Laser-induced incandescence: Particulate diagnostics for combustion, atmospheric, and industrial applications

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Abstract

The understanding of soot formation in combustion processes and the optimization of practical combustion systems require *in situ* measurement techniques that can provide important characteristics, such as particle concentrations and sizes, under a variety of conditions. Of equal importance are techniques suitable for characterizing soot particles produced from incomplete combustion and emitted into the environment. Additionally, the production of engineered nanoparticles, such as carbon blacks, may benefit from techniques that allow for online monitoring of these processes.

In this paper, we review the fundamentals and applications of laser-induced incandescence (LII) for particulate diagnostics in a variety of fields. The review takes into account two variants of LII, one that is based on pulsed-laser excitation and has been mainly used in combustion diagnostics and emissions measurements, and an alternate approach that relies on continuous-wave lasers and has become increasingly popular for measuring black carbon in environmental applications. We also review the state of the art in the determination of physical parameters central to the processes that contribute to the non-equilibrium nanoscale heat and mass balances of laser-heated particles; these parameters are important for LII-signal analysis and simulation. Awareness of the significance of particle aggregation and coatings has increased recently, and the effects of these characteristics on the LII technique are discussed.

Because of the range of experimental constraints in the variety of applications for which laser-induced incandescence is suited, many implementation approaches have been

developed. This review discusses considerations for selection of laser and detection characteristics to address application-specific needs. The benefits of using LII for measurements of a range of nanoparticles in the fields mentioned above are demonstrated with some typical examples, covering simple flames, internal-combustion engines, exhaust emissions, the ambient atmosphere, and nanoparticle production. We also remark on less well-known studies employing LII for particles suspended in liquids.

An important aspect of the paper is to critically assess the improvement in the understanding of the fundamental physical mechanisms at the nanoscale and the determination of underlying parameters; we also identify further research needs in these contexts. Building on this enhanced capability in describing the underlying complex processes, LII has become a workhorse of particulate measurement in a variety of fields, and its utility continues to be expanding. When coupled with complementary methods, such as light scattering, probe-sampling, molecular-beam techniques, and other nanoparticle instrumentation, new directions for research and applications with LII continue to materialize.

Keywords: Laser-induced incandescence; Particulate diagnostics; Nanoparticles; Nanoscale heat transfer; Soot; Black carbon

1. Introduction

The past several decades have been marked by a growing concern over the effects of combustion-generated particles on human health and the environment. Numerous scientific studies have indicated that soot particles, commonly produced during the incomplete combustion of hydrocarbons, pose a serious public health risk, e.g., [1]. The effects of atmospheric soot emissions have gained a great deal of recent attention, stemming from growing concerns over the contribution of black-carbon particles to climate change. Most atmospheric particles reflect incoming solar radiation and tend to partially counteract global warming; soot particles, in contrast, strongly absorb in the visible and near infrared and are associated with significant positive radiative forcing leading to global warming. Models predict that the radiative forcing from atmospheric soot is ~65% that of CO₂, the most abundant greenhouse gas [2]. Relative to CO₂, however, soot has a short lifetime in the Earth's atmosphere, and reducing soot emissions into the atmosphere has been proposed as a near-term climate-change mitigation approach [3]. Black carbon can also settle on snow and ice, and thus decrease their reflectivity. In addition to increasing the radiative forcing because of this albedo effect, the additional surface absorption can accelerate melting of snow and ice, which, in turn, further enhances surface absorptivity and global warming [2, 4].

Hydrocarbon combustion is the main source of atmospheric soot emissions, and, although soot chemistry has been studied extensively over decades, some of the primary mechanisms in soot formation, growth, and oxidation are uncertain [5-8]. Controlling soot emissions

will require focused efforts to understand soot formation and oxidation under a variety of combustion conditions. Combustion is also the main source for industrially manufactured carbon nanostructures and provides various opportunities for the synthesis of non-carbon nanoparticulate materials [9]. Experimental studies of particulate formation in the context of both reduced emissions and targeted synthesis rely heavily on the ability to measure and characterize particles over a wide range of conditions.

Whereas incipient soot is close to spherical, small (1–10 nm), and composed of large organic species with high hydrogen-to-carbon ratios, mature soot is composed of primary particles of 10–50 nm in diameter with fine structures similar to polycrystalline graphite (low hydrogen-to-carbon ratios) [10-14]. These primary particles are covalently bound into branched-chain aggregates of tens to hundreds of nanometers in size. These aggregates are non-spherical and are characterized by fractal dimensions typically in the range of 1.7–1.9 [11, 14-18]. The non-sphericity of these particles complicates optical measurements of them. In addition, these particles can be coated with semi-volatile coatings in the combustor, exhaust stream, or atmosphere, which can significantly change their physical characteristics and optical properties. Mature soot, whether coated or not, absorbs strongly in the ultraviolet (UV), visible, and infrared (IR) wavelength ranges, is a refractory material with a sublimation point of about 4000 K, and is insoluble in polar and non-polar solvents. Atmospheric scientists refer to particles with these characteristics as "black carbon" [2, 19, 20]. When we speak of carbonaceous particles or soot without further specification in this text, we mean the type of solid particles combustion scientists refer to as "mature soot" and the atmospheric community has defined as "black carbon". To reduce redundancy and

confusion, we avoid the commonly used expression "refractory black carbon" or "rBC" because all black carbon particles are refractory.

Laser-induced incandescence (LII) has become a workhorse for *in situ* measurements of mature-soot particles in combustion and ambient atmospheric environments. This technique involves heating soot particles with a high-power, pulsed or continuous-wave (CW) laser to temperatures high enough to emit measurable quasi-blackbody radiation. This emission is recorded either with gated (time-integrated) detection techniques, often for the spatially resolved visualization of particle-distribution or volume-fraction measurements, or with time-resolved detection methods, particularly for primary-particle size or single-particle measurements. One advantage of LII is that it is particularly sensitive to, and highly selective for, soot particles because they absorb strongly and can be heated to temperatures of ~4000 K before they sublime. LII with pulsed and CW lasers (called pulsed LII and CW LII in this paper, respectively) are based on the same physical processes, but the experiments and the data analyses are different. They will be treated in parallel throughout the paper.

Alternative methods for measuring the concentrations of particulate matter and particle sizes include *in situ* optical and probe-sampling online and *ex situ* approaches. Direct comparison of the existing techniques is complicated because they use different physical effects for detection and thus respond in different ways to the chemical composition (e.g., volatile vs. non-volatile components) and morphology (measuring primary-particle sizes, aggregate sizes, or mass). Sampling techniques are additionally influenced by artifacts that may occur during sampling (condensation, formation of additional particles,

agglomeration) and the modification of the structure of particles when deposited on surfaces and exposed to environmental conditions. Nevertheless, all of these techniques have inherent strengths and potential weaknesses and characteristics that can be exploited for selected applications or combined with LII in order to extend ranges of applicability.

Similarly to LII, photoacoustic techniques take advantage of the large absorption cross section in the ultraviolet, visible, and infrared for selective measurements of soot particles [21, 22]; these techniques, however, do not heat the particles to the same degree, are influenced by other absorbing species, and thus do not have the same selectivity. Elastic light scattering (ELS) is even less selective than photoacoustic approaches and will give signal from other particles, such as fuel or water droplets. ELS can, however, yield information about aggregate sizes and morphologies [15, 23-30]. Coupling these optical techniques can be a powerful approach for *in situ* and *ex situ* particle characterization.

Measuring light extinction, which results from a combination of absorption and scattering, provides absolute soot-volume fractions if the optical properties are known [31-34]. It is the most direct and commonly employed method to measure soot concentrations in flames and is thus often used to calibrate LII volume-fraction measurements [35-39]. Applications of extinction often employ a standard approach of scanning a laser beam through a flame or other sample or using full-field extinction [40] with an expanded light bundle. In axisymmetric flames extinction is usually analyzed with an Abel inversion procedure to obtain local concentrations. In non-premixed flames application of this inversion may be challenging [41] as regions of low soot concentration may be surrounded by annular zones of high soot load. Considerable improvement in full-field extinction techniques has been

achieved by the introduction of diffused light illumination, mitigating effects of beam steering [42, 43]. For atmospheric applications, particle concentrations are generally too low to measure using single-pass extinction, and multi-pass approaches, such as cavity ring-down spectroscopy (CRDS), are used [44-46]. CRDS can be sensitive enough to measure single particles [47]. As an alternative to extinction, a number of emission techniques have been introduced to investigate soot in flames [48-50]. These techniques also suffer from the inherent challenges of retrieving spatially-resolved information from integrated signals. For these types of studies, LII has the advantage of applicability in an imaging mode for the determination of spatial distributions of particles.

For measurements of concentrations of carbon particles in engine exhaust and the atmosphere, various techniques exist that are extensively treated in several review articles [20, 51-53]. Methods employed in these disparate research areas have converged in recent years because drastically reduced particulate-matter emissions from engines yield relevant concentrations in engine exhaust that are similar to those in the atmosphere. In the field of engine-exhaust analysis, the traditional procedure is Constant Volume Sampling (CVS), combined with the deposition of particulate matter on a filter [54]. Besides simple gravimetric analysis, various alternative approaches allow for a determination of the composition of the matter collected. Filter-based approaches are also very common for atmospheric measurements [20, 46]. Another popular approach in this area relies on the aethalometer principle, where the opacity of a filter paper after loading is measured optically. Other instruments in this field include photoacoustic spectroscopy, mentioned

above, and various forms of microbalances. For further information the reader is referred to recent reviews [20, 51, 53, 54].

For particle-size measurements, LII provides information about primary-particle sizes.

These measurements are sometimes complemented by *ex situ* measurements of aggregate-size distributions by differential-mobility sizing using a scanning mobility particle sizer (SMPS). Here, particles are charged and size-selected according to their mobility in the carrier gas within an electric field; particles within the various size fractions are subsequently counted with a condensation particle counter (CPC). This technique yields the electric mobility diameter, which is the diameter of a sphere that would balance the electric force with the drag force on a charged particle in an electric field moving with a flow field. For a soot aggregate, which is not spherical, the mobility diameter can be related to the radius of gyration [55], but the relationship depends on the particle morphology [14] and the properties of the carrier gas. Aggregate sizes can similarly be measured using an aerodynamic particle sizer, the measurement concept of which is based on measurements of drag forces from particle velocities in an accelerating carrier gas, e.g., [56, 57]. These measurements are usually made for single particles in the inlet of an aerosol mass spectrometer (AMS) following an aerodynamic lens system, e.g., Refs. [58, 59].

Alternatively, the aerodynamic aerosol classifier (AAC) measures the aerodynamic size by balancing aerodynamic drag forces with centrifugal forces generated between rotating drums at atmospheric pressure [60, 61]. Particle-mass distributions can similarly be measured using a centrifugal particle-mass analyzer (CPMA) or aerosol particle mass analyzer (APM), which classifies charged particles subjected to a centrifugal force

generated between rotating drums by their mass-to-charge ratio [62, 63]. An attractive feature of the AAC is that it does not require charging the particles and thus is not confounded by multiple charging effects common to the SMPS, CPMA, and APM.

All these measurements require extractive sampling and delivery to the measurement device. Dilution techniques are often employed in an attempt to avoid interaction of particles in the sampling lines and condensation of volatile species. In practice, avoiding condensation and nucleation in the extraction probe is difficult. Sampling techniques that gather particle samples from flames and hot gases, e.g., by thermophoretic sampling on a surface that is exposed to the gases for a short period, have been developed for atmospheric and high-pressure systems [64, 65], but measures need to be taken to avoid or account for particle modification by condensation on the cool sampling surface. For sampled particles, imaging techniques, such as transmission electron microscopy (TEM) and scanning electron microscopy (SEM), have been invaluable for characterizing aggregate morphologies and sizes and primary-particle sizes, e.g., [13, 66-72]. X-ray spectroscopic techniques, such as near-edge x-ray absorption fine-structure spectroscopy (NEXAFS), e.g., [73-75], and x-ray photoelectron spectroscopy (XPS), e.g., [68, 76, 77], can also provide considerable information about soot-particle fine structure and chemical composition, e.g., [68, 73]. X-ray scattering techniques, such as small- and wide-angle x-ray scattering (SAXS and WAXS, respectively), have also been used for *in situ* measurements of soot-particle size and morphology [75, 78-86].

Online measurements of particles from flames and atmospheric samples have been carried out with molecular-beam and particle-beam techniques, in which an aerosol sample is

locally extracted from a flame or the atmosphere, and further reaction is interrupted by rapid expansion of the sampled aerosol into vacuum or by rapid dilution with an inert gas. A particle-laden beam is formed and sent into an AMS or a particle-mass spectrometer (PMS) [87-89], which is a variant of the classical mass spectrometer for gaseous species and very small particles [90]. A fraction of the particles in the sample are charged either by collisions with ions or electrons, by thermionic emission, or by photoionization. In the size range of interest for studying primary particles, multiple charging is unlikely according to Fuchs' theory [91]. The particle beam is directed through a velocity filter and an adjustable electric field. The particles are deflected according to their kinetic energy and charge towards a Faraday cup, where their number is counted by measuring charges. This arrangement has been used in combination with LII measurements in low-pressure and atmospheric-pressure flames [92-95]. Alternatively the particles are vaporized thermally or photolytically, and the particle constituents are analyzed in a mass spectrometer, providing detailed information about particle composition, e.g., [92, 96-104].

Under some conditions, LII has been combined with complementary, non-optical techniques for particle characterization and for intercomparison of alternative methods, e.g., [69, 70, 95, 105]. The combination of methods may further be used to obtain even more information about the morphology of complex particles. Whereas elastic light scattering (ELS) or its combination with LII yields radii of gyration of soot aggregates, alternative methods may provide an aerodynamic or an electrical mobility diameter, all demonstrating different dependencies on the structure of complex particles.

Additional methods may also be useful for the calibration of LII measurements. Besides extinction, such methods include the absolute intensity approach, requiring knowledge of the particle temperature from multi-wavelength pyrometry in order to determine mass and volume concentration [105-107], and comparison to gravimetric filter methods for mass or volume concentration in *ex situ* applications [108]. Single-particle CW instruments are generally calibrated using a mobility size-selected particle distribution of a carbonaceous material that gives similar behavior to atmospheric particles, such as agglomerated-fullerene particles [109-111].

This review updates and expands on previous reviews [20, 112-115]. We focus here on fundamental aspects of the technique and its implementation, highlighting new developments over the past several years. In particular, we have included sections on the effects of aggregation and particle coatings on LII signals, LII from non-carbonaceous materials, and the application of the technique to atmospheric particulates.

2. Principle and development of LII

Laser-induced incandescence is generated by heating particles with a high-energy or high-power laser. As particles interact with the laser and absorb light, their temperatures increase, leading to a strong non-linear increase in thermal radiation, i.e., incandescence, with temperature according to Planck's law. Figure 1 shows a typical experimental setup used to measure *in situ* time-resolved particle temperatures and pulsed-LII signals. There are several key components to an LII setup for either pulsed or CW LII. The details of implementation will vary between CW or pulsed approaches, but the fundamental roles of

the components are the same. Figure 1 demonstrates these components for pulsed LII. They include

1. The laser system: Particles are very commonly heated with the fundamental beam of a pulsed Nd:YAG laser at 1064 nm. The second harmonic (i.e., 532 nm) can be used to heat the particles, but visible (and shorter) wavelengths are often avoided for combustion conditions because of potential interferences from laser-induced fluorescence (LIF) from polycyclic aromatic hydrocarbons (PAHs) (see Section 4.1.1). Sometimes an injection-seeded laser is used for applications that require a smooth laser temporal profile (see Section 4.1.4).
2. Optics for laser-beam intensity control: The laser beam intensity can be attenuated to control the fluence using a combination of a half-wave plate, which rotates the laser polarization, and a thin-film polarizer, which transmits only one linear component of the polarized beam and rejects the other. The setup shown in Fig. 1 has two thin-film polarizers for better rejection of the off-axis polarization and better fluence control. Sometimes a thin-film polarizer is placed before the half-wave plate to pass the dominant polarization of the laser output and reject portions of the beam that may have scrambled polarization caused by strain birefringence in the oscillator rod.
3. Optics for managing the spatial profile of the laser beam: The laser beam can be used directly, or it can be shaped for a specific application. For imaging of particle distributions, the beam can be expanded in one direction and reduced in the other using cylindrical lenses to produce a laser-sheet profile (see, for example, Fig. 26). For applications that require a top-hat (i.e., homogeneous) spatial profile, an optical layout like the one shown in Fig. 1 is

often used. The near-field output of the laser is relay imaged to an aperture using a two-lens telescope. The first lens is one focal length away from the laser output coupler. The distance between the first lens and the second is the sum of their focal lengths. In Fig. 1, the lenses have the same focal length, and the telescope neither expands nor reduces the beam (i.e., one-to-one imaging is employed in the first telescope). The aperture is separated from the second lens by the lens's focal length. A true image of the beam at the output coupler is formed at the position of the aperture, where the beam is collimated. The aperture passes only the center of the beam, which is relatively flat if the full output of the laser is Gaussian or quasi-Gaussian. A second telescope relays the image of the aperture to the detection volume. In the example shown in Fig. 1, the second lens has a focal length that is one half that of the first lens, and the beam is therefore reduced by a factor of two. Any number of two-lens image-relaying telescopes can be used in series to relay the near-field beam quality to the detection volume. The spatial filtering aperture can be placed at the front end of any of these telescopes to be imaged to the detection volume. An example of the beam spatial profile at the detection volume is shown in the inset. In the setup in Fig. 1, the laser power and beam spatial profile are monitored, and the laser beam is collected in a beam dump after passing through the sample.

4. The sample: In Fig. 1, the beam is passed through a flame, where it interacts with soot.

5. Detection optics: In Fig. 1, the signal is split and is passed through imaging optics similar to those in the laser beam. Figure 1 shows two-lens telescopes that include achromatic lenses separated from each other by the sum of the focal lengths. The first lens is a focal length away from the flame, and the second lens is a focal length away from the detector.

Dichroic filters are generally placed in the collimated region between the lenses. Other imaging configurations are also common. In Fig. 1, there is a notch filter to block laser scatter and filters selected for two-color LII (see Section 4.2.2).

6. The detector(s): Figure 1 shows two photomultiplier tubes (PMTs) connected to an oscilloscope for time-resolved detection. There are many detector configurations tailored to the application. Alternatively, there could be CCD cameras for imaging the soot distribution (see Section 4.2).

Examples of the time dependence of the particle temperature and resulting LII signal are shown in Fig. 2 for pulsed-laser heating of flame-generated soot. At low laser fluences (Fig. 2a), particle temperatures increase throughout most of the laser pulse until the absorptive heating rate is balanced by rates of other cooling mechanisms. The cooling is dominated by conduction to the surrounding atmosphere at atmospheric and higher pressures. After the laser pulse, the signal decays relatively slowly at atmospheric pressure and flame temperatures. In the low-fluence regime, when laser fluences are increased, signal decay rates following the laser pulse increase because the difference between the peak particle temperature and the ambient temperature increases. At higher pressures and lower gas temperatures, conductive cooling rates and LII signal decay rates are much faster. At high fluences (Fig. 2b), particles sublime during the laser pulse, and LII-signal-decay rates are predominantly controlled by the rate of mass loss. At these fluences, the rate of signal decay from mass loss depends on laser fluence; once the particle reaches the sublimation point, the signal-decay rate increases substantially with increasing laser fluence, which shifts the peak signal to earlier times with increasing laser fluence. An example of this

behavior, which is a distinguishing feature of pulsed-LII signals in the high-fluence regime, is shown in Fig. 3 from Ni *et al.* [116]; Curves 2, 3, and 4 are in this regime.

At high laser fluences, particle temperatures reach a maximum close to the sublimation temperature, as shown in Fig. 4. Under flame conditions for pulsed LII, spectrally inferred temperatures are often higher than the equilibrium sublimation temperature of ~ 4140 K at which C_3 sublimates, and particles sometimes appear to reach temperatures of ~ 4460 K at which C_2 sublimates [117-121]. The reason for this behavior is not well understood and requires further work. The peak-LII signals saturate when the sublimation temperature is reached (see Fig. 4). This behavior has been observed in pulsed LII in several studies [117, 120-123]. The peak temperature inferred from spectrally resolved incandescence, however, depends on the wavelength dependence of the emissivity used to fit the spectra [117, 118, 123, 124]. The current uncertainties in this wavelength dependence convey an uncertainty of ~ 200 K to the soot peak temperature in the high-fluence regime for mature soot [118, 123] and an even higher uncertainty for less mature soot [123]. These fluence curves were recorded with a top-hat laser-beam profile and a fast detector. Influences of the experimental conditions on these fluence curves are discussed in Section 4.

The underlying processes are the same for CW-laser heating of the particles. In the most common implementation of this technique, individual particles are injected into the cavity of a CW-laser beam, and the broadband-LII signal is recorded as the particles traverse the beam. The intra-cavity-beam intensity is very high (in the range of 1.7×10^5 – 1×10^6 W/cm² [125, 126]), and the particles traverse the beam for ~ 10 μ s before they exit the beam or are fully vaporized [125, 127-130]. A typical CW-LII temporal profile is shown in Fig. 5. The

laser-particle interaction is determined by the drift time of the particle through the laser beam [131] and is comparable to particles interacting with a high-fluence laser pulse ($\sim 3 \text{ J/cm}^2$) with a pulse duration of $\sim 10 \text{ }\mu\text{s}$ [130]. Peak CW-LII signals demonstrate similar behavior to that shown in Fig. 4a when the laser power is varied [126]. Ideally, under normal operating conditions, the particles are heated to the sublimation temperature and are fully sublimed, and changing the laser power simply changes the amount of time it takes to sublime the particle. Current measurements with this technique commonly use the approach developed by Stephens *et al.* [132] and implemented for atmospheric measurements by Schwarz *et al.* [125-127] and Moteki and Kondo [129, 131]. This approach is employed in the Single-Particle Soot Photometer (SP2, Droplet Measurement Technologies), which is shown in Fig. 6.

During either pulsed- or CW-LII-signal collection, the particle can also heat by oxidation or cool by radiative emission. Other processes can influence the signal, including changes to the optical properties of the particle during the heating and cooling process, e.g., by annealing of the particle fine structure or changes to the aggregate morphology or composition. These processes are reviewed in more detail in the next section.

The first published demonstration of the application of LII to measure soot involved the use of a TEA CO_2 laser to heat carbon-black particles [133]. The signals were temporally resolved and analyzed using an energy-balance model. Weeks and Duley [133] proposed that the temporally-resolved LII signal could be used to infer particle size. A few years later Eckbreth [134] and Burakov *et al.* [135] developed energy- and mass-balance models to describe laser-induced heating, incandescence, and saturation of LII signal from soot by

vaporization of the particle and used temporal LII profiles measured in flames to validate their models. Bukaty *et al.* [136, 137] and Melton [138] later expanded the model analysis to include other heating and cooling mechanisms. This early work provided a basis for much of the development performed over the next decade for applications of LII to study soot in engine cylinders [139-144], engine exhaust [106, 108, 145, 146], laminar and turbulent flames [24, 36, 67, 116, 147-162], and other environments [163-165]. These demonstrations of the applicability of LII to particle measurements under adverse conditions were followed by an explosion of work applying LII to a wide variety of combustion and atmospheric conditions.

Will *et al.* [120, 166, 167] and Roth *et al.* [156, 168-171] made significant advances in the modeling of time-resolved signals for primary-particle sizing, an approach now usually referred to as TiRe-LII. Following the earlier modeling work of Weeks and Duley [133], Eckbreth [134], Burakov *et al.* [135], Bukaty *et al.* [136, 137], and Melton [138], Bengtsson *et al.* [172, 173], Smallwood *et al.* [121, 146, 174, 175], and Michelsen [176, 177] developed detailed models to serve as a means to extend the technique to a wider variety of conditions, including high pressures and low temperatures, by understanding and targeting the largest sources of uncertainties. This work was complemented and followed by a considerable body of work to develop models for predicting LII signals under a wide range of conditions [35, 129, 132, 145, 147, 154, 178-190]. Vander Wal *et al.* [13, 37, 191] performed extensive surveys of experimental parameters for optimizing the application of the technique, used LII coupled with laser-induced fluorescence (LIF) measurements to distinguish soot precursors from mature soot particles in flames [158, 160, 192], and

applied the technique to carbon nanotubes and other materials [193, 194]. Snelling and coworkers [106, 107, 195] developed the technique of auto-compensating LII, which enabled quantitative measurements of volume fraction without the need for calibration via another soot measurement technique, such as extinction or gravimetric analysis. De Iuliis *et al.* [122] also recognized the benefits of this approach, and it has subsequently been adopted by others. Witze *et al.* coupled LII with elastic scattering measurements to measure particle-size changes from laser-induced sublimation [26] and to infer the fraction of volatile coatings on exhaust particles [196, 197]. Atmospheric black-carbon measurements usually employ an approach developed by Stephens *et al.* [132] in which individual particles are injected into the cavity of a CW laser, and the LII signal is recorded simultaneously with elastic scatter from the particle. Stephens *et al.* [132], Schwarz *et al.* [125, 126], and Moteki and Kondo [129] demonstrated that the magnitude of the peak of the LII signal provides information about the mass of an individual particle, and the relative timing of the LII and scatter signals provides some information about volatile coatings. Alternatively, atmospheric measurements can be performed with a high-sensitivity variant of pulsed LII [198, 199].

A large body of work has been published on the applications of LII to combustion, industrial, and atmospheric environments. In this review we give examples of some of this work to provide the reader with an overview of what has been done and where the field currently stands. With careful experimental design and appropriate calibration, LII measurements of volume and mass concentration can be made without a detailed heat-transfer model or understanding of all the fundamental processes. However, this

fundamental understanding is essential for an accurate determination of primary-particle sizes, reduction of measurement uncertainties, identification of artifacts, and resolution of issues and limitations associated with LII measurements. We conclude with a perspective on current underlying issues and limitations, and suggest possibilities for areas of development.

3. Fundamental understanding

3.1. Underlying processes

LII measurements entail heating particles to temperatures at which incandescence is readily observable. These temperatures are generally in the range of 2500-4000 K. Under these conditions a number of processes can be initiated and have significant impact on the LII signal. A great deal of work has focused on developing an understanding of these processes and models capable of predicting LII signals under a variety of conditions; we will refer to these papers in the respective sections of this manuscript. In this section we discuss the processes that are involved, or are likely to be involved, in determining the temporal evolution of the LII signal and highlight uncertainties associated with our incomplete understanding of these processes. These individual processes contribute to the energy and mass balance of the particle. Figure 7 provides a summary illustration of these processes. Many researchers have implemented the underlying equations into in-house software to predict and analyze LII signals. With LIISim, one system has been made freely available [200]; this system allows the user to calculate LII-signal decays for variable conditions and provides data-analysis capabilities for uploaded experimental data. The Community LII

Modeling Environment (CLiiME) [201] will also be publicly available soon. The CLiiME code uses object-oriented modern Fortran and is modular, open-source, and user modifiable, and the default model has been validated over a wide range of conditions and can be applied to either pulsed- or CW-LII configurations.

3.1.1. Energy and mass balance

The time evolution of the particle temperature and diameter are calculated by solving the time-dependent energy and mass balance equations including the contributions from a number of mechanisms. The energy-balance equation can be summarized as

$$\frac{dU_{\text{int}}}{dt} = \dot{Q}_{\text{abs}} + \dot{Q}_{\text{rad}} + \dot{Q}_{\text{cond}} + \dot{Q}_{\text{sub}} + \dot{Q}_{\text{ox}} + \dot{Q}_{\text{ann}} + \dot{Q}_{\text{therm}} , \quad (1)$$

where U_{int} is the internal energy of the particle, t represents time, $\dot{Q}_{\text{abs}}$ is the absorptive-heating rate for a single primary particle, $\dot{Q}_{\text{rad}}$ is the radiative-cooling rate, $\dot{Q}_{\text{cond}}$ is the conductive-cooling rate, $\dot{Q}_{\text{sub}}$ is the evaporative-cooling rate, $\dot{Q}_{\text{ox}}$ is the oxidative-heating rate, $\dot{Q}_{\text{ann}}$ is the heating rate from annealing, and $\dot{Q}_{\text{therm}}$ is the thermionic-cooling rate. In most models the rate of change of the internal energy of the particle is expressed as

$$\frac{dU_{\text{int}}}{dt} = \rho_s c_s \frac{\pi d_p^3}{6} \frac{dT}{dt} = M c_s \frac{dT}{dt} , \quad (2)$$

where ρ_s is the particle density, c_s is the particle specific heat, d_p is the primary-particle diameter, T is the particle temperature, and M is the particle mass. Liu and Snelling [175], Hiers [184], and Michelsen *et al.* [202] derived a modified expression with an extra term that accounts for the loss of some ability to store sensible heat when the mass of the particle

is reduced, but this term is canceled in Eq. (1) by extra terms introduced in similar expansions of terms for sublimation [175, 184, 202] and oxidation [184, 202]. The interested reader is referred to the respective papers; in the following discussion, the straightforward version of the balance is used. Substituting Eq. (2) into Eq. (1) and solving for temperature yields

$$\frac{dT}{dt} = \frac{1}{M c_s(T)} (\dot{Q}_{\text{abs}} + \dot{Q}_{\text{rad}} + \dot{Q}_{\text{cond}} + \dot{Q}_{\text{sub}} + \dot{Q}_{\text{ox}} + \dot{Q}_{\text{ann}} + \dot{Q}_{\text{therm}}). \quad (3)$$

The mass loss is also solved for explicitly, i.e.,

$$\frac{dM}{dt} = \left(\frac{dM}{dt} \right)_{\text{sub}} + \left(\frac{dM}{dt} \right)_{\text{ox}}, \quad (4)$$

and is related to the diameter via the density according to

$$d_p = \left(\frac{6M}{\pi \rho_s} \right)^{\frac{1}{3}}. \quad (5)$$

Solving these coupled differential equations yields the mass, diameter, and temperature, which are needed to calculate signal.

The LII signal is produced by radiative emission from a heated particle. The signal per primary particle S is calculated using the Planck function modified by the emissivity ϵ_λ to account for deviations from a perfect blackbody, a function to account for the wavelength-dependent efficiency of the detection system Σ_λ , and a factor to account for the solid angle detected, i.e.,

$$S = \Omega \pi d_p^2 \int_{\lambda} \varepsilon_{\lambda} \frac{2\pi h c^2}{\lambda^5 \left[\exp\left(\frac{hc}{\lambda k_B T}\right) - 1 \right]} \Sigma_{\lambda} d\lambda, \quad (6)$$

where λ is the emission wavelength, h is the Planck constant, c is the speed of light, and k_B is the Boltzmann constant. The emissivity is given in the Rayleigh approximation, where $d_p \ll \lambda$, by

$$\varepsilon_{\lambda} = \frac{4\pi d_p E(m)}{\lambda}, \quad (7)$$

where m is the complex index of refraction and $E(m)$ is the refractive-index function for absorption discussed in Sec. 3.2.2. The absorption function $E(m)$ depends on wavelength, and, based on Kirchhoff's law, also describes the emission properties of a particle. The signal is usually calculated over some narrow or broad wavelength range in the ultraviolet, visible, or near-infrared spectral regions.

3.1.2. Laser absorption and radiative emission

The most important process for LII is the initial step involving the heating of the particle by absorption of the incident laser light. In the absence of any nonlinear absorption mechanism, this process is straightforward and easily calculated, as long as the absorption cross section is known. The absorption cross section is the geometric cross section, i.e. the cross-sectional area, πd_p^2 in the case of a spherical particle, multiplied by the so-called absorption efficiency that represents the ratio between the true and the geometric cross section. For a primary particle in the Rayleigh regime ($d_p \ll \lambda_L$ where λ_L is the laser wavelength), the absorption cross section is given by

$$\sigma_{\text{abs}} = \frac{\pi^2 d_p^3 E(m)}{\lambda_L}. \quad (8)$$

One important implication, e.g., relevant for the measurement of soot-volume fraction by LII, is that the absorption cross section for small particles scales with particle volume. This cross section will change if the particle size changes (e.g., if the particle starts to sublime and shrink or undergoes swelling with increasing temperature and decreasing density) or if the index of refraction and $E(m)$ change (e.g., if the particle anneals to a more ordered form of carbon). The absorptive-heating rate (in units of J/s) is then calculated according to

$$\dot{Q}_{\text{abs}} = \sigma_{\text{abs}} E_e(t), \quad (9)$$

where $E_e(t)$ is time-dependent irradiance of the incident laser.

The radiative-cooling rate is calculated using the Planck function integrated over all wavelengths, i.e., Eq. (6) with $\Sigma_\lambda = 1$. If there is no wavelength dependence to $E(m)$, the radiative-cooling rate is given by [176]

$$\dot{Q}_{\text{rad}} = -8\Gamma(5)\zeta(5) \frac{\pi^3 d_p^3 (k_B T)^5 E(m)}{h(hc)^3}, \quad (10a)$$

where the value of the gamma function of 5, $\Gamma(5)$, is 24, and the value of the Riemann zeta function of 5, $\zeta(5)$, is 1.0363, and

$$\dot{Q}_{\text{rad}} = -\frac{198.97\pi^3 d_p^3 (k_B T)^5 E(m)}{h(hc)^3}. \quad (10b)$$

A wavelength dependence for $E(m)$ can also be taken into account in a similar, more complicated expression [176]. The radiative-cooling rate is normally significantly smaller than other cooling rates, such as the conductive-cooling rate at atmospheric pressure and above and the evaporative-cooling rate at high laser fluence. Under vacuum at low laser fluences, however, this mechanism is the dominant cooling mechanism. Recent experimental work has been performed under vacuum conditions to study this mechanism in more detail [174, 183, 203], but more work needs to be done to improve the experimental results and model comparisons.

3.1.3. Conduction

For CW LII and at low and moderate laser fluence for pulsed LII, conductive cooling is the most important processes for heat loss by the particle, particularly at atmospheric pressure and above (for the relative importance of the various cooling processes as a function of particle temperature, see Section 3.1.8). Understanding heat transfer by conduction is critically important for inferring primary-particle sizes from pulsed-LII decay rates. Conductive cooling also competes with absorptive heating on the timescales relevant for CW-LII detection and can limit the minimum particle size detectable using this approach [126, 130]. Considerable work has been devoted to identifying an appropriate conductive-heat-transfer mechanism for LII applications. In the free-molecular-flow regime (where the mean free path is much greater than the particle diameter), the conductive-cooling rate can be expressed as [176, 204, 205]

$$\dot{Q}_{\text{cond}} = \frac{-\pi d_p^2 \alpha_T p_0}{RT_0} \sqrt{\frac{RT_0}{2\pi W_a}} \left(C_p - \frac{R}{2} \right) (T - T_0), \quad (11)$$

where p_0 is the ambient pressure, T_0 is the ambient temperature, R is the universal gas constant, W_a is the molecular weight of air, C_p is the heat capacity of air at constant pressure, and α_T is the thermal-accommodation coefficient. A more detailed version of this expression, which accounts for the temperature dependence of the heat capacity, is given by [202]

$$\dot{Q}_{\text{cond}} = \frac{-\pi d_p^2 \alpha_T p_0}{RT_0} \sqrt{\frac{RT_0}{2\pi W_a}} \left[\int_{T_0}^T C_p(T') dT' + \frac{R}{2}(T - T_0) \right], \quad (12)$$

where the additional $R(T - T_0)$ accounts for the work associated with the expansion of the heated gas molecules leaving the surface. In many LII models the heat capacity is assumed to be constant; this assumption has little impact on the calculated value of the conductive-cooling rate [202]. Neglecting the work of expansion, however, may decrease the conductive-cooling rate by 22–29% [202].

Whereas free-molecular-regime conduction models are valid for flames at atmospheric pressure and below, transition- and continuum-regime models are more accurate in simulation of conduction for room temperature or high-pressure applications. A review of these models by Liu *et al.* [206] recommends the Fuchs boundary-sphere model [91] for modeling particle conductive cooling in LII applications because it has greater accuracy throughout the transition regime and beyond, i.e., to large particle-to-gas temperature ratios. Many models use an alternative mechanism introduced by McCoy and Cha [207] and used in the model developed by Melton [138]. Transition-regime approaches, such as the Fuchs boundary-sphere model [91] and the McCoy-Cha [207] mechanism, are preferred for high-pressure applications. Work by Kuhlmann *et al.* [167] suggests that, at atmospheric

pressure and an ambient gas temperature of 300 K, the Fuchs boundary-sphere model [91] predicts conductive-cooling rates as much as 20% higher than those calculated using the McCoy-Cha approach [207] for typical primary-particle sizes of ~ 30 nm, as shown in Fig. 8. Bambha and Michelsen [130] demonstrated that the McCoy-Cha approach [207] can be used to reproduce CW-LII temporal profiles at room temperature and atmospheric pressure, as long as shielding to individual primary particles is explicitly taken into account.

The McCoy-Cha expression [207] assumes a transition regime between the free-molecular regime and the continuum regime and is represented as

$$\dot{Q}_{\text{cond}} = -\frac{2\kappa_a \pi d_p^2}{d_p + GL} (T - T_0), \quad (13)$$

where κ_a is the thermal conductivity of the bath gas, L is the mean free path, and G is a function of the heat-capacity ratio $\gamma = C_p/C_v$ and is given by

$$G = \frac{2(9\gamma - 5)}{\alpha_T(\gamma + 1)}. \quad (14)$$

Equation (13) can be shown to be equivalent to Eq. (11) when $GL \gg d_p$ [204]. The expression given in Eq. (14) is appropriate for monatomic bath gases. Modifications to this model, which account for conductive cooling by polyatomic gases, have also been employed to reproduce room temperature, atmospheric pressure, CW-LII temporal profiles [130].

The value used for α_T has a significant effect on the calculated conductive-cooling rate.

Section 3.2.1 gives an overview of the uncertainties associated with this parameter.

Snelling *et al.* [121] have shown that models need to account for changes in the local gas temperature resulting from heat transfer to the surrounding bath gas under some conditions. This effect has been further investigated experimentally by Nordström *et al.* [208]. With respect to conductive cooling, much of the current work focuses on understanding the impact of aggregate size [69, 130, 167, 206, 209-213], primary-particle polydispersity [206, 212, 214-216], bridging between primary particles [213], aggregate morphology [70, 130], and particle maturity [217, 218] on pulsed-LII-signal decay rates and CW-LII-signal timing; the work on the influence of aggregate size and structure is reviewed in Section 3.3. A number of studies have also targeted high-pressure applications for which this influence is extremely important [219-224].

3.1.4. Sublimation and other vaporization mechanisms

At high laser fluences evaporative-heat loss is the most significant cooling mechanism. More importantly, however, sublimation and other vaporization mechanisms lead to rapid decreases in LII signal because of mass loss during the laser pulse. These mechanisms are the most poorly understood of the mechanisms that have a strong influence on LII signals. Most LII models are not able to reproduce the rapid signal decays attributable to evaporative-mass loss or the plateau in the peak temperature at high fluences (shown in Fig. 4).

The majority of LII models represent the evaporative-cooling rate as [138, 146, 204]

$$\dot{Q}_{\text{sub}} = \frac{\Delta H_v}{W_v} \left(\frac{dM}{dt} \right)_{\text{sub}}, \quad (15)$$

where H_v is the enthalpy of formation of the sublimed carbon clusters, and W_v is the average molecular weight of the sublimed carbon clusters. The mass-loss rate attributable to sublimation is given by

$$\left(\frac{dM}{dt}\right)_{\text{sub}} = \frac{-\pi d_p^2 W_v \alpha_M p_v}{RT} \left(\frac{RT}{2\pi W_v}\right)^K, \quad (16)$$

where α_M is the mass-accommodation coefficient, p_v is the average saturation partial pressure of the sublimed carbon clusters, and K is a constant that is typically equal to 0.5 but is sometimes given a value of 0.4 to account for non-idealities of the bath gas and desorbed clusters [180]. Some models use temperature-dependent parameterizations for the average mass, enthalpy of formation, and partial pressure for the sublimed carbon clusters [146, 202], whereas others use the specific mass, enthalpy of formation, and partial pressure derived from the Clausius-Clapeyron equation for each carbon cluster sublimed [171, 176]. A summary of sublimation sub-models is given in [204].

In order to reproduce the fast signal decay rates for pulsed LII at high fluence, however, a much faster mechanism must be included in the model [176, 186, 188, 204]. Such a mechanism is also required to keep the particles from superheating to temperatures well above the sublimation temperature (see Fig. 4). One approach is to use a photodesorption mechanism that limits absorption of photons to states that thermalize, and allows photons to excite states that lead to non-thermal mass loss. One such mechanism is described in [176], and a similar mechanism is described in [204].

Experimental studies have demonstrated that soot aggregates do not disaggregate (i.e., break into individual primary particles) when laser heated, even at high fluences [67, 70, 73, 152, 191, 192]. Measured changes in soot scattering and absorption cross sections have indicated that vaporization leads to particle-size reductions during laser irradiation [26, 119, 178, 225, 226]. These results are consistent with conclusions drawn from transmission electron microscopy (TEM) images of laser-irradiated soot particles, which have demonstrated aggregates with smaller primary particles but the same morphology as non-irradiated particles [67, 73, 152, 191, 192]. Further support is provided by scanning mobility particle sizer (SMPS) measurements of laser-heated soot particles, which showed formation of new particles with amorphous carbon fine structure (as determined by TEM), suggesting condensation of vaporized carbon clusters [73]. Despite these studies, considerably more work is needed to understand and experimentally confirm these mechanisms.

3.1.5. Oxidation

Oxidation can lead to both particle heating and mass loss, but this process is much slower and less significant than either laser heating or evaporative-mass loss at high laser fluences. Its rate is on the time scale of conductive cooling, however, and it tends to slow signal decay rates for pulsed LII at low fluences [186, 202]. Neglecting oxidation could thus have an impact on primary-particle sizing at low fluences. Nevertheless, most LII models do not include this mechanism [204]. The oxidative-heating term can be expressed as [176, 202, 204]

$$\dot{Q}_{\text{ox}} = \frac{(\Delta H_{\text{ox}} + \alpha_T C_p^{\text{CO}} T)}{W_1} \left(\frac{dM}{dt} \right)_{\text{ox}}, \quad (17)$$

where H_{ox} is the enthalpy of the reaction $\text{C} + \frac{1}{2}\text{O}_2 \rightarrow \text{CO}$ (at a reference temperature T_{ref}), which is equivalent to the enthalpy of formation of CO, α_T is the thermal-accommodation coefficient, C_p^{CO} is the heat capacity of CO, and W_1 is the molecular weight for a carbon atom. In some references [176, 204], an expression similar to Eq. (17) was used, but H_{ox} was defined relative to the reaction $2\text{C} + \text{O}_2 \rightarrow 2\text{CO}$. The expression given by Eq. (17), however, does not account for the temperature of the incoming O_2 [184, 202]. Michelsen *et al.* [202] provided a more detailed expression that explicitly accounts for the incoming O_2 , i.e.,

$$\dot{Q}_{\text{ox}} = \frac{1}{W_1} \left\{ \Delta H_{\text{ox}} + \int_{T_{\text{ref}}}^{T_f^{\text{CO}}} \left[C_p^{\text{CO}}(T') + \frac{R}{2} \right] dT' - \frac{1}{2} \int_{T_{\text{ref}}}^{T_0} \left[C_p^{\text{O}_2}(T') + \frac{R}{2} \right] dT' \right\} \left(\frac{dM}{dt} \right)_{\text{ox}}. \quad (18)$$

The rate of mass loss through oxidation can be expressed according to

$$\left(\frac{dM}{dt} \right)_{\text{ox}} = \frac{-\pi d_p^2 W_1 2k_{\text{ox}}}{N_A}, \quad (19)$$

where N_A is the Avogadro constant, and k_{ox} is the rate constant for $2\text{C} + \text{O}_2 \rightarrow 2\text{CO}$. Of course, under combustion conditions, concentrations of radical species OH and O can be significant, and rates of oxidation by these species may contribute substantially to overall oxidation rates [227-229]. In the atmosphere, NO_2 can also oxidize soot [230]. At LII temperatures, however, oxidation by O_2 is rapid and nearly diffusion controlled, and using reactions with O_2 as a proxy for oxidation by all oxygenated species is not a bad

approximation. Other chemical reactions at the surface, such as growth mechanisms, happen on timescales longer than those important for LII detection.

3.1.6. Annealing

Numerous experimental studies have demonstrated that the fine structure of mature soot is very similar to polycrystalline graphite with graphite crystallites preferentially aligned parallel to the particle surface near the surface and more randomly ordered near the center of the particle [10, 12, 73, 231-233]. Several studies have shown that these particles can anneal when irradiated with a laser, such that graphite crystallites originally ~2–5 nm in length and 3-10 graphite layers deep form more extended layered structures with fewer defects [68, 73, 191, 192, 234]. At some fluences extension of these crystallites includes introduction and migration of five- and seven-membered ring structures, which allow curvature of these graphitic layers, such that the primary particles can form carbon-onion structures with hollow cores [68, 73, 191, 192, 234].

Annealing is expected to be exothermic and will thus heat the particle. A more important contribution from this process is its potential influence on all of the other processes because it changes the physical structure of the particle on the fine scale. Little is known about this mechanism, its rate, its energetics, and how it affects other physical parameters, such as the particle's optical properties. Although this mechanism could have a very significant impact on LII signals, it is disregarded in most LII models because so little is known about how to take it into account. The heating rate for this mechanism can generally be expressed as

$$\dot{Q}_{\text{ann}} = \frac{-\Delta H_{\text{ann}} k_{\text{ann}} N_d}{N_A}, \quad (20)$$

where H_{ann} is the enthalpy for annealing, k_{ann} is the rate constant for annealing, and N_d is the number of defect sites in the particle. The one model that includes annealing assumes that the particle anneals at low temperatures by migration of interstitial atoms that combine with defects within the graphite planes [176]. The particle anneals at high temperatures by in-plane vacancy migration. The number of defects is solved for explicitly. A more detailed description of this mechanism can be found in [176].

3.1.7. Thermionic emission

Thermionic emission has a relatively small influence on the particle-cooling rate and is generally disregarded in LII models [204]. One reason to include this mechanism, in addition to accounting for its impact on the energy balance of the particle, is to understand and predict the charging of the particle caused by laser heating. The expression for this term is based on a Richardson-Dushman approximation given by [182, 204, 235]

$$\dot{Q}_{\text{therm}} = -\frac{4\phi m_e (\pi d_p k_B T)^2}{h^3} \exp\left(\frac{-\phi}{k_B T}\right), \quad (21)$$

where ϕ is the work function, and m_e is the electron mass.

3.1.8. Overview of the relative importance of the energy-loss processes

Table 1 provides an overview of the effects of the mechanisms involved in the energy and mass balance of the particle. The mechanisms that have the most impact on the energy balance are laser absorption for heating, conduction and sublimation for cooling, and

sublimation for mass loss. These mechanisms are highlighted in bold type in Table 1. The main effects on the LII signal for pulsed and CW LII are listed separately.

For pulsed LII, a critical parameter for defining the influence of each mechanism is the laser fluence, and the most relevant fluence ranges for each mechanism are listed in Table 1. The fluence ranges listed in Table 1 are approximate, as there is a transition from low fluence to high fluence around a peak soot temperature of about 4000 K, above which sublimation becomes dominant. The relevant fluence ranges depend on a number of factors, such as laser wavelength, particle maturity, and ambient temperature and pressure. Figure 9 demonstrates the importance of these mechanisms at atmospheric pressure, as predicted by an energy- and mass-balance model [186] that includes all of the mechanisms discussed in Section 3.1. For these calculations, the model assumed a primary-particle diameter of 33 nm and used a measured laser temporal profile with a pulse duration of 8.3 ns (FWHM), a wavelength of 532 nm, and fluences of approx. 0.03 and 1.00 J/cm². On this timescale, the shape of the laser profile is approximately represented by the absorption rate at low fluences, i.e., the blue line in Fig. 9a. Figure 10 demonstrates the increasing importance of conductive cooling with increasing pressure at temperatures below ~4000 K and the importance of evaporative cooling at higher temperatures.

For CW LII, the integrated absorption over the laser-particle interaction time is relatively high, such that the particle, or most of it, is expected to reach the sublimation point. For this application approach, the impact of each mechanism will depend on the state of the particle as a function of the laser-particle interaction time. Figure 11 shows an example of the relative importance of these heating and cooling mechanisms during the laser-particle

interaction time, as predicted by an LII model that includes all of the mechanisms described in Section 3.1 [130]. Calculations were performed for a mature soot particle with a fractal dimension of 1.9, mobility diameter of 126 nm, mass of 0.415 fg, and monodispersed primary-particle size of 13 nm. The model used a measured laser-particle interaction time with a laser wavelength of 1064 nm. At atmospheric pressure and room temperature, conductive-heat loss is the dominant cooling mechanism. The calculations are shown for an aggregate of primary particles with varying exposed surface area. The result presents the average for this distribution of primary particles [130].

3.2. Physical properties and key parameters

In this section we evaluate some of the key parameters used in LII modeling. Because of both their central role in LII models and the on-going debate on the actual values to be employed, we will focus on the thermal accommodation coefficient α_T and the optical absorption function $E(m)$ of the complex index of refraction m , but will also remark on density ρ and specific heat c_s of the particle. As in most instances in this review, the discussion will be essentially restricted to refractory (i.e., solid) carbonaceous particles, mostly mature soot. A general overview of the different parameters used by various groups in the application of LII can be found in Michelsen *et al.* [204], where the status in 2007 is reflected. We are especially interested in the origin of the values in use in order to critically evaluate their reliability and to ease the way to assessing the accuracy of the values obtained on the basis of past and future experiments and modeling activities.

3.2.1. Thermal accommodation coefficient

The thermal accommodation coefficient α_T is a key parameter for particle sizing by LII. For conditions under which the dominant cooling mechanism is conduction to the surrounding atmosphere, α_T is central to linking the temperature-decay rate to primary-particle size in pulsed LII and for distinguishing between morphology effects and coating effects on signal timing in CW LII [130]. A similar connection also exists for the particle density ρ_s and specific heat c_s , but α_T has generally been assumed to contribute more significantly to uncertainties in LII models than other parameters. The accommodation coefficient describes the extent to which gas molecules exchange vibrational, rotational, and translational energy with a surface during surface-scattering events. Given a Maxwell-Boltzmann distribution of initial states, the initial energy of bath-gas molecules striking the surface can be characterized by an initial temperature T_0 . Likewise, the Maxwell-Boltzmann distribution of states, following interactions with the particle surface, can be characterized by a final temperature T_f . For a surface temperature of T , the accommodation coefficient is defined as [236]

$$\alpha_T = \frac{T_f - T_0}{T - T_0}. \quad (22)$$

When gas-surface collisions are completely elastic, i.e., no energy is exchanged, this parameter has a value of 0. When gas molecules are fully equilibrated with the surface during these interactions, this parameter reaches a value of 1.

No direct measurements have been made of the thermal-accommodation coefficient with soot, but a few direct measurements have been made on carbon surfaces, which serve as a good surrogate for mature soot. The accommodation coefficient has been measured to be 0.26 for room-temperature N_2 interacting with a graphite surface at 1220–1270 K [237] and 0.20–0.25 for CH_4 interacting with amorphous carbon surfaces at 850 and 1400 K [238]. The latter value is suspicious because CH_4 thermally decomposes at temperatures above 1240 K [239, 240]. This process is catalyzed by carbon surfaces such that decomposition occurs at temperatures closer to 900 K in the presence of carbon black [239, 240].

Furthermore, the relevant temperatures for LII measurements often range from either room temperature (~ 300 K) or flame temperatures (1700–2000 K) to laser-heated-particle temperatures of 2500–4000 K. The accommodation coefficient is expected to depend on surface temperature and gas temperature [236, 241], and extrapolating lower temperature measurements of α_T to higher temperatures is difficult without additional information.

Michelsen [242] used state-to-state measurements of NO scattered from graphite surfaces as a function of surface temperature and incidence energy to derive partial accommodation coefficients for vibrational, rotational, and translational degrees of freedom. These partial accommodation coefficients were combined into a surface-temperature and gas-temperature-dependent global accommodation coefficient, as shown in Fig. 12.

Measurements were made at surface temperatures as high as 700 K and gas temperatures as high as 1650 K. The results were extrapolated to higher temperatures to yield an estimate of 0.18–0.21 for α_T at LII-relevant surface temperatures and typical flame-gas temperatures.

Although the trends with surface temperature and gas temperature are consistent with

previous measurements for H₂, Kr, Xe, and CH₄ on graphite (see [242] and references therein), the data available for such a study are limited, and no data are available to confirm the high-surface-temperature extrapolation.

Daun and coworkers have performed molecular-dynamics simulations [243, 244] to derive α_T . The results indicate that the dependence of the accommodation coefficient on surface temperature is insignificant at surface temperatures above 2400 K, as shown in Fig. 13 [244]. These results do not extend to surface temperatures that overlap with available data, however, and have not been confirmed with experiments at these higher temperatures. Nevertheless, the molecular dynamic simulations for rotational and translational accommodation are qualitatively consistent with the results shown in Fig. 12 from experimental data extrapolated to higher temperatures. The results from the molecular-dynamics simulations for monatomic gases suggest that α_T increases with increasing gas temperature [243], which is consistent with the trends inferred from surface scattering measurements [242]. The results for N₂, however, indicate that α_T decreases with increasing gas temperature [244], which is inconsistent with surface scattering measurements [242]. However, the paucity of measurements as a function of gas temperature makes these trends difficult to confirm.

At ambient pressures greater than or equal to atmospheric pressure and temperatures well below the sublimation temperature of ~ 4000 K, the pulsed-LII-signal decay rate is believed to be largely determined by the conductive-cooling rate. Given a model that can accurately calculate the conductive-cooling rate and pulsed-LII-signal decay rate for a known primary-particle size, one should be able to infer the thermal-accommodation coefficient from a

measured pulsed-LII-temporal profile, assuming that the accommodation coefficient is independent of particle-surface temperature, the LII model is accurate, and there are no other complicating factors. Unfortunately most models have difficulties reproducing pulsed-LII-signal decay rates during the first 50 ns or so [121, 245, 246], the thermal-accommodation coefficient is likely to be temperature dependent [121, 236, 241, 242], and there are many other ill-defined factors that influence this decay rate, such as aggregate size [69, 167, 206, 209-212] and morphology [70, 130] (see Sec 3.3), particle composition [217, 218, 246, 247], oxidation and annealing [176], and other coupled parameters with large uncertainties. These factors make it challenging to derive a physically meaningful thermal-accommodation coefficient from pulsed-LII-temporal profiles.

A number of studies have produced estimated values for α_T from fits of current models to LII temporal profiles measured under specific conditions with known (or assumed) particle sizes [121, 167, 169, 180, 181, 245, 246, 248-250]. Kuhlmann *et al.* [167, 250] have pointed out that LII-inferred values depend strongly on the heat-transfer model used to derive them. Somewhat detached from its original physical meaning, the accommodation coefficient may often be regarded as a central calibration constant in LII particle sizing. Values of α_T used for a vast range of experimental conditions include 0.05–0.5 for a number of monatomic and polyatomic bath gases [245], 0.1–0.5 for various bath gases [248], 0.20–0.25 for an aggregate [167, 250], 0.22–0.34, depending on particle composition [246], 0.28 [180] (where the original value of 0.07 given there results from a deviating definition of α_T) and 0.37 for soot in a laminar diffusion flame [249], 0.38 [181], 0.36–0.44, varying inversely with fluence and peak surface temperature [121], 0.38–0.44 for an

isolated primary particle [167, 250], and 1.0 [169]. Values below 0.1 appear to be associated with measurements on polyatomic gases that tend to thermally decompose at temperatures above ~ 1000 K [245].

As mentioned above, these values also depend on a number of other factors. For example, the dependence of α_T on soot maturity is not well established, but recent results from Bladh *et al.* [217, 247] and López-Yglesias *et al.* [218] suggest that α_T decreases with increasing soot maturity. These results are supported by theoretical and experimental studies of surface roughening and enhanced gas-surface energy transfer at graphite surfaces when hydrogen is bound to the surface [238, 251-256]. Michelsen and coworkers [218] derived an expression for α_T that depends on the surface maturity of the particle. Using this expression, they demonstrated that the previously unexplained rapid pulsed-LII signal decay over the first ~ 50 ns after the laser pulse [121, 245, 246] could be explained by enhanced conductive cooling for surfaces with relatively high hydrogen content, which decreases, as does α_T , as the particle surface is oxidized and annealed [218]. The data used to derive this expression are limited, however, and did not include an easily quantifiable assessment of soot maturity, such as H/C ratio or other metric for long-range order of the fine structure of the surface; more work is necessary to verify and quantify the dependence of α_T on soot maturity.

Calculated signal-decay rates for pulsed LII and signal timing for CW LII are also dependent on associated uncertain parameters, such as particle density and specific heat. Because the choice of both α_T and heat-transfer model strongly influences particle sizes

inferred from measured pulsed-LII temporal profiles, care must be taken when comparing particle sizes derived from experiments using different LII models in the analysis.

Although the uncertainties associated with α_T are considerable, values of this parameter in common use in current LII models span the relatively narrow range of 0.23–0.37 [204] for mature soot in a flame with a well-ordered surface fine structure. Numbers approaching the value of 0.9 used in the original work of Melton [138] have been inferred for soot at room temperature when the particle-surface fine structure is disordered [218]. Melton’s value originates from a corresponding value from McCoy and Cha [207], who in turn state that, with the choice of this value, their theory “matches the data of Harbour as reported by Hidy and Brock”.

These large uncertainties in the value of α_T have a significant impact on the accuracy of pulsed-LII-particle sizing and the reliability of analysis of CW-LII-signal timing and shape related to particle morphology and composition. The impact of these uncertainties highlights the need for further work to focus on the determination of a physically-derived α_T that can be incorporated into appropriate heat-transfer models. A better understanding and description of the thermal-accommodation coefficients relevant for LII and their dependence on particle characteristics and measurement conditions may finally evolve if carefully conducted experiments are accompanied by, and are used to validate, a combination of molecular dynamics and Monte-Carlo simulations.

3.2.2. Absorption function

The refractive-index function for absorption $E(m)$ is critically important for calculating laser absorption, and hence particle-heating rates, i.e., Eq. (8); $E(m)$ is just as important for calculating radiative emission, i.e., Eq. (10), and hence LII signals, i.e., Eqs. (6) and (7).

This parameter is given by

$$E(m) = -\text{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\} \quad (23)$$

where m is the complex index of refraction, i.e., $m = n - ik$.

This parameter is of central importance when calibrating LII signals for soot-volume-fraction measurements through comparison with extinction measurements. This calibration is only valid if the value of $E(m)$ is the same for the calibration (extinction) and LII-measurement conditions. This parameter, however, may be sensitive to a number of factors, such as wavelength [119, 249, 257-260], particle composition [70, 218, 246, 247, 259-261], and particle temperature [119, 226], and controlling for some of these factors during calibration may be difficult.

A wavelength dependence of m imparts a wavelength dependence to $E(m)$. For single-color time-resolved pulsed-LII applications, uncertainties in the magnitude of $E(m)$ directly affect the predicted peak temperature and calculated particle-cooling rate, which determines the particle size inferred. The wavelength dependence of $E(m)$ is much more critical than its absolute value in two-color pulsed-LII primary-particle sizing, where temperatures are inferred from fits of the Planck function to measured signal in two wavelength regions.

Under these conditions, uncertainties in the wavelength dependence of $E(m)$ lead to uncertainties in inferred particle temperatures [107, 118], cooling rates, and inferred primary-particle sizes.

Because knowledge of the optical properties of soot is also important for light extinction and scattering measurements of soot in flames, combustion exhaust streams, and the atmosphere, there is a wide body of literature covering these properties. We refer the interested reader to several excellent reviews in this area [15, 262, 263]. Here we will focus on particular aspects of these optical properties directly relevant to LII measurements.

For the determination of $E(m)$ the knowledge of m is sufficient, but not necessary, and there are several ways to measure $E(m)$. One way of measuring $E(m)$ is via extinction measurements. The problem inherent in this approach is that scattering must be taken into account, as the extinction cross section σ_{ext} of a particle ensemble is the sum of the absorption and scattering cross sections, σ_{abs} and σ_{sca} , respectively. Although the single-scattering albedo $\omega = \sigma_{\text{sca}}/\sigma_{\text{ext}} = \sigma_{\text{sca}}/(\sigma_{\text{sca}} + \sigma_{\text{abs}})$ for carbonaceous particles is small under many conditions, it is non-negligible. Reported values are in the range of 0.19–0.47 in the visible [119, 263–267] and 0.006–0.077 at 1064 nm [119, 130], depending on particle size and maturity. Therefore, for a reliable determination of σ_{abs} and $E(m)$ from extinction, scattering properties have to be considered, which in turn requires an appropriate description of soot aggregates, typically based on a fractal approach (see Sec. 3.3), and knowledge of the refractive-index function for scattering

$$F(m) = \left| \frac{m^2 - 1}{m^2 + 2} \right|^2. \quad (24)$$

In the end, the determination of the two refractive-index functions $E(m)$ and $F(m)$ is equivalent to determining both the real and imaginary parts of m .

To a first approximation, the particle temperature change upon laser heating, $T_{\max} - T_0$ (with peak temperature $T_{\max}$ and initial temperature T_0), is directly proportional to $E(m)$, as long as sublimation and other effects do not play a role. Because the LII signal depends strongly on particle temperature, the LII signal itself can be used with an LII model to infer $E(m)$. A number of recent experiments have addressed the determination of $E(m)$ or its wavelength dependence based directly on LII measurements. One relatively early example for such an approach is the work of Snelling *et al.* [249]. From a pyrometric measurement of the soot peak temperature in a diffusion flame, they found $E(m) = 0.42$ for 1064 nm under the assumption of a wavelength-independent absorption function and $E(m) = 0.395$ when using a linear dependence of $E(m)$ on λ . Later similar experiments [121] yielded a value of 0.4 for 532 nm excitation in a diffusion flame, and de Iuliis *et al.* [268] obtained a value of 0.29 at 1064 nm in a diffusion flame. Beyer and Greenhalgh [183] performed low-fluence LII measurements under high-vacuum conditions (10^2 Pa) for particles extracted from a diffusion flame. Because conductive cooling is greatly reduced under vacuum, where radiative cooling dominates, their experiments had decay times of several tens of microseconds. From the radiative emission, they were able to obtain an estimate for an average $E(m)$ value of 0.4 weighted over the wavelength range of emission.

Measuring LII at two laser wavelengths offers a convenient method for testing the wavelength dependence of $E(m)$ for these two wavelengths, as first proposed by Therssen *et al.* [269]. The idea is to induce an identical particle temperature increase (a proxy for total energy absorbed) at two laser wavelengths and to calculate the $E(m)$ ratio from the ratio of fluences required to achieve this target temperature change. The identical maximum temperature is determined by matching the peak LII signal, the signal-decay rate, or the inferred-particle temperature for the two wavelengths [119, 218, 260, 261, 269, 270]. Therssen *et al.* [269], Michelsen *et al.* [119, 218, 271], and Bejaoui *et al.* [270] measured values in the range of about 0.84 – 1.0 for the ratio $E(m, 532 \text{ nm})/E(m, 1064 \text{ nm})$ for mature soot in diffusion and premixed flames.

Because of structural and compositional changes due to variations in the maturity of the soot, $E(m)$ is expected to change with height above the burner (HAB), especially in the lower regions of premixed flames and the central regions of diffusion flames. The ratio $E(m, 532 \text{ nm})/E(m, 1064 \text{ nm})$ appears to decrease with increasing soot maturity, as shown by the results of Clon *et al.* [261] (see Fig. 14) and Lpez-Yglesias *et al.* [218]. In order to study the effects of compositional changes of $E(m)$ with HAB in a premixed flame, Clon *et al.* [261] used a low-pressure ($\sim 0.03 \text{ MPa}$) premixed $\text{CH}_4/\text{O}_2/\text{N}_2$ flame to extend the soot inception zone. As shown in Fig. 14, they observed a strong decrease in the $E(m)$ ratio for 532 and 1064 nm with increasing HAB until a constant value was reached. The absolute values, however, for the ratio $E(m, 532 \text{ nm})/E(m, 1064 \text{ nm})$ found in this low-pressure, premixed flame are exceptionally high, exceeding unity, in contrast to investigations in atmospheric, diffusion and premixed flames. Lpez-Yglesias *et al.* [218] observed a similar

trend, i.e., a lower $E(m)$ ratio for mature soot in the edges of co-flow diffusion flames relative to the less mature soot in the centers of the flames. This approach requires care to reduce the influences of interferences from LIF [218, 261]. In addition, use of UV wavelengths can yield misleading results because such wavelengths can lead to photodissociation rather than direct heating [272, 273]. Nevertheless, these results are consistent with many studies using other techniques that have demonstrated a shift in the absorption cross section to longer wavelength with increasing soot maturity, i.e., decreasing H/C ratio and increasing percentage of sp^2 hybridization [43, 259, 274-280].

In addition to a change in the wavelength dependence of $E(m)$, the magnitude of $E(m)$ also changes with soot maturity. The absorption cross section or $E(m)$ increases with increasing soot maturity [218, 246, 247, 261, 275-277, 281-283], and a higher laser fluence is required to bring less mature soot to similar LII-signal levels (for pulsed LII) and temporal behavior (for CW LII) as more mature soot [218, 246, 261, 283]. For pulsed LII this dependence results in a shift in fluence curves (i.e., peak LII vs laser fluence) to higher fluence with decreasing particle maturity, i.e., increasing hydrogen content [218, 246, 283]. Figure 15a shows results from Bladh *et al.* [284] who measured peak pulsed-LII signal at different locations in a premixed ethylene flame. The peak LII is shifted to higher fluence at lower heights above the burner (HAB), where the soot is less mature and particles sizes are smaller, as indicated by faster LII-signal decay rates (Fig. 15b). This behavior may be entirely attributable to a dependence of the absorption cross section on soot maturity [283] or to a combination of factors, including changes in density and specific heat with changes in soot maturity [218]. For a leaner flame, the peak LII does not reach a plateau and

increases nearly linearly, as shown in Fig. 15c. These results are accompanied by very fast signal-decay rates that do not change with HAB, as shown in Fig. 15d. The primary-particle size inferred from these data was ~ 1 nm [284]. Although unusual, nearly linear fluence curves have been reported previously by Mouton *et al.* [285] under similar flame conditions. Bladh *et al.* [284] suggest that this behavior may be related to physicochemical transformations of the particles during laser heating or less efficient laser heating of these small particles. Identifying the reasons of this behavior will require further investigation.

In another study Bladh *et al.* [247] investigated the variation of $E(m)$ with HAB in a premixed atmospheric flame for an incident wavelength of 1064 nm by two-color LII and found an increase from about 0.21 for low positions in the flame (with a corresponding primary-particle size of about 5 nm) up to 0.45 at large HAB. Eremin *et al.* [286] studied the variation of $E(m)$ with particle size during the formation of carbon (and iron) particles in a shock-tube experiment and found very low values ($E(m) \sim 0.05$) for the smallest particles with sizes of a few nanometers increasing up to 0.25 (again at 1064 nm) for the largest particle (about 20 nm). These results appear to have large uncertainties, however, because multiple parameters were varied simultaneously to derive both $E(m)$ and particle size. In addition, other parameters, such as particle density, could easily be changing with particle size, and these parameters could be very strongly coupled to a value for $E(m)$ derived in this manner, making $E(m)$ more of an empirical fit parameter for these conditions. Values of $E(m)$ depend on soot maturity, and the magnitude of $E(m)$ and its wavelength dependence can vary dramatically, both with location within a flame, and between diffusion and premixed flames (premixed flames tend to produce less mature soot

than diffusion flames). Care should therefore generally be taken to note the conditions under which $E(m)$ is determined.

With the high temperatures involved in LII measurements, other effects relevant for absorption must also be considered. Particles may expand or change in fine structure if annealing occurs. It is far from trivial to isolate the effects on $E(m)$ from those of other parameters, such as size, density, and heat capacity. Michelsen *et al.* [119] coupled LII and extinction measurements at laser wavelengths of 532 and 1064 nm to derive the fluence dependence of the scattering and absorption cross sections at these wavelengths. Their experimental results indicate that the absorption cross sections increase with increasing fluence until the point at which the particle starts to sublime (see Fig. 16), and the scattering cross sections decrease with increasing fluence. The magnitude of the increase in absorption cross section is consistent with a swelling of the particle because of changes in density with temperature, but this trend could also be attributed to a change in the optical properties of the particle. Michelsen *et al.* [119] attributed the scattering trend either to a change in the optical properties of the particle or to a change in the morphology of the particle with temperature. Thomson and coworkers [226, 287] confirmed these trends in extinction and scattering cross sections. They attempted to invoke Maxwell Garnett theory to derive a temperature-dependent $E(m)$, which suggested that particle expansion could not explain the absorption-cross-section enhancement with increasing temperature. Maxwell Garnett theory, however, addresses the problem of calculating an average index of refraction of a system comprised of inclusions, such as soot particles in a medium, e.g., air [15, 288-290], and cannot be used on a microscopic scale to predict the temperature

dependence of m or $E(m)$ [291]. More work is thus required to provide a full understanding of the changes in both the absorption and scattering cross sections with particle temperature.

A large body of work has demonstrated that coatings on soot particles can have a significant influence on their optical properties. A non-absorptive coating on a soot particle is expected to increase both the scattering cross section [197, 292-294] and the absorption cross section [262, 263, 292, 294-297]. These predicted enhancements in scattering and absorption cross sections have been confirmed experimentally [97, 197, 298-305].

Aggregate morphology may also influence soot optical properties. As the fractal dimension (i.e., particle compactness) increases, the absorption cross section is predicted to decrease for fractal dimensions below 2 [15, 294, 306] and increase for fractal dimensions above 2 [307]. Increases in scattering cross sections with increasing fractal dimension are not well understood; scattering cross sections are predicted to either increase [307, 308] or decrease [15, 197, 294, 309] with increasing fractal dimension. Optical effects of coatings or morphology could have a considerable influence on the time evolution of CW-LII signals because particle composition and morphologies can vary widely for atmospheric application, but more work needs to be done to confirm such effects.

3.3. Influence of aggregation

In the initial years of development, LII models, including Melton's [138] original description of the basic LII process, only considered isolated spherical particles or those in point contact and neglected effects of aggregation on the various mechanisms involved in LII. The issue of aggregation was originally addressed by Vander Wal *et al.* [13], who compared primary-particle sizes inferred from pulsed-LII-signal-decay rates and TEM

images at various heights in diffusion flames and found partial agreement, depending on the position in the flame, an effect they attributed to the degree of aggregation. Filippov and Rosner [310] went much further and argued that under certain conditions, namely, if particles are in the near-continuum regime, aggregate-size distributions might be inferred from the decay rates of pulsed-LII temporal profiles. Although no evidence for such an application is known, this statement highlights the fact that researchers started to think about the influence of particle structure on LII.

Soot aggregates and many other types of nanoparticles synthesized in flames may be characterized by a fractal-like approach [18, 311-313], where the number N_p of primary particles per aggregate can be described by

$$N_p = k_f \left(\frac{2R_g}{d_p} \right)^{D_f} . \quad (25)$$

Here, D_f is the fractal dimension, which for soot aggregates is typically in a range of ~ 1.7 – 1.9 [11, 14-18], and k_f is the fractal prefactor, a proportionality constant of order unity [15]. Literature values span a wide range of values for the fractal prefactor [17, 314]. In some cases this spread is due to a different formulation from that expressed in Eq. (25).

Experimental results often yield a value for k_f of 1.9 – 2.8 [18, 315, 316], which tends to be larger than that obtained from simulations. The enhanced experimental value may be attributable to particle overlap found in real aggregates [17, 314, 317].

Filippov *et al.* [209] investigated the influence of aggregate size and morphology on heat-transfer rates for free-molecular and continuum regimes. The effects of aggregate size on the decay rate of pulsed-LII signals were later incorporated into LII models, and the

consequent impact on estimations of primary-particle size parameters was investigated [211, 249]. Liu *et al.* [206] illustrated that distributions of both aggregate size and primary-particle size influence the decay rate of pulsed-LII signals (see Fig. 17). This study showed that polydispersity in the aggregate size has little impact in the free molecular regime but becomes important in the transition regime, and polydispersity in primary-particle diameter in general is always important and should be considered. Liu *et al.* [206] further introduced the concept of the equivalent heat-transfer sphere to model the effect of shielding on heat conduction between aggregate particles and the surrounding gas.

Apart from the overall aggregate structure, the overlap or bridging effect needs to be considered in particle sizing by pulsed LII or assessing LII-signal timing in CW LII as it reduces the surface area available for conductive-heat transfer. The particle overlap may be characterized by an overlap parameter $C_{ov} = (d_p - d_{ij}) / d_p$ [17, 317], where d_{ij} may be regarded as the distance between the centers of two particles. For point contact $C_{ov} = 0$, complete sintering/coalescence would result in $C_{ov} = 1$. The value of C_{ov} depends on formation characteristics and, when measured in a flame, on height above the burner, but values in the range of $C_{ov} \approx 0.15\text{--}0.25$ may be regarded as common for many soot samples [318-320]. Because the decay rates of pulsed-LII signals depend on the specific-surface area, such a reduction in free surface directly influences signal-decay rates, which, in turn, influence inferred primary-particles sizes. If an overlap parameter is known, this reduction may be calculated in a straightforward manner when employing a realistic thermal-accommodation coefficient. If, however, an effective-accommodation coefficient is used from previous calibration measurements of LII vs. TEM data, this value implicitly takes

into account this surface reduction, as long as the calibration is performed on particles with the same morphology and ambient conditions. As a kind of second-order effect, bridging between primary particles also influences shielding phenomena in heat transfer, as recently analyzed by Johnsson *et al.* [213]. Under some conditions, e.g., under fuel-rich premixed combustion conditions and in exhaust streams [321-323], soot aggregates can have a more compact morphology, which significantly reduces the effective surface area and can substantially reduce the pulsed-LII signal-decay rates [70], as demonstrated below.

Generally, the aggregate structure may influence most of the individual processes introduced in Section 3.1; in practice, however, these effects only appear to be important for heat conduction and, to a lesser extent, for the absorption of laser radiation. One would certainly expect an effect of aggregation on the sublimation process; the expanding vapor cloud from one particle will interact with that from a neighboring particle. Why this topic has not been discussed so far may be explained by the typical conduct of particle-sizing experiments in the low-fluence regime, on the one hand, and by the general difficulty in adequately describing sublimation with LII on the other hand. As an additional effect, analysis of scattering signal recorded simultaneously with CW LII suggests that the radius of gyration of the particle increases significantly during sublimation; the effect has been hypothesized to be similar to the expansion of a kernel of popcorn when the vaporization of water in the kernel causes the kernel to pop [130].

Light absorption is usually described by the Rayleigh-Debye-Gans approximation for fractal aggregates (RDG-FA) [15, 315], which postulates that primary particles in an aggregate absorb light independently of the aggregate, i.e., as if they were isolated particles.

The absorption cross section of an individual primary particle in turn is simply computed using the Rayleigh approximation, given by Eq. (8). Several studies, e.g., [21, 306, 324-328], compared more exact numerical models with the RDG-FA approximation and found that RDG-FA under-predicts laser absorption for conditions typical for LII by roughly 10% for a small aggregate of ~ 100 primary particles with a fractal dimension less than 2. The resulting effect is equivalent to that from a corresponding error in the absorption function $E(m)$, see Section 3.2.2. By Kirchoff's law of thermal radiation, emission of radiation should also be affected by aggregation. Because radiation provides only a minor contribution to the energy balance under standard conditions, effects to the energy balance of the particle may be safely neglected. Aggregation may influence the LII signal, however, and has been predicted to impart a temperature-dependent bias in the incandescence [326]. In two-color LII, the emission properties are differently affected for the two wavelengths, but this effect is second-order compared to the wavelength dependence of the emission function. Liu and Smallwood [210] state that the effect may be neglected for laminar diffusion flames at atmospheric pressures, but may become important when considering aggregates with broader size distributions and larger primary particles, such as found in high-pressure flames and atmospheric particles.

In practice, however, the most important effect of aggregation to LII applications is the reduced conductive heat transfer. For quantification, a shielding factor $\eta = \dot{Q}_{\text{agg}} / (N_p \dot{Q}_{\text{cond}})$ is introduced, relating the conductive-cooling rate $\dot{Q}_{\text{agg}}$ for an aggregate to that from N_p independent primary particles. A number of studies, e.g., [206, 209, 213, 249, 329], have carried out molecular dynamics simulations to quantify this effect. Although different

in the detailed approach and in their quantitative results, all of these investigations predict a strong decrease in heat conduction for the initial addition of primary particles and a leveling off when the aggregate consists of tens or hundreds of monomers (see Fig. 18).

Experimental investigations of the relationship between aggregate size and heat transfer are scarce. In a study with 14 different carbon blacks, Kuhlmann *et al.* [167, 250] could demonstrate a clear correlation between the number of primary particles per aggregate and an effective-accommodation coefficient derived from the comparison of LII and TEM data. Based on Fuchs's heat-transfer model [91] and the shielding approach by Snelling *et al.* [249], they extrapolated the data to derive a thermal-accommodation coefficient for an isolated particle of $\alpha_T = 0.38 \pm 0.03$. Bladh *et al.* [69] used aggregates from a soot generator based on a propane diffusion flame with similar primary particles but various degrees of aggregation and could qualitatively demonstrate reduced heat conduction with increasing aggregation.

Because of this shielding effect, the fractal dimension also influences the conductive-cooling rate. More compact aggregates have a smaller effective surface area. Bambha *et al.* [70, 130] have shown that the conductive-cooling rates and pulsed-LII-signal decay rate can be substantially reduced for aggregates with larger fractal dimensions. They extracted soot from a co-flow diffusion flame, size-selected it for a mean aggregate size of 150 nm, cooled it, coated it with oleic acid, and then removed the coating in a thermodenuder, which caused the aggregates to become more compact, increasing the fractal dimension from an average of 1.9 for the uncoated particles (Fig. 19, inset) to 2.3–2.4 for the coated-denuded particles (Fig. 19, inset). The curves in Fig. 19 show a comparison of pulsed-LII temporal

profiles for uncoated and processed (compact) particles and demonstrates the change in pulsed-LII-signal-decay rate caused by the aggregate morphology change. The compact particles have a much lower conductive-cooling and signal-decay rate.

Such shielding effects can also influence CW-LII signals. Figure 20 shows average CW-LII temporal profiles from an SP2 using particle distributions generated in the same way as those used for the study shown in Fig. 19 [130]. Because the SP2 measures single particles, it is possible to identify the particle size associated with a specific temporal profile, which enables studies of aggregate-size effects. Conductive cooling competes very effectively with absorptive heating on timescales relevant for CW LII in the SP2 configuration.

Particle shielding reduces conductive-cooling rates and leads to more rapid heating of particles, which shifts LII temporal profiles to earlier times. Figure 20 shows that the leading edge of the LII temporal profile appears earlier for more compact and larger aggregates. Bambha and Michelsen [130] were able to reproduce these CW-LII temporal profiles with an LII model that explicitly accounted for a distribution of surface areas for primary particles in an aggregate. The results of this study demonstrate increased shielding with increasing aggregate size and fractal dimension.

3.4. Influence of coatings

Soot particles released into exhaust streams can be coated with semi-volatile liquid coatings condensed onto the surface, which can be comprised of a number of components, including unburned fuel, lube oil, sulfuric acid, water, and other combustion by-products [330]. Fresh uncoated soot particles are hydrophobic, but particles emitted into the atmosphere can accumulate semi-volatile coatings that can allow them to become hygroscopic and effective

cloud condensation nuclei [2]. Following aging in the atmosphere, soot particles can have a very broad range of coatings, which can affect their optical properties and optical-detection methods, such as LII. Non-absorptive coatings on soot particles have been shown to increase scattering and absorption cross sections (see Sec 3.2.2). Coatings on soot particles have also been demonstrated to lead to significant changes in particle morphology. As detailed above, uncoated mature-soot particles have a branched-chain morphology typically described by a fractal dimension of 1.7–1.9 [11, 14-18]. When coated with oxygenated hydrocarbons or sulfuric acid, this morphology changes such that the particles collapse, the fractal dimension increases, and the mobility diameter decreases [16, 70, 96, 197, 298, 299, 301, 302, 304, 305, 331-334]. Depending on the coating, these changes are either reversible [299] or irreversible [70, 299, 301, 304, 333, 334] when the coating is removed by a thermodenuder.

Volatile coatings can also have an effect on LII signals [70, 97, 129, 197, 335, 336]. Case and Hofeldt [335] speculated that volatile coatings found on engine-exhaust particles could reduce LII signals at low laser fluences. Witze and coworkers [197, 336] used a modification of the LII model of Michelsen [176] to predict the effects of coatings on pulsed-LII signals. Their modeling results confirmed the hypothesis of Case and Hofeldt [335] and predicted that volatile coatings will have a significant influence on the magnitude of the pulsed-LII signal at low laser fluences and very little effect at high laser fluences. Preliminary experimental results with coatings of sulfuric acid confirmed these predictions [336] and indicated that the additional energy needed to volatilize the coating delays the heating of the particle to incandescence temperatures. At low fluences a large fraction of

the available laser energy is used to remove the coating, whereas at high fluences there is plenty of energy in the laser pulse to remove the coating and heat the particle to the sublimation temperature.

Similar results have been obtained for CW LII [97, 129]. Under these conditions there is typically plenty of energy to remove a coating and heat the particle to the sublimation temperature, but the process happens over microseconds. Moteki and Kondo [129] demonstrated that the effect of the coating for these studies is to delay the onset of LII signal in time until the coating has vaporized. An example of this effect is shown in Fig. 21. The results for an uncoated particle (shown in Fig. 5) are reproduced in Fig. 21a for reference. This figure shows that a coating of ~ 195 nm of oleic acid causes the LII to shift to later times relative to the scattering signal [129, 131].

Bambha *et al.* [70] have recently demonstrated that coatings of oleic acid on flame-generated soot shift the fluence curves for pulsed LII to higher fluence, i.e., higher fluences are required to heat the particles to a particular temperature when a coating is applied to the particle, as shown in Fig. 22a. The curves shift to higher fluence with increasing coating thickness until the coating is heavy enough ($>75\%$ by mass) that the laser-heated particles do not fully vaporize the coating, at which point the curves start to shift to lower fluence with increasing coating thickness, as shown in Fig. 22b. The restructuring of the particle with coating is nearly reversible if a laser removes the coating. The coatings must be heavy enough for rapid vaporization to forcefully open the particle, a process that also leads to particle fragmentation of primary-particle chains from the aggregate. Denuded particles shift to slightly lower fluences; the reason for this enhanced LII signal is likely the

reduction in conductive-cooling rate, caused by the reduction in the effective surface area, as discussed in Section 3.3.

4. Implementation of LII

4.1. *Excitation*

4.1.1. Laser wavelength

LII is typically initiated with a visible or near-infrared (IR) laser, largely because of the ready availability of high-energy lasers in these wavelength regions. The fundamental and second harmonic of the Nd:YAG laser are the most common wavelengths used, but longer wavelengths have been successfully employed [133]. The shorter the excitation wavelength and the higher the laser fluence, the higher is the probability of fluorescence from electronically excited evaporated species or from the gas-phase environment, and thus IR wavelengths are generally preferred. The suppression of interfering signal is simplified with excitation in the near infrared. In an unknown experimental situation it is helpful to check the emission spectra of the signal before choosing detection filters to ensure that no strong molecular emission bands are visible within the detection range on top of the broadband LII signal (e.g., [120, 159, 337, 338]). Wavelengths in the visible and ultraviolet (UV) region can excite electronic states of polycyclic aromatic hydrocarbons (PAHs), which then emit broadband fluorescence, particularly when the PAHs are vibrationally and rotationally hot, e.g., [339-344]. UV wavelengths can lead to fluorescence from a host of other species, e.g., [345] and references therein. Far UV wavelengths have been shown to preferentially lead to photodissociation of soot over particle heating and incandescence [272, 273].

Recommended filter wavelength ranges for soot detection are shown in Table 2. These spectral regions are generally chosen to eliminate the most prominent spectral interferences in soot measurements, which arise from Swan-band emission from excited C_2 at 468, 516, 550, and 580–620 nm [151, 337, 346]. High fluences can lead to soot sublimation and interferences from C_2 Swan-band emission at these visible and even near-IR wavelengths [120, 151, 159, 337, 338]. Swings-band emission from excited C_3 can also interfere with LII measurements at wavelengths of 360–440 nm [337]. Although LIF from PAHs and other species is commonly assumed to be red-shifted relative to the excitation wavelength, the fluorescence can be shifted to the blue if multi-photon excitation is involved or the fluorescing species end up in a lower rovibronic state than the one in which they started, i.e., they are anti-Stokes shifted [117, 151, 337, 347-350]. Polarization-discrimination approaches can be used to reduce LIF interferences from species such as PAHs [117].

4.1.2. Laser fluence

The LII signal depends nonlinearly on the particle temperature. For pulsed LII at low fluences, the maximum temperature reached by the particle is approximately linearly dependent on the laser fluence, i.e., ignoring minor heating and cooling mechanisms during laser heating, the maximum temperature reached $T_{\max}$ can be approximated by [119, 168]

$$M \int_{T_0}^{T_{\max}} c_s(T') dT' \approx \sigma_{\text{abs}} F_L, \quad (26)$$

where F_L is the laser fluence. For CW LII, conductive cooling is always competing with absorptive heating, and the expression to determine the peak LII signal is more involved.

Assuming that the specific heat is constant over this temperature range, performing the integration, substituting Eqs. (5) and (8) for the diameter and absorption cross section, and rearranging gives [283]

$$T_{\max} \approx \frac{6\pi E(m)F_L}{\rho_s c_s \lambda} + T_0. \quad (27)$$

The LII-signal magnitude increases nonlinearly with fluence until the particle reaches the sublimation temperature. The peak of the time-resolved signal becomes independent of fluence above the fluence at which $T_{\max}$ reaches the sublimation point. As shown in Eq. (27), the peak LII signal reached increases with fluence, and the fluence at which this plateau is reached depends on the laser wavelength, the initial particle temperature, and the particle composition, which determines $E(m)$, ρ_s , and c_s [119, 218, 260, 261, 269, 270, 283]. An example of the fluence dependence of the peak pulsed-LII signal and $T_{\max}$ is shown in Fig. 4 for laser wavelengths of 532 and 1064 nm.

At high fluences, the peak-pulsed-LII signal is independent of fluence for a laser beam with a homogeneous spatial distribution, but the signal-decay rate increases nonlinearly with fluence because the sublimation rate increases with increasing fluence. At these fluences, the gated-LII signal decreases because of the rapid decay rate. The fluence dependence of a gated signal thus depends on the gate width and timing relative to the laser. A gated signal tends to increase with fluence, reach a plateau (at $\sim 0.2 \text{ J/cm}^2$ at 1064 nm), and decrease at higher fluences ($> 0.2 \text{ J/cm}^2$ at 1064 nm) [67, 116, 177, 212]. Volume-fraction measurements are often made in the plateau region; this approach makes the experiment less sensitive to shot-to-shot variability in the laser energy and to attenuation through the

sample. High laser fluences also make the pulsed-LII measurements less sensitive to particle coatings [70]. An alternative approach is two-color LII, which usually employs lower fluence but measures particle temperature directly, compensating for changes in laser energy and the presence of coatings. Particle-sizing measurements tend to be made at low fluences to avoid the complications of sublimation and large changes in particle size during the laser pulse.

4.1.3. Laser spatial profile

The laser spatial profile can have a significant effect on the LII signal. From the perspective of signal analysis, a top-hat (i.e., homogeneous) spatial profile is conceptually the most straightforward configuration to implement because all particles at any point in the detection volume will receive the same laser fluence (see the inset in Fig. 1), but a top-hat profile may not be the simplest beam profile to use in practice. Some high-energy pulsed lasers generate a Gaussian or quasi-Gaussian profile, and the beam used for CW LII is also Gaussian. Such a profile can be turned into a top-hat in pulsed LII by selecting the center of the beam with an aperture or by using a specially designed beam shaper. A top-hat profile will distort as it is propagated in free space. As shown in Fig. 1, a series of two-lens telescopes can be employed to relay the image of the aperture or beam-shaper output to the detection volume without diffraction effects [351]. Although the beam will be collimated at the detection volume, the final image will have a finite depth of field. Starting with a Gaussian or quasi-top-hat multimode or injection-seeded high-energy pulsed laser and employing relay imaging of an aperture has been effectively demonstrated to produce a top-

hat profile that is maintained over a depth-of-field of several mm [70, 113, 117, 283, 352, 353].

For pulsed-LII volume-fraction measurements at high fluences, a one-dimensional Gaussian (i.e., sheet) profile can provide gated signals that are nearly independent of fluence [35, 116, 147, 159, 177]. For a two-dimensional (i.e., cylindrically symmetric Gaussian) beam, gated pulsed-LII signals have been shown to increase monotonically with increasing fluence [24, 150, 173, 177, 354]. The signal will decrease in the high-intensity center of the beam with increasing fluence as the particles sublime. At the same time the signal will increase in the low-intensity wings of the beam where particles have not reached the sublimation point [173, 177, 354, 355]. This effect was demonstrated by Delhay *et al.* [355] who recorded pulsed-LII signal in a backward configuration collinear with the laser beam. An example of their results is shown in Fig. 23 for several laser pulse energies using a Gaussian laser-beam spatial profile at 1064 nm. At the lowest laser energy, the maximum signal is seen at the peak of the laser spatial profile in the center of the beam. With increasing pulse energy, the region with detectable LII signal increases in size, and, at a high laser-pulse energy (right panel), the maximum intensity is moved to the wings of the laser spatial profile. These effects often compensate for each other for a Gaussian sheet. There is a higher fraction of low-intensity wings for a cylindrical Gaussian profile, and such a profile typically generates a gated signal that does not reach a plateau and continues to increase with increasing fluence [24, 150, 173, 177, 354]. Bladh *et al.* [212] have shown that a top-hat profile is predicted to yield the least error in soot-volume-fraction

measurements with gated detection at low fluences, and a two-dimensional Gaussian beam gives slightly less error than a top-hat or Gaussian sheet at high fluences (see Fig. 24).

4.1.4. Laser temporal and spatio-temporal profile

The vast majority of pulsed-LII measurements are made using high-energy pulsed Nd:YAG lasers with a 7–10 ns pulse duration. Many of these lasers are multimode, and temporal profiles tend to exhibit considerable mode structure. An example is shown in Fig. 25 for a laser with an unstable-resonator-cavity design; multimode lasers with stable resonators may exhibit even more complex mode structure [356]. This mode structure is random and varies from shot to shot. Injection seeding the laser provides a smooth temporal profile, which is compared with the multimode profile in Fig. 25. At low fluences the multimode structure has little effect on the LII signal. At high fluences the mode structure on the laser pulse leads to a minor decrease (of <10% for most fluences) in the average LII signal [357]. For most LII applications, an injection-seeded laser is not necessary. Injection seeding is only important if the data are being collected to validate a time-dependent LII model or study the fine details of the temporal evolution of the LII signal.

There are a large number of mechanisms that can influence LII-signal generation, and understanding these processes in detail requires collecting data under a wide range of conditions. Such data are necessary for validating LII models. Shorter pulses may be useful in isolating heating and cooling processes that naturally occur on different timescales.

Michelsen [353] used the second harmonic of a Nd:YAG laser with a 65-ps pulse duration and a streak camera with an 8-ps temporal resolution to study the time evolution of the signal on a shorter timescale. Relative to the laser pulse the LII signal behaves in time very

similarly to the typical nanosecond LII. The results suggest, however, that such short laser pulses may be more susceptible to generating interferences by multiphoton absorption [353]. Pulses with femtosecond durations can lead to electronic excitation on a timescale shorter than that at which electrons can thermalize with the lattice and heat the particle. In a bulk solid this process initiates rapid local heating and localized material ablation [358]. In a nanoparticle, significant ionization can occur, which can cause Coulomb explosion at high laser fluences [359]. Kaldvee *et al.* [360] recently used picosecond LII for enhanced range resolution in a single-ended detection geometry.

For volume-fraction measurements, a laser with a longer pulse duration may be more effective. Stephens *et al.* [132] developed an approach based on intracavity irradiation of single particles injected into the cavity of a CW laser. This approach allows particles to be heated to the sublimation point using a compact laser system. Figure 5 shows an example of LII and ELS temporal profiles compared with the laser-particle interaction time using such a system. The laser interaction time is determined by measuring the laser scatter from a non-absorbing particle as it drifts through the laser beam; this drift time is on the order of microseconds. The long interaction time provides sufficient signal for detection of single particles.

This approach to CW LII tends to be impractical for combustion systems where *in situ* measurements in flames, engine cylinders, or other combustors are required. The current implementation of this technique also provides reduced sensitivity to soot particles with a fractal dimension of 1.8 and a mobility diameter less than ~ 140 nm (see Sections 4.2.3 and 5.4), which makes it undesirable for most combustion measurements. Ditaranto *et al.* [361]

performed pulsed-LII measurements on soot in a flame using a laser with a variable pulse length and temporal profile. Their results demonstrate extended generation of LII over the pulse duration of up to 900 ns. These LII temporal profiles display interesting and unexpected behavior, i.e., a second unexplained temporal mode, which will require further investigation to characterize and understand. Black [362] demonstrated that a 1060 nm fiber laser with a high-repetition-rate CW-equivalent power of 100 W could be used to measure LII from soot in the exhaust of an aero-engine.

The high-energy pulsed Nd:YAG lasers typically used for LII measurements employ a cavity design with a geometrically unstable resonator configuration. This laser cavity is usually coupled with variable-reflectivity mirrors to produce high quality beam profiles [351]. This cavity design has been shown to generate a spatial profile that varies throughout the pulse [352, 363-365]. The timing of the pulse varies by nanoseconds with radial position in the beam. The center of the beam emerges from the laser first, and the edges of the beam can be delayed from the center by more than 10 ns. Passively Q -switched stable resonators and microchip lasers demonstrate similar spatio-temporal beam-profile behavior [366, 367]. The time-dependent transverse-mode structure in a multimode laser can also lead to evolution of the spatio-temporal profile with time, but this behavior is less predictable and is more complex for a stable resonator than for an unstable resonator [356]. Evolution of the spatio-temporal profile during the laser pulse can have a substantial effect on time-resolved LII and elastic scattering measurements if not taken into account in the analysis or measurement design [352].

4.2. Detection

LII detection depends to a large extent on the purpose of the measurements. Volume-fraction measurements are typically derived from the intensity information of time-integrated pulsed-LII measurements or accumulated histograms of peak CW-LII signals from individual particles. These measurements are implemented differently from pulsed-LII particle-size measurements based on time-resolved (TiRe-LII) measurements. An alternative approach is the absolute-intensity method, which employs time-resolved measurements at multiple wavelengths in the measurement of concentration (mass and volume), specific-surface area, and primary-particle diameter (see Section 4.2.1). Additionally, the approach for measurements of point-wise information is different from the approach used for one or two-dimensional imaging. Furthermore, there is a choice between single- and two- (and even three-) color measurements with the purpose of accurately determining particle temperature from pyrometry. In strongly absorbing environments, additional detection of attenuation or local-light intensity (through extinction measurements and measurements of local elastic scattering) has been used to evaluate local laser fluences. In all these approaches, choices must be made concerning integration times, time resolution of detectors, and detection-wavelength ranges, which can depend on the measurement situation. In strongly attenuating environments, effects of signal trapping on the results should be investigated.

4.2.1. Volume and mass fraction

For many initial applications of LII, the main focus was on the determination of the soot-volume fraction. Signal was collected on a photomultiplier tube (PMT), resulting in point

measurements [138, 150]. One-dimensional line images were acquired in turbulent flames via line-imaging onto a camera [149]. A common approach for pulsed LII, however, uses two-dimensional imaging with a gated intensified CCD camera equipped with appropriate detection filters, which detects signal from a plane illuminated by a laser-light sheet (e.g., [24]). Typical detector arrangements are orthogonal to the laser direction, at an oblique angle to increase the measurement volume [108], or in backward geometry for single-ended applications, such as the detection in exhaust plumes of jet engines [38]. Kaldvee *et al.* [360] recently investigated the use of LII for volume-fraction measurements in a configuration for range-resolved single-ended detection using LII on a picosecond timescale.

The underlying reason for the approximate proportionality between the maximum LII signal and soot-volume fraction is derived from the fact that soot primary particles are volume absorbers and emitters, i.e., in the Rayleigh approximation the energy absorbed and emitted by them scales with volume and not with surface as for macroscopic objects (see Sec 3.1.2). If all particles reach the maximum signal at the same peak temperature (i.e., the sublimation point), the peak signal will correlate with the volume or mass of the particle. Melton's original analysis of pulsed LII [138] included some dependence of signal magnitude on particle size, and relevant experimental parameters and conditions must be controlled or taken into account [113] to obtain a dependable measure for volume fraction. For gated pulsed-LII measurements, the choice of the detection gate width and timing relative to the laser pulse are important. To discriminate LII signal against LIF, e.g., from C₂ or PAHs, and elastically scattered laser light, delayed detection (relative to the laser

pulse) has been suggested. The comparably long pulsed-LII-signal decay can then be separated from luminescence from short-lived excited states and scattering [148, 162]. However, experimental [116, 153, 212, 368] and theoretical work [154, 369, 370] showed that long or delayed detection gates bias the LII signal towards the contribution from large particles because the signal from the small particles decays faster. Therefore, the most common compromise is to discriminate against interfering signals spectrally with appropriate filters, to use a laser wavelength in the near infrared (1064 nm), and to use short (50 ns) detection gates that overlap with the laser pulse (i.e., prompt detection). Using short-gate times is also desirable for high-pressure applications [177, 221]. Conductive-cooling rates increase with increasing ambient pressure and decreasing ambient temperature, thus speeding up the LII-signal decay rates and reducing the time-integrated intensity [177, 221]. Furthermore, for time-integrated detection, if calibrations are carried out under conditions that are different from the ambient application conditions, long and delayed-detection gates deteriorate the validity of the calibration [177]. However, with short-gate times imperfections in the performance of gated detectors can have an influence on the signal integration and hence the resulting volume-fraction information [371, 372].

Measuring the soot-volume fraction f_V based on the pulsed-LII signal intensity relies heavily on the knowledge of the dependence of the signal on laser fluence and of the local laser fluence itself. The dependence of pulsed-LII signal on the laser fluence is nonlinear and complicated (see Figs. 2, 3, 4, 15, and 22) and might vary with the maturity of the soot and may therefore change as a function of height above the burner or with particle composition between flame and exhaust measurements [284, 285]. A calibration against

alternative measurements (such as laser extinction or electron microscopy) is not universal for a wide variety of measurement situations. Common understanding is that, if fluence curves are similar everywhere in the zone of interest, a fixed proportionality factor between the LII signal and f_V can be assumed. If this requirement is not met (e.g., Figs. 15 and 22), correcting for the compositional effects of the fluence dependence of the signal is a challenge.

In measurement situations with significant laser attenuation, the local laser fluence changes, and, for relating measured signals to f_V , information about laser attenuation is necessary or highly desirable for reconstructing the local laser fluence and thus correcting the detected pulsed-LII signal. Approaches that use the local pulsed-LII signal as a measure of the local laser attenuation for corrections in a post-processing step provide an experimentally simple method, that is, however, based on many assumptions [155, 373]. Measurements of the total attenuation of the beam after passing through the interaction volume cannot fully correct for local variations of the laser fluence and their nonlinear influence on the LII signal. For relatively low levels of attenuation, one does not need to be concerned about the influence of attenuation on laser fluence. Measuring attenuation in an LII-experiment may not only help in monitoring the laser fluence, but can also serve as calibration for LII and offer additional information when combined with elastic scattering. The RAYLIX technique introduced by Bockhorn, Suntz, and co-workers [162, 374], see Fig. 26, records both variations in the light sheet intensity for fluence control and local extinction for the calibration of the LII-signal, and uses this information in a further step to evaluate particle

number densities and sizes from simultaneous measurement of two-dimensional Rayleigh scattering and LII.

Using laser fluences in the plateau zone (see Sections 4.1.2 and 4.1.3), where the pulsed-LII signal appears independent of the laser fluence, is a pragmatic way to circumvent the requirement of knowing local laser fluences. A plateau in fluence dependence of the pulsed-LII signal can be reached with Gaussian laser beams through the counteracting effects of a decreasing signal per unit f_V in the center of the laser beam as the particles sublime and the increasing size of the detection volume signal increases at the periphery of the beam (see Section 4.1.3). This method is frequently applied in practical situations for measuring f_V where laser attenuation is substantial and difficult to assess, e.g., due to strong spatial fluctuations of the soot distribution. In turbulent flames beam steering and trapping may be so significant that, even when working in the high-fluence regime, considerable effects on the derived soot concentration may result. Sun *et al.* [375] report on a possible underestimation in soot volume fractions of up to 30% in an atmospheric turbulent jet-flame. Figure 27 shows images of the spatial profile of a laser sheet beam after passing through an air jet (Fig. 27a) and through a turbulent flame (Figs. 27b-f).

To address some of the limitations with high-fluence pulsed LII that have been identified, a low-fluence pulsed-LII technique, autocompensating LII (AC-LII) can be applied. AC-LII employs time-resolved two-color pyrometry, low laser fluence, and an absolute-intensity calibration [107, 195, 369]. AC-LII measures the soot-particle temperature with two-color time-resolved pyrometry (see Section 4.2.2). Absolute calibration of the entire detection system, from the probe volume to the PMT, is achieved by placing a source of known and

traceable spectral radiance at the probe-volume location and recording the signals on the PMTs [107]. This approach results in a calibration factor for each PMT that relates the measured signal to units of spectral radiance ($\text{W m}^{-3}\text{sr}^{-1}$). With the absolute-intensity calibration and the temperature, f_V is determined as [369]

$$f_V = \frac{S_p}{\Omega \lambda w_b} \frac{\lambda^6 \left(\exp \left(\frac{hc}{k\lambda T} \right) - 1 \right)}{12\pi c^2 h E(m)}, \quad (28)$$

where S_p is the absolute-intensity-calibration factor, and w_b is the width of the laser beam in the sample volume. The mass concentration can be determined by multiplying the soot-volume fraction by the soot-material density, ρ_s . This technique allows determination of the soot-volume fraction and mass concentration without the need to correlate the result to another measurement of soot, such as extinction.

The use of low fluence (peak temperatures below 4000 K) in AC-LII ensures that substantial sublimation of the soot is avoided. The direct measurement of soot-particle temperature is necessary to permit the use of low fluence, as the LII signal varies dramatically with fluence in the low-fluence regime. However, by measuring time-resolved temperatures on a shot-by-shot basis, the technique automatically compensates for any effect that could cause a change in the peak-soot-particle temperature reached during the LII process. Possible effects that cause variations in the measurement environment include fluctuations in local bath-gas temperature, laser energy, attenuation of the laser beam, and the presence and quantity of condensed volatile material. To ensure that all soot particles are exposed to the same laser fluence and thus heated to the same peak temperature,

methods described in Section 4.1.3 are applied to generate a homogeneous laser-beam spatial profile.

CW-LII measurements are also generally made in the plateau region to ensure that particles reach the sublimation point. Using lower laser powers tends to reduce sensitivity to smaller particles and to particles with semi-volatile coatings [126, 376]. For CW LII compositional effects are a particularly serious challenge. Little work has been done to assess interference effects, and particle coatings have been shown to have a significant influence on temporal profiles [97, 128, 129]. The peak signal is used to measure particle size and mass fraction, and the impact of composition on the peak signal is currently not well understood.

Instrument calibrations are often performed with materials, such as agglomerated-fullerene particles, that do not mimic the composition and fine structure of combustion-generated particles, and these materials can give widely varying results [109-111]. Routine calibration with combustion-generated particles, e.g., Diesel exhaust, is also cause for concern because the composition of these particles is not well controlled and can be highly variable [110].

4.2.2. Primary-particle size

Determination of primary-particle sizes or primary-particle-size distributions with pulsed LII is based on the particle-cooling rate after laser heating, which results in a primary-particle-size dependent decay rate of the LII signal, measured using time-resolved detection (TiRe-LII) [36, 157, 166]. Other early applications that used this time-dependent signal variation focused on the two-dimensional detection of the LII signal at two different delay times after the laser pulse with two gated cameras [36, 157, 166]. Because cooling rates depend on the specific surface area, the ratio of the signal detected at two different delay

times is mainly determined by primary-particle size. At high laser fluences, signal decay rates may be strongly affected by sublimation of particles. Because of the uncertainties in modeling sublimation, measurements for primary-particle size should preferably be carried out in the low-fluence regime. In this regime, particle cooling is dominated by heat conduction. With heat conduction dominating, the temperature decay of an isolated primary particle may well be approximated by a simple exponential decay as the change of temperature with time is proportional to the temperature difference between the particle and the bath gas. The situation is more complicated for the LII signal decay, yet under certain conditions a single-exponential decay may be regarded as a first rough estimate as well [168]. Cenker *et al.* [377] have recently evaluated the errors introduced by applying this single-exponential approximation under various conditions.

In low-fluence LII the initial signal-decay rate is sometimes faster than expected based on current LII models. This effect has been called “anomalous cooling”, and the reasons for this behavior are currently not well understood [121, 245, 246]. One explanation for this behavior is that the thermal-accommodation coefficient and, hence, the conductive-cooling rate is enhanced for surfaces with relatively high hydrogen content; this enhancement decreases as the particle surface is oxidized and annealed while the particle is hot, i.e., during the initial part of the decay curve [218]. As a consequence, the initial part of the signal curve is sometimes neglected in the analysis.

The evaluation of the signal decay requires knowledge of the peak temperature, which is either calculated based on the soot absorption properties (see Section 3.1.2) or inferred from a pyrometric measurement of the particle temperature (see below). The determination

of primary-particle sizes is generally complicated by particle aggregation (see Section 3.3). If the aggregates are within the RDG-FA limit, large and small particles within these aggregates are heated up to the same final temperature, as suggested by Eq. (27). The measured or simulated temperature is then valid for all particles in the probe volume, provided that the probe volume is homogeneously illuminated. For some researchers, this assumption is an essential element of their analysis, and all particles are assumed to cool down from the same temperature. With increasing delay after the laser pulse, however, a dispersion in temperature develops because of the size-dependent cooling rates, and the measured effective temperature is biased by the larger primary particles [206].

Primary-particle sizes are generally inferred by comparison of measured pulsed-LII signal decays with simulated ones based on an LII model, e.g., [166, 180, 181, 185, 217].

Determining the distribution of primary-particle sizes from the decay rate of the pulsed-LII signal, however, is an ill-posed problem because very different distributions may result in similar signal traces. As pointed out in [113], the mathematical reason for this ambiguity is that a superposition of similar nearly exponentially decaying functions yields an almost exponential decay. During cooling, a particle ensemble with an initially homogeneous temperature distribution becomes inhomogeneous. Small particles cool faster than large ones, and, therefore, the late decay is determined by the large particle fraction. Rather than a direct inversion, the usual approach in time-resolved LII is to use a parameterization of the distribution and to determine the characteristic parameters of such a distribution.

For primary-particle sizes, lognormal-size distributions are regarded as a good approximation for various flame conditions [35, 113, 214, 222, 378], and the distribution is

typically narrow [319, 379]. However, a lognormal-size distribution may not always be the best assumption. Normal distributions are also often employed. Banerjee *et al.* [380] recently pointed out that multi-lognormal distributions may show better agreement for some specific environments. Several approaches have been developed that correlate characteristic parameters of a distribution, e.g., the count-median diameter (CMD) and the width of the distribution (σ), with coefficients from a fit to the LII signal decay, e.g., [214, 377, 381, 382], often with an emphasis on the analysis of various phases of the decay. Figure 28 shows primary-particle-size distributions measured using TEM images from samples collected at different HAB in an atmospheric ethylene-air laminar-diffusion flame. LII-signal-decay curves at these locations were fit assuming a monodisperse distribution of primary-particles sizes (green vertical lines). The data were also sequentially fit at times >300 ns to estimate the contribution from large primary particles and at times <300 ns to estimate the contribution from small primary particles [377]. At high pressure, the situation becomes more complicated because significant conductive heat-loss occurs during the laser pulse. Therefore, small particles in the ensemble reach lower peak temperatures compared to large ones, causing a systematic error in the experimental determination of temperature and thus in the data analysis that assumes initially homogeneous temperatures. An approach to estimate the distributions of primary-particle diameter based on the decay of the effective temperature identified that the initial temperature-decay rate of a soot particle ensemble at the moment of the peak-soot-particle temperature is inversely proportional to the Sauter mean diameter d_{32} of the poly-disperse primary-soot particles. The distribution width is then determined from the decay at late times [214]. A survey and comparison of various

methods for the determination of particle-size distributions has been given by Daun *et al.* [383].

Particle cooling is even more complicated in aggregates because the individual particles are shielded by their neighbors, which makes the cooling rate dependent on the number of primary particles per aggregate and aggregate morphology (see Section 3.3), and the effects of the primary-particle-size distribution may in practice be masked by the aggregate effects. In cases where particle morphology may vary, LII may need to be combined with ELS to determine effective aggregate surface areas as input to the LII-decay analysis, see Section 4.2.3. The analysis is complicated by the fact that typically aggregate-size distributions in flames are rather broad [384].

Measuring primary-particle sizes at elevated pressures and low temperatures can be particularly challenging because the enhanced conductive-cooling rate strongly influences the particle-energy balance during the laser pulse [176, 220, 385, 386]. Because of the strong desire to understand soot formation under high-pressure conditions, considerable work has been devoted to developing pulsed-LII primary-particle sizing for these conditions [220, 222, 223, 385-387].

The competition between the conductive-cooling rate and absorptive-heating rate may be the major limiting factor in the detection sensitivity of small particles, even at atmospheric pressure and elevated temperature. A number of studies have demonstrated the use of pulsed LII to measure particles that are just a few nanometers in size at atmospheric and lower pressures and at slightly elevated pressures [95, 156, 169, 247, 284, 285]. Most of these measurements were made at elevated temperatures as well. Several of these studies

have made use of pulsed LII to study soot formation and growth in flames and shock tubes [95, 169, 247, 284, 285], taking advantage of the ability to make *in situ* measurements without perturbing the combustion system by sampling. The only restrictions on the use of pulsed LII for studies of soot formation are that (1) the particles must be refractory and (2) the particles must absorb the laser light. Results of Minutolo, D'Alessio, and co-workers [388-391] suggest that incipient hydrocarbon particles are transparent to UV and visible light. Absorption cross sections are even smaller at longer wavelengths, e.g., [259]. Semi-volatile hydrocarbon particles are unlikely to be able to withstand temperatures 1000-2000 K above flame temperatures required for LII detection [392]. Grotheer *et al.* [92] have demonstrated that hydrocarbon-based incipient soot particles vaporize or dissociate during laser heating. The results of Stirn *et al.* [95] similarly suggest a sensitivity to soot particles >3 nm but negligible sensitivity to hydrocarbon-based incipient particles in this size range. The balance between conductive cooling and absorptive heating is likely to have a much more significant impact on the detectible size limit for CW LII and for pulsed LII at high pressures and/or low temperatures.

For point measurements the entire pulsed-LII-signal-decay curve can be measured with fast PMTs in combination with a transient recorder, such as a digital storage oscilloscope. The temporal resolution must be sufficient to capture the decay, which is on the order of a few hundred nanoseconds for soot in atmospheric-pressure flames, but is shorter at higher pressures or lower ambient gas temperatures. At low fluence, for which decay rates are relatively slow, time-resolved measurements can be made as coarsely as in 10-ns intervals, although steps of 1 ns or less are often used.

Typical “fast” PMTs have rise times of 1–1.5 ns and cannot closely follow the rapid signal increase during the heating of soot. Therefore, the signal evaluation is often restricted to the signal decay after the signal peak. In this case, for an accurate evaluation of the pulsed-LII decay curves, the peak-particle temperature $T_{\max}$, which occurs during or at the end of the laser pulse, must be provided as an input parameter for the LII-signal model. Calculating this temperature based on the absorption cross section of the soot particle, the laser fluence, and the photo-induced processes during laser heating is associated with large uncertainties. Therefore, the time-dependent temperature [107] or the peak-particle temperature [180, 393] is often measured via two-color pyrometry [107, 180, 393]. The pulsed-LII signal is detected at two wavelengths or wavelength ranges λ_1 and λ_2 . Assuming a top-hat beam profile and identical temperatures for all particles in the ensemble, the signal ratio at these two wavelengths is related to the particle temperature [106, 394] according to

$$T = \frac{hc}{k_B} \left(\frac{1}{\lambda_2} - \frac{1}{\lambda_1} \right) \cdot \left[\ln \left[\frac{S_p(\lambda_1, T)}{S_p(\lambda_2, T)} \frac{K_2(\lambda_2)}{K_1(\lambda_1)} \frac{\varepsilon(\lambda_2)}{\varepsilon(\lambda_1)} \frac{\lambda_1^5}{\lambda_2^5} \right] \right]^{-1} \quad (29)$$

Here, h is the Planck constant, c is the speed of light, k_B is the Boltzmann constant, S_p is the detected emission signal from the particles at the two detection wavelengths, and K_1 and K_2 are calibration constants that account for the spectral sensitivity of the detectors at the two wavelengths. The emissivity, ε , given in Eq. (7), is a function of the refractive index of soot. The absolute value of $E(m)$ is not required here; only the ratio of $E(m)$ at the two detection wavelengths is needed. The ratio $\varepsilon(\lambda_2)/\varepsilon(\lambda_1)$ can be approximated by λ_1/λ_2 assuming that $E(m)$ is constant between the two wavelengths. The validity of this approximation can introduce some uncertainty to the inferred temperatures [118, 395]. A careful choice of the

detection wavelengths is necessary. A theoretical study by Liu *et al.* [124] compares the different strategies and suggests the 400/780 nm combination based on the respective temperature effects for low to moderate fluences. They especially stress the advantage of using a short-wavelength option for the blue channel. This study, however, did not consider the effect of spectral interferences at higher fluences that could influence the choice of the detection wavelength in a particular experiment. The reasons for the experimentally chosen wavelength combinations therefore mainly are to avoid interferences from C₂ Swan-band emission (see Sections 4.1.1 and 4.2.1) and to choose a spectral range where the detection system is most sensitive. For (the commonly used) detection at 450 and 550 nm, C₂ interference can be relevant for wide bandpass filters. There is no universal best solution. Depending on the measurement situation, the potential contribution of interfering species, and the efficiency of the detection scheme (such as collection solid angle and the angle between the laser propagation direction and the detection axis), a compromise must be made between temperature sensitivity and signal-to-noise ratio. To increase signal collection at long wavelengths, it may also be advisable to use a longpass filter instead of a bandpass filter [396].

TiRe-LII measurements [380, 383] with PMTs are limited to pointwise detection. The spatial distribution of particle sizes can be revealed only with repeating the measurements at different locations; this approach is not possible for rapidly fluctuating situations like those in unsteady flames. Will *et al.* [36, 157, 166] developed a strategy for two-dimensional particle sizing, where pulsed-LII signals are acquired at two delay times after the laser pulse with gated intensified cameras and where the particle size is deduced from

the local signal ratio with the obvious restriction that only average particle sizes can be obtained. This technique provided particle-size distributions with high spatial resolution for laminar flames at atmospheric pressure. For elevated pressure, Cenker *et al.* [385] presented a theoretical study of the best choice of detection parameters for two-time-step detection. Streak cameras allow TiRe-LII measurements along a line [397], and the technique has been recently extended to multi-time-step detection with a multi-chip camera, which improves the possibilities of reconstructing the LII-signal decay, as has been demonstrated in single-shot imaging measurements by Sun *et al.* [398] in turbulent flames.

4.2.3. Aggregate size

Although there may be a significant influence of aggregate size and structure on time-resolved pulsed-LII results (see Section 3.3), depending on the heat-conduction regime, the primary-particle size is the main size parameter to be determined from pulsed LII. TiRe-LII can be a unique and powerful technique in this context. Nonetheless, measuring aggregate size is equally important for developing a full understanding of particle formation; knowledge of aggregate size and morphology is also vital for an improved measurement of primary-particle size via LII. The aggregate-size distributions are typically assumed to be log-normal, although it has been suggested that a double log-normal may be a better descriptor for soot aggregates [72].

Besides various approaches for a direct determination of aggregate sizes and morphologies from light scattering alone [15], there are a few combinations of LII and light scattering that allow for a measurement of aggregate size. With the possibility of measuring volume fractions by LII, the combination of LII and light scattering may provide the diameter of a

volume-equivalent sphere, similarly to scattering/extinction measurements. Quay *et al.* [24] used this approach for pointwise measurements of soot-particle diameters in a diffusion flame and obtained good agreement with previous results from scattering and extinction. Will *et al.* [36, 157] performed two-dimensional measurements on a diffusion flame and could obtain a complete qualitative picture of aggregate sizes in such a flame; no calibration of scattering was performed to provide quantitative analysis. Though addressing the determination of aggregate sizes, these initial experiments, however, did not account for an explicit structure factor for light scattering. Later Reimann *et al.* [399] took up these early approaches and performed a quantitative 2D determination of aggregate sizes in the form of radii of gyration R_g in a premixed flame (see Fig. 31). They pointed out that these measurements are limited to a maximum of $qR_g \approx 1$, with q the modulus of the scattering vector, as a general restriction from the dependence of the structure factor on aggregate size. For larger values of qR_g , the sensitivity of the structure factor on particle size is drastically reduced. With their conventional perpendicular arrangement of laser sheet and direction of observation, the maximum average size accessible was limited to about 60 nm. Ma and Long [400] recently extended this approach and combined two-dimensional multi-angle light scattering with multi-wavelength line-of-sight attenuation and previous TiRe-LII measurements to obtain a comprehensive characterization of particle parameters and soot optical properties in a co-flow laminar non-premixed flame. Snelling *et al.* [29] used a compact set-up combining pointwise LII and light scattering at an angle of 35° . This approach provided sensitivity to particle sizes of about 200 nm. They compared radii of gyration from this combination with those obtained from two-angle light scattering with

good agreement for a given form of the particle distribution. Crosland *et al.* [401] extended these studies using LII and two-angle ELS to infer soot-volume fraction, primary-particle diameter, and aggregate radius of gyration simultaneously. Compared to pure light scattering, integrating LII into these measurements on aggregate sizes has the obvious advantage that other relevant quantities, such as concentration, primary-particle size, and fractal dimension, become accessible. Aggregate sizing from light scattering alone, however, does not rely on the refractive index. A simultaneous application of both approaches thus allows for a consistency check regarding m .

Where applications allow for extractive sampling from combustion systems or the ambient atmosphere, CW LII provides the capability for measuring aggregate size on a particle-by-particle basis. Measurements using the SP2 allow resolution of mass distributions over mass ranges of ~ 0.5 –300 fg [126, 130]. This mass range is equivalent to aggregates with $N_p \approx 8$ –5000 primary particles with diameter $d_p = 40$ nm. For a typical fractal dimension D_f of 1.75, this equivalent aggregate would have a mobility diameter of ~ 110 –2000 nm, based on the relationship [55, 402]

$$D_m = d_p N_p^{0.46}. \quad (30)$$

Although this technique lacks sensitivity to particles on the high and low end of the size spectrum, it has the advantage that it allows measurements of individual particles, which is very attractive for studies of inhomogeneous or broad particle distributions. Volume fractions or mass distributions are derived from histograms of signals from individual particles. Figure 29 shows a comparison of the mass distribution of soot sampled from a co-flow diffusion flame measured by the SP2 and by a centrifugal particle mass analyzer

(CPMA) [130]. The corresponding mobility size distribution, measured with an SMPS, is also shown. Particles were size selected for a mobility diameter of 125 nm and had a fractal dimension of ~ 1.9 . Figure 29 demonstrates excellent agreement between the CPMA and SP2 mass distributions at particle masses between 0.52 and 10 fg and some discrepancies at masses lower than 0.52 fg and mobility diameters smaller than 140 nm.

4.2.4 Particle composition and mixing state

Because LII has some sensitivity to soot maturity (see Sections 3.2.1 and 3.2.2) and volatile coatings (see Section 3.4), LII signal behavior can potentially provide compositional information. This area of development is much less mature than other applications, such as primary-particle sizing or volume-fraction and mass-distribution measurements. Mixing state refers to how components of an aerosol distribution with an inhomogeneous composition are combined. In an external mixture of particles, soot particles mingle with other materials as separate and distinct particles. In an internal mixture, soot particles are mixed with other materials within individual particles. With respect to internal mixtures, small soot particles could be well mixed with other materials within each particle; they can exist in a core-shell configuration with soot sitting in the center of the particle surrounded by a coating; or they can be segregated from the other material, perhaps attached to the surface of a particle of another material. The mixing state of soot has a strong influence on its optical properties and is of great interest for climate studies, e.g., [2, 295, 403, 404].

Early in the development of pulsed LII for applications to engine exhaust, Case and Hofeldt [335] noted that the volatile coatings found on most engine-exhaust soot particles could perturb pulsed-LII signals, particularly at lower laser fluences and in the wings of Gaussian

laser beams at all fluences (see Section 3.4). They combined LII with elastic-light scattering (ELS) measurements and demonstrated differences in the relative response of these techniques with engine load. They suggested that the combination of LII and ELS could be used to determine the volatile fraction of particle mass by measuring the delay in the onset of LII caused by vaporization of the volatile coating.

Witze *et al.* [196, 197] developed an extension of pulsed LII called laser-induced desorption with elastic-laser scattering (LIDELS) to measure soot-volume fraction and volatile-coating fraction of exhaust particles in real time. This technique combines LII and ELS using two laser pulses of comparable energy separated in time. The first laser pulse provides an ELS signal for inferring the total particle-volume fraction. This pulse is also absorbed by the soot core particles, causing the particles to heat sufficiently to desorb the volatile coating. The ELS signal from the second pulse is used to infer the volume fraction of the non-volatile core particles. LII is recorded simultaneously with the ELS signals for complementary measurements of volume fraction of the non-volatile core particles.

Using pulsed LII, López-Yglesias *et al.* [218] propose exploiting the shift in fluence curves to higher fluences for less mature soot (e.g., Fig. 15) as a proxy for soot maturity. Similarly, it might be possible to determine the presence of volatile coatings from the shift to higher fluences of the fluence curves caused by such coatings (e.g., Fig. 22). Coupling pulsed LII with ELS for these applications might allow the effects of soot maturity to be isolated from the similar effects of volatile coatings on fluence curves. In any case, much more work needs to be done to determine whether such an approach will be viable.

CW LII can also provide information about volatile coatings. As shown in Fig. 21, temporal LII profiles recorded by the SP2 are delayed in time when significant amounts of volatile coating are present on the particles, and some studies using the SP2 instrument have been focused on extracting this information [128, 129]. Detection of scattered laser light as the particle traverses the intra-cavity beam provides additional information about the particle as it heats. If the particle has a significant volatile coating, some of the energy absorbed by the particle will be lost to vaporizing the coating, and the particle will take longer to reach detectable incandescence temperatures. The LII signal will be delayed in time, but the scattered signal will not be susceptible to such a delay. The difference in time between the LII and scattering signals can be used to determine whether or not the particle has a thick coating [97, 128, 129]. This technique is promising but has not yet been shown to be quantitative. In addition, because of conductive-cooling effects, small aggregate sizes and open aggregate morphologies (e.g., fractal dimensions of ~ 1.8) can lead to LII-signal delays for uncoated particles similar to those observed for coated particles; these effects can mimic the effects of volatile coatings and complicate the analysis [130]. Schwarz *et al.* [405, 406] identified coated particles by a reduction in the scattering signal prior to the peak in the LII signal, suggesting that, as the particle heats, the volatile coating is vaporized, and scattering decreases.

Recent results have suggested that CW LII can be employed to derive additional information about particle-mixing state. As noted above, LII signals of soot particles coated with a semi-volatile or volatile coating will be delayed in time as the coating is vaporized from the laser-heated particle. This time delay is called the "lag time" [407, 408]. Sedlacek

et al. [407] observed that, under some conditions, LII signals can appear early relative to the laser-scatter signal, as shown in Fig. 30, i.e., they demonstrate negative lag times. Sedlacek *et al.* [407] hypothesized that negative lag times may be attributable to soot particles sitting on the outside of larger particles; they can heat and vaporize early during the laser-particle interaction time without conducting too much heat to the other material within the particle. Generally, the scattering signal is detected much earlier than the LII signal for particles with mixed composition, as shown in Fig. 21. Figure 30 shows an example of a particle for which the LII signal appeared well before the peak of the scattering signal. Although negative lag times have been observed by other groups, e.g., [408], they are not common, and further work is needed to firmly identify the cause of negative lag times.

5. Applications

5.1. Flames

Pulsed LII has been applied to a broad variety of flames. Atmospheric laminar flames have played the most prominent role in the development of the technique, and two types of flames are of particular importance. Jet-diffusion flames have been used extensively to demonstrate the favorable characteristics of pulsed LII for the two-dimensional visualization of the sooting properties of flames. Premixed burners, mostly of a McKenna type, have also been employed for the development of various implementations of LII as these flames show an essentially one-dimensional character, i.e., to a first approximation, properties of the flame only change with height above the burner. An intercomparison of data derived by LII from several groups for three different burners (McKenna burner with a

stabilization plate; two diffusion burners – Gülder and Santoro laminar co-flow burners) is documented by Schulz *et al.* [113].

For laminar diffusion flames, a number of early papers investigated the visualization and quantitative measurement of soot concentration in an imaging fashion [116, 149, 159, 409]. Employing pointwise measurements, Quay *et al.* [24] addressed the combination of LII and light scattering for the determination of volume-equivalent diameters of soot aggregates. Soon thereafter, Will *et al.* [36, 157] succeeded in a two-dimensional measurement of primary-particle sizes in an ethylene diffusion flame. Using LII combined with light scattering, they were able to describe the characteristic features of the formation, aggregation, and oxidation of soot particles. Many other investigations employing laminar diffusion flames followed; recent examples include a study on the relationship between the prompt LII signal and volume fraction [410], the application of two-color LII in an imaging fashion [395], and the combination of LII with light scattering [29] and extinction [119]. Diffusion flames have often been the object of investigation under the especially challenging conditions of microgravity, e.g., [164, 370, 411], with LII allowing for an efficient assessment of the differences of soot formation in buoyant and non-buoyant flames.

Likewise there is a long history of pulsed-LII usage in laminar premixed flames with a number of early studies addressing fundamental issues of LII, e.g., [35, 151, 159]. Some recent examples include studies focusing on the combination of pulsed LII and light scattering [29, 213, 399] (see Fig. 31), a basic assessment of the method, a study of the

fluence effects using LII and ELS [283], a comparison with other techniques [412], and a measurement of optical properties of soot [247].

There have been a number of LII studies on these types of flames at both low and high pressures. Investigations at pressures as low as 12 kPa, e.g., [180, 261, 413], have focused on issues of calibration of LII for low volume fractions, the extension of the soot formation zone for ease of observation, and the general features of soot formation under these conditions. There are a larger number (e.g., [221, 414, 415]) of LII experiments at elevated pressures up to 4 MPa with a focus both on LII modeling under these conditions and on the influence of pressure on the sooting characteristics of the flames investigated.

Generally, pulsed LII has been widely applied for the fundamental investigation of soot formation in flames and the development and testing of numerical models. Desgroux *et al.* [347] recently presented an excellent review on the use of LII and other optical methods in this context, and we refer the interested reader to this paper. Applications include the characterization of the soot propensity of various fuels as derived from the soot-volume fraction measured by LII and frequently used, e.g., by McEnally and Pfefferle [416, 417] and the concurrent theoretical and experimental description of soot distributions in non-premixed flames [418-420].

Given the ability of pulsed LII to provide instantaneous two-dimensional characterization of the soot distribution, particular interest is directed at measurements in turbulent flames. Simple jet-diffusion flames are often used for these studies, which range from visualization [147] and measurement of soot-volume fractions [116] in the early years to the statistical analysis of soot parameters [421] and combinations of pulsed LII with other optical

techniques for a simultaneous determination of temperature, species concentration, and flow fields [185, 422-424]. Amongst the multitude of flames investigated by LII, pool fires with diameters up to 2 m constitute both particularly challenging and spectacular case studies [425, 426]. Other recent applications of pulsed LII include investigations on the soot-turbulence interaction in the soot-formation region of non-premixed jet flames [427] and the development of surrogates for real fuels [428] and of pulverized coal flames [429]. Pulsed LII is also well suited for studies in turbulent flames because of the ability to perform single-shot imaging under many conditions. Pulsed LII can also be coupled with other imaging techniques, such as planar LIF (PLIF) detection of selected gas-phase species. Figure 32 shows an example of UV LIF detection of PAHs coupled with pulsed LII.

5.2. Internal combustion engines

Reducing soot formation is an important goal for the development of modern internal-combustion engines. Soot originates from diffusion controlled (Diesel-like) combustion and pool fires. While traditionally the focus was on Diesel engines only, soot emissions from direct-injection gasoline engines recently became an important topic in the light of new emissions legislation. Because the soot emitted from engines depends in a complicated way on the in-cylinder conditions, exhaust-gas sampling measurements do not provide the information that is required for understanding the formation process in sufficient detail. The soot that is emitted from engines depends on two consecutive processes, soot formation (and its spatial inhomogeneities) and soot oxidation. Soot oxidation can fail because of local quenching of the diffusion flame that surrounds (and, in the best case, burns) the soot or because of overall low temperatures in the expansion stroke that cause reactions to

become too slow for full soot oxidation within the time available. While soot formation has been visualized successfully based on the light emission during the hot combustion phase, e.g., in Diesel sprays, the soot oxidation phase tends to be invisible for this technique because gas-phase (and hence particle) temperatures are too low to generate sufficiently strong thermal radiation [430]. Therefore, there is a strong interest in using LII for quantitative and qualitative in-cylinder measurements. With respect to the field of combustion in Diesel engines, great interest has been devoted to the investigation of droplet and spray combustion. Papers in this field include studies on single, fiber-supported [163] or freely falling [116] droplets and soot formation from Diesel sprays in high-pressure chambers or rapid-compression machines [65, 223, 431, 432]. The applications of LII to exhaust measurements are treated independently in Section 5.3. Applications range from qualitative imaging that is used for developing conceptual models of soot formation and oxidation, quantitative imaging of soot-volume fractions, point measurements of particle size with the purpose of generating data for model validation, and fast measurements in the exhaust gas that outperform conventional soot-measurement techniques in terms of real-time capability and dynamic range. Quantifying LII from engine measurements is especially challenging because of the strong variations in ambient pressure and temperature depending on the timing of the measurement in an engine cycle.

Motivated by engine measurements and the goal of performing them under more “idealized” conditions, measurements have been carried out in shock tubes [165, 169], rapid compression machines [432], and Diesel sprays injected into high-pressure chambers [223, 431]. Often these experiments simplify quantitative studies that are aimed at the

development of models. These model experiments allow decoupling of the chemistry from mixture formation and turbulence and provide clean environments in which sprays are not influenced by wall effects, and windows remain clean for extended periods of time. From a diagnostics point of view, these experiments have the advantage of better control of measurement conditions, better optical access, and, because of the comparably high level of reproducibility (in contrast to typical engine cycles), improved options to correct for influences such as laser and signal attenuation.

Because of the large number of applications reported in the literature, it is not possible to fully cover all work. We will therefore present typical examples and pioneering work.

5.2.1. Qualitative imaging of soot-volume fraction

Early measurements demonstrated the applicability of LII to in-cylinder measurements in an optically accessible square-piston engine in combination with NO-LIF imaging [433]. Pioneering work about the temporal and spatial distribution of in-cylinder soot formation and oxidation was carried out by Dec and coworkers [140, 143, 434], which led to the formulation of a conceptual model of DI Diesel combustion [144]. Conceptual models for “classical” Diesel processes were expanded to cases with high air entrainment and exhaust-gas recirculation, where virtually no engine-out soot can be detected, and where the LII measurement showed that soot was only observed in the recirculation zones close to the bowl perimeter [435].

The simultaneous measurement of the distribution of OH LIF and soot (via LII) provided information about soot oxidation by the diffusion flame, and its failure once the flame is

quenched at the wall [436]. Figure 33 shows an example of OH and LII distributions in an engine cylinder; this example demonstrates that qualitative imaging measurements can be powerful for obtaining a conceptual understanding of processes.

While the measurements in Fig. 33 were made in individual engine cycles, the effect of exhaust-gas recirculation (EGR) in a heavy-duty engine was investigated with a sequence of LII measurements. For these measurements a laser cluster was used to generate a sequence of eight laser pulses for mapping the soot distribution with an 8-chip framing camera with a time resolution of 1° crank angle [437]. Because of the high repetition rate of laser pulses required, however, the measurements are perturbed by the measurement technology [438].

5.2.2. Quantitative imaging of soot-volume fraction

To obtain absolute soot-volume fractions from pulsed LII, signal intensities must be calibrated. Therefore, the accuracy of the measurement is strongly influenced by the accuracy of the calibration method. For calibration of pulsed LII measurements, in principle, two approaches are feasible. The semi-quantitative measurement, where the signal is proportional to the soot-volume fraction, can be compared to a calibration measurement using pulsed LII, e.g., in a standard flame. Alternatively, the measurement under the conditions of interest is compared to an independent evaluation of the soot-volume fraction using a different technique, e.g., laser extinction. In the first case, a calibration burner (that has been characterized before) must be placed in the detection volume without changing the optical setup. A more critical limitation, however, is that calibrating measurements in a high-pressure environment with an atmospheric-pressure flame, such as in [141, 352, 439],

can be problematic, not only because the pressure-dependent heat-conduction must be considered [222], but also because optical properties of the soot can be different.

Some of these uncertainties can be reduced by using light extinction as the calibration method. However, this technique depends on the knowledge of the refractive index of soot, which can differ significantly between different experiments [440]. For calibration with extinction, however, the experiment requires optical access from three sides, and beam steering can cause additional losses in transmitted signal. In addition, the LII measurement must cover the full interaction length where the laser light could potentially be attenuated to connect the measured attenuation with the integrated soot mass along the beam path.

Neither calibration strategy is ideal, but combining them can further reduce the associated uncertainties of each. In small-bore optical Diesel engines, additional problems arise from rapid window fouling. Additionally, measurements late in the cycle can be compared to exhaust-gas measurements of particulate concentration. In all cases, in-cylinder measurements will have significantly larger errors compared to measurements under stationary atmospheric-pressure conditions.

In addition to the variation of injection pressures, ambient density, and pressure [223, 441, 442], the effect of nozzle geometry [373], including cluster nozzles [443], has been investigated with pulsed LII in high-pressure combustion vessels, and quantitative data have been used for the validation of soot-formation simulations [444]. Because, in high-pressure cells, Diesel combustion can be decoupled from flow-field effects, the influence of fuel composition on soot formation has been investigated with pulsed LII [445].

Combinations of pulsed LII measurements with simultaneously performed laser diagnostics have been reported to investigate the interaction of fuel concentration and flows and to gain additional information about the soot field. The correlation of soot-volume fraction and mixture formation has been investigated with combined measurements of soot by pulsed LII and fuel concentration via laser-induced exciplex fluorescence imaging [446, 447]. The influence of bulk-flow structures on soot formation has been investigated by combined pulsed LII and PIV (particle-image velocimetry) measurements in a swirl-supported, direct-injection Diesel engine [448, 449]. Combinations of pulsed LII and Rayleigh scattering were used to determine relative soot diameters from in-cylinder [450] and high-pressure spray-chamber measurements [373].

Other applications include low-temperature Diesel combustion [451] and, with increasing regulations of particle emissions from spark-ignition engines, the interest in LII imaging has recently been expanded to gasoline engines. Direct-injection gasoline engines and engines with low-temperature combustion and high dilution with exhaust gases are of particular interest [430]. In the work of de Francqueville *et al.* [452] a combination of pulsed LII and laser extinction has been applied to gasoline direct-injection engines, where the extinction signal was used to calibrate the LII soot-volume-fraction measurements. Under these measurement conditions, peak-soot-volume fractions are typically lower than in conventional Diesel combustion. In contrast, the high soot-volume fraction in Diesel sprays can severely attenuate laser and signal light through absorption and scattering and thus influence quantitative measurements [453]. For improving the quantification of in-

cylinder soot-volume-fraction measurements, strategies for correcting for signal trapping have been developed for optically thick media [155, 414, 454-456].

5.2.3. In-cylinder Diesel particle sizing

Several experiments have focused on the determination of soot primary-particle sizes using time-resolved-pulsed-LII measurements. In-cylinder soot particle-size measurements were carried out in a single-cylinder Diesel engine. The pulsed-LII signal decay from the poly-disperse soot-particle ensemble was evaluated based on the assumption of either mono-disperse or lognormal [457] and lognormal or multi-lognormal particle-size distribution. It was found that the time dependence of the LII signal contains sufficient structure to allow retrieval of details of the particle-size distribution and that a multi-lognormal size distribution approach yielded the best results [380]. Based on this approach, the typical increase and decrease of soot-particle sizes in early and late phases of the combustion process were observed [458]. However, with increasing pressure, the relevant time domain becomes shorter and more difficult to analyze. At high pressure, conductive cooling during particle heating causes additional complexity, which might cause a particle-size dependence of the peak particle temperature at the end of the laser pulse [386]. Theoretical analyses of the effects for pressures up to 60 bar have been carried out by several groups [176, 220, 385, 386].

Figure 34 shows the results of in-cylinder particle-size measurements in a two-stroke Diesel engine [171]. The time-resolved-pulsed-LII signal was detected at 550 and 694 nm after excitation at 1064 nm with a laser fluence of 0.10 J/cm^2 . The count-median diameter (CMD) is in the range of 30 to 75 nm and increases up to a crank angle (CA) of about 10°

CA and decreases again towards a value of about 30 nm at 100° CA after top dead center (TDC). This behavior can be explained by particle formation and subsequent particle oxidation. The geometric standard deviation of the size distribution, σ_g , is constant at a value of 1.1 up to a crank angle of 70° CA and then increases towards about 1.32. The in-cylinder measurements with LII were compared with TEM measurements from samples collected with a thermophoretic sampler in the engine exhaust. Good agreement was demonstrated between the pulsed-LII-inferred sizes at 100° CA after TDC and the TEM determined primary-particle sizes in the exhaust gas.

An application to measurements in a heavy-duty Diesel engine was reported by Bougie *et al.* [458]. Boiarciuc *et al.* [459] have used two-color time-resolved LII measurements in a Diesel engine for the determination of soot-volume fraction and primary-particle diameters. In addition, particle-size measurements are finding their application in gasoline direct-injection engines, indicating the increased interest in understanding and avoiding soot emissions from this type of engine [460].

5.3. Exhaust gases

Pulsed LII has also been shown to be very useful for measurements of soot in engine exhaust. Case and Hofeldt [145, 335] demonstrated the advantages of pulsed LII for measuring soot in Diesel exhaust with high sensitivity for engine particulates, high time resolution relative to the exhaust stroke and associated transient conditions, and relative insensitivity to variable operating conditions. Snelling and coworkers [105, 106, 108, 146] and Witze and coworkers [105, 409, 461, 462] further developed the technique for Diesel and spark-ignition engine exhaust, and Schraml *et al.* [463-465] extended its applicability

to Diesel-exhaust-particle sizing using time-resolved pulsed LII. Jenkins *et al.* [466] and Black and coworkers [38, 467-469] similarly demonstrated the applicability of LII for aircraft-engine exhaust-particle measurements.

Engine-exhaust particulates are often coated with significant volatile coatings. As described in Section 4.2.4, Case and Hofeldt [335] combined pulsed LII with elastic-light scattering (ELS) measurements to study emissions as a function of engine load and noticed differences in the response of LII and ELS, which they attributed to volatile coatings. Witze *et al.* [196, 197] later combined pulsed LII with ELS in a technique called laser-induced desorption with elastic laser scattering (LIDELS) for real-time measurements of soot volume fraction and volatile-coating fraction of exhaust particles (see Section 4.2.4).

Commercial instruments, namely the LI²SA (Esytec) and the LII 200 and the LII 300 (Artium Technologies) have been evaluated against numerous other nanoparticle measurement instruments for the measurement of mass concentration in vehicle emissions [115, 470, 471]. In the study by Durbin *et al.* [470], pulsed LII was evaluated for potential application as an on-board emissions-measurement system for emissions from heavy-duty Diesel trucks. Work on gas-turbine-exhaust emissions measurement has led to an international-standard method that uses LII for the measurement of nonvolatile-particulate matter (nvPM) [472]. This standard procedure includes calibration of mass concentration by correlation with elemental carbon (EC) determined by thermo-optical analysis as prescribed in NIOSH 5040 [282]. Research to support this standard has shown, using extractive sampling from the exhaust to transfer particles to the instrument, that pulsed LII

is effective in measuring nvPM (solid combustion-generated nanoparticles) from commercial aircraft engines [473, 474].

CW LII has also been used for studies of exhaust gases from on-road vehicles (e.g., [199, 475]) and ships (e.g., [476]). These measurements tend to be made in the ambient atmosphere. Although they have some limitations on detection of particles in the size range typical of Diesel and gasoline engine emissions (see Section 5.4), they provide measurements with fast (1-s) time response and high sensitivity to a significant fraction of Diesel-engine particulate emissions and can provide highly valuable measurements, particularly when combined with complementary techniques.

5.4. Ambient atmosphere

Some early applications of pulsed LII focused on characterization of ambient particulates. Filippov *et al.* [156] subjected lab air to laser pulses and found that the size distribution obtained from the time-resolved signals was shifted to smaller values with increasing pulse energy, a fact they attributed to the possible disintegration of clusters. More recently, there have been a number of measurements of atmospheric black carbon by LII [125, 128, 198, 199, 477-481]. Lu, Brook, Smallwood, and coworkers [198, 199] developed a high sensitivity variant of the pulsed-LII approach pioneered by Snelling, Smallwood, Liu, and co-workers for use in combustion studies [107]. The soot-volume fraction detection limit for the instrument developed by Smallwood and co-workers [107, 198, 199], known as the high-sensitivity LII (HS-LII) instrument, is 8×10^{-15} , which is equivalent to 15 fg/cm^3 for a distribution of particles [198], and Migliorini *et al.* [481] recently developed a pulsed-LII instrument with a detection limit of 200 fg/cm^3 . The lower limit on particle size is currently

unknown, but several groups have pushed the size limit for pulsed LII to particle sizes of a few nanometers in diameter at elevated detection volume temperatures and pressures near atmospheric and lower [95, 156, 169, 247, 285, 369]. The SP2 CW-LII instrument described above has a detection limit of 10 fg/cm^3 (according to the DMT website).

Figure 35 shows a comparison of transportation-emissions measurements from the SP2 and HS-LII instruments [199]. Because the SP2 instrument may be insensitive to a significant fraction of particles in the size range emitted by some transportation sources, its measurements tend to be lower than those of the HS-LII instrument (by as much as 80%) under conditions in which the main sources of black-carbon particulates are predominantly light-duty gasoline vehicles [199, 475]. Agreement between the instruments is better (to within 40%) for conditions when the traffic contains a larger fraction of heavy-duty Diesel vehicles, and a greater proportion of the particles are within the measurement size range of the SP2. The contrast in the measurements aided in assessing the relative contributions of gasoline and Diesel vehicles to the detected emissions. These results are consistent with a studies by Holder, Yelverton, and coworkers [475, 482].

How well the SP2 captures these smaller particles depends on factors that include the alignment of the instrument and laser power [376, 483]. Figure 36 shows a comparison of SP2 instruments demonstrating the fall-off in sensitivity of the instruments for small-particle sizes. When the instruments are well aligned, and the laser power is high, the fall-off starts to occur at $\sim 0.5 \text{ fg}$, which is consistent with the results shown in Fig. 29. When the instrument is poorly aligned or the laser power is low (dashed lines), the instrument

loses sensitivity to relatively large particles of <2 fg, which corresponds to a mobility diameter of ~ 250 nm for a particle with a fractal dimension of ~ 1.8 (see Fig. 29).

Recent work has exploited the sensitivity of the SP2 instrument to coatings on atmospheric particles discussed in Section 3.4 [128, 129]. Attempts have been made to correlate the amount of volatile coating with the difference in time between the LII and scattering signals [97, 128, 129] or with changes in the scattering signal as the particle heats [405, 406]. The results are promising, but more work is required to verify that this technique can be quantitative. In addition, a study of mature soot extracted from a flame demonstrated that lag times used to infer thick coatings are similar to those observed for small aggregates relative to large aggregates and for particles with a fractal dimension of 1.9 relative to those with a fractal dimension of 2.3-2.4 [130], indicating that particle morphology and size effects can mimic the effects of coatings.

5.5. Engineered nanoparticles

The vast majority of LII applications have focused on soot, despite early investigations on the application of LII to non-carbonaceous material [156, 193]; even the very first LII report by Weeks and Duley [133] included a signal from an aluminum aerosol. Soot and other carbonaceous particles are especially suitable for LII application because of their high sublimation temperature of ~ 4000 K and their appreciable imaginary part of the refractive index. In combination, these two properties allow for high peak-particle temperatures, resulting in a pronounced thermal emission in the visible spectral range with convenient detection.

Because of an increasing interest in the synthesis of engineered nanoparticles in the gas phase, a wide range of materials may potentially become a subject for the application of LII. In this context an *in situ* measurement of particle sizes is of particular interest because it offers the possibility of process monitoring and, eventually, process control. The first such use of pulsed LII was described by Dankers *et al.* [484] and Sommer *et al.* [485], who applied the technique in various industrial production and test reactors for carbon-black manufacturing, as shown in Fig. 37. Because of the extremely high solid concentration in industrial reactors, as compared to flames, a different detection concept had to be employed. They used a collinear approach and a beam splitter to perform excitation and detection through the same window mounted to a flange in the reactor, which was continuously purged. In these investigations Dankers *et al.* [484] could demonstrate a high sensitivity (<1 nm) of the particle sizes measured to fluctuations and changes in the reactor settings. They also obtained excellent correlation of the sizes measured *in situ* with adsorption measurements, which allowed characterization of the specific-surface area of the final product.

Besides these successful measurements on particles similar to soot, only a few approaches are reported in the literature on other carbonaceous material. Allouis *et al.* [189, 486] investigated the formation of larger, micron-sized particles in Diesel and heavy oil spray flames, namely so-called cenospheres (hollow particles) and pleospheres (filled particles). They developed appropriate models for the LII signals from these particles and were able to discriminate between these particles and much smaller soot particles, which were present at much higher concentrations. They could also obtain reasonable estimates for the sizes of

various particles but concede that the accuracy of the sizes obtained is low, as one would anticipate in view of the very large parameter set used. Vander Wal *et al.* [194] performed an extensive study on the dependence of the pulsed-LII signal from carbon nanotubes on excitation and detection parameters. Mitrani and Shneider [487] performed studies of pulsed LII on multi-walled carbon nanotubes (MWCNTs) suspended in air in order to develop a diagnostic for *in situ* sizing. They observed that large MWCNTs cooled faster than smaller ones, which is opposite the behavior of soot, and concluded that MWCNT sizing will require a detailed heat-transfer model including nonstationary heat conduction, sublimation, defect formation, annealing, and oxidation. LII or similar approaches are sometimes used in the investigation of carbon-nanotube formation. For example, with the use of CW lasers in this process for the ablation of carbonaceous material from a solid target, the resulting thermal radiation in the plume is analyzed for particle temperature [488, 489]. Cau *et al.* [490] reported on the measurement of soot-particle sizes via time-resolved pulsed LII in such a process.

Metals comprise another class of nanoparticles targeted for LII investigations. Studies performed so far cover a wide range of materials, including silver [156, 491], copper [492], titanium [193], molybdenum [193, 493, 494], tungsten [193], nickel [495], and iron, e.g., [169, 193, 494, 496, 497]. In a relatively early study Vander Wal *et al.* [193] addressed the general viability of pulsed LII for selected metal materials and investigated the spectral signatures of these metal particles under various excitation and detection schemes. They highlighted the need for a careful interpretation of the spectra obtained because of the overlap between atomic and molecular emission on the one hand and incandescence on the

other hand. Vander Wal [68] also pointed out that laser-induced microplasmas of particles may produce a pseudo-blackbody emission, which might be misidentified as LII.

Additional complexities for application of LII to metals results from lower vaporization temperatures and melting points and variations of the heat capacity with temperature, particularly for some metals, such as iron [496]. With metal particles a laser-induced change in particle morphology is obvious, and laser irradiation may thus also be used to induce partial sintering in metal-particle aggregates [491, 495]. When aggregates from noble metals are subjected to laser radiation, however, one must be aware that plasmonic resonances may result in local field enhancements with the consequence of strongly nonuniform heating within an aggregate [491]. Applications of LII to metal-nanoparticle sizing are growing, but significant persistent limitations are caused with large uncertainty associated with values of the thermal-accommodation coefficient α_T for most combinations of particle material and bath gas. Effective-accommodation coefficients are usually obtained by matching the time-resolved-pulsed-LII signals to independently determined particle sizes [496, 498], and molecular dynamics (MD) simulations [499] might eventually provide *a priori* knowledge for future applications. Nevertheless, this approach requires a critical assessment of uncertainties and experimental-verification approaches.

For other classes of particles, material reports on successful LII applications are scarce. Eom *et al.* [500] and Sipkens *et al.* [494] measured the size of silicon particles as a representative of elemental semiconducting materials during the growth in low-pressure plasma reactors. For oxidic and ceramic materials a number of issues exist that obviously prevent a broader applicability; issues include adverse optical properties, a lack of

knowledge of important physical properties, including the thermal-accommodation coefficient, and the superposition of other emissions upon the LII signal. In an early investigation Filippov *et al.* [156] performed measurements on a TiN aerosol produced from commercial particles dispersed through a shock-tube set-up. Although a reasonable time-resolved-pulsed-LII signal could be observed, the particle sizes retrieved with a major mode near 100 nm were one order of magnitude larger than the specified primary-particle sizes. The authors ascribe this finding to the generation of very compact aggregates during the dispersion process. No published reports exist on LII investigations on silica (SiO_2) particles, the oxidic nanoparticles with the highest production rate, which is probably due to the negligible imaginary part of the refractive index in the visible and near infrared. A few studies, however, have been carried out for titania (TiO_2), the second-most industrially important oxidic particulate material. Sommer *et al.* [485] measured time-resolved pulsed-LII signals from nanoparticles produced in a laser-vaporization process, but could not evaluate these signals because of interference from signals of the original coarse material. For titania particles formed in a flame-synthesis process, Zizak and co-workers [501, 502] obtained characteristic signal-decay curves, which reasonably correlated with heights in the flame, but a spectral analysis showed that the signal is dominated by narrow-band emission rather than broad-band thermal radiation. They preferred to call their method “laser-induced emission” rather than laser-induced incandescence. Lehre *et al.* [397] succeeded in a quantitative analysis of LII signals from manganese and iron oxide particles produced in a laser-vaporization reactor. In this case effective-accommodation coefficients, which were not stated explicitly, were obtained by a fit to particles sizes obtained from TEM.

In summary, LII is very suitable for monitoring carbon-black production. When it comes to non-carbonaceous material, however, serious complications can limit the applicability of the technique. Despite more uncertain “optical” properties of other particles, future applications may be expected. In this context, however, knowledge of the thermal-accommodation coefficient and low-temperature phase changes represent significant difficulties for quantitative particle sizing by pulsed LII.

5.6. Suspensions

Although laser-induced incandescence is commonly linked to the measurement of nanoparticles in the gas phase, there are a large number of investigations that deal with the properties of nano-sized particles in a condensed phase after intense laser irradiation. As in the early work of McEwan and Madden [503], which investigates the change of the refractive index of a colloid after a nanosecond laser pulse, many studies use ink or more defined dispersions of carbon blacks. Other early papers include those of Mansour *et al.* [504] and Nashold and Powell Walter [505], which received hundreds of citations and deal with the application of these suspensions as “optical limiters”. The idea here is to find a medium so that the radiant flux transmitted increases linearly with increasing irradiance for low values and approaches a limiting value for high irradiance. Many of the studies in this context focus on the mechanisms of optical limiting and heat transfer, including the formation of a microplasma around the heated particles, the generation of acoustic waves and the formation of bubbles from the liquid evaporated [503-506]. The term “laser-induced incandescence” for the investigation of these phenomena in the liquid phase was

probably used for the first time by Zelensky in 1999 [507], who also performed related studies with borate glass doped with carbon particles [508].

In 2007, Sommer and Leipertz [509] carried out an investigation devoted to the pulsed-LII-signal decay times of carbon-black dispersions. For the ten carbon blacks employed, they found a linear correlation between the LII-decay time and the primary-particle size from TEM. They also identified a plateau region where both the maximum LII signal and the decay time did not depend on the laser fluence. In these measurements, which were performed for one type of carbon black, they also found lack of dependence of the decay time on the dispersion liquid, which was limited to ethanol, isopropanol, and methoxy-nonafluorobutane; these dispersion liquids exhibited distinctively different liquid thermal conductivities. As a tentative explanation for this behavior the authors mentioned similar heat conductivities in the gas phase. No model has been proposed, however, that would allow for the determination of particle size from LII in liquids. One must conclude that sufficiently comprehensive models for LII in liquids would certainly be even more complex than those for gases. Moreover, well-established methods exist for nanoparticle sizing in liquids such as Dynamic Light Scattering, which might explain the limited interest in further exploiting LII in this context.

There is a great deal of interest in measuring black carbon in snow, ice, and rain. CW-LII measurements have been used to make these measurements by aspirating a liquid sample of rainwater, melted snow, or melted ice containing black carbon [510-512]. After the nebulized particles are dried, they are measured by the SP2 as an airborne sample. Much of

the development in this technique is currently devoted to sample preservation and preparation and elimination of coagulation effects [513-517].

6. Current research and future directions

LII has become the workhorse for soot-volume fraction and mass-concentration determination in combustion and atmospheric applications, and its utility continues to expand into new application areas under a wider range of conditions. With additional work coupling LII with other techniques, its usefulness may be further extended into characterization of aggregate size, morphology, and composition. Considerable progress has been made in recent years on the development of a detailed understanding of LII. Nevertheless, uncertainties about the technique limit its reliability and quantitative application under some conditions, and further research will be required to narrow these uncertainties to advance the technique.

Of general importance is the determination of reasonable detection limits for volume-fraction measurements under a variety of measurement conditions. Of particular current interest is the minimum particle-size limit accessible by LII and the effects of particle composition and aerosol mixing state on LII signals. A related point of interest is associated with the volatility limit for particle detection, which defines the applicability of LII to incipient-soot formation or brown-carbon detection. This topic is related to understanding the efficiency with which LII can be used for measurements of different forms of pure carbon, such as amorphous carbon [110], which are likely to vaporize at much lower temperatures.

Effects of particle composition include the application of LII to materials other than carbon, which would allow application of LII to a wide range of manufactured nanoparticles.

Composition effects also include inhomogeneous particles. As discussed in Section 3.4, volatile coatings on carbonaceous-soot particles can strongly influence LII signals under some conditions, but the understanding of these effects is far from complete. These effects could have substantial implications for several LII applications, particularly for those associated with engines, exhaust streams, and the ambient atmosphere, where coatings are prevalent.

There are a range of uncertainties associated with time-resolved measurements and primary-particle sizing. One of the most important issues is the wide variability in results from different LII models [204], which are used to analyze LII data to derive primary-particle sizes, especially in the case of poly-disperse size distributions. Current efforts are being directed at determining the critical energy- and mass-balance mechanisms and how they should be implemented in models and developing model-validation protocols. Further work needs to focus on measuring the most important parameters included in these models, such as temperature-dependent thermal-accommodation coefficients of soot in various bath gases and temperature-, wavelength-, and soot-maturity-dependent optical properties of soot. As summarized in Section 3.3, one area of particularly active research focuses on the effects of aggregate size on primary-particle sizing. More recent work highlights the additionally important influence of aggregate morphology on pulsed-LII-signal-decay rates and, hence, on inferred primary-particle sizes. Because of the growing interest in engine applications, accurate modeling of LII and experimental verification under high-pressure

conditions is critically important. Similarly, there is a need for reliable LII models for low-temperature conditions relevant to ambient particles and exhaust streams.

Another area of active research is the application of CW LII to ambient atmospheric particles and particles suspended in snow, ice, and rainwater. For all of these applications, reliable instrument calibration is critical and is subject to considerable uncertainties. More effort is needed to identify calibration standards with fine structure and characteristics similar to particles of interest. In addition, assessing the reliability of studies of particle composition and mixing state using CW LII is also an area of growing interest.

Optical (light extinction and scattering), probe-sampling, and online molecular-beam techniques provide complementary information about soot-particle sizes, morphology, composition, and mixing state. The benefits that are connected to their combination with LII have not been fully explored. With the aim of fully characterizing the properties of gas-borne particles, the field is open for further development of useful combinations of techniques that might be included in easy-to-use instruments, e.g., for atmospheric and exhaust-gas analysis.

While LII is mostly understood as a method to investigate a *per se* unknown particle ensemble based on a good physical understanding of the underlying physical processes and parameters, it also has the potential to be seen in the opposite direction. Once well-defined particle samples are available (well-characterized in composition, morphology, and size distribution), pulsed and CW LII provide ways to probe physical characteristics and mechanisms.

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Summary of symbols and parameters

c	speed of light ($2.998 \times 10^8 \text{ m s}^{-1}$)
C_{ov}	primary-particle overlap parameter
c_s	specific heat of the solid particle ($\text{J kg}^{-1} \text{ K}^{-1}$)
C_p	molar heat capacity of air at constant pressure ($\text{J mol}^{-1} \text{ K}^{-1}$)
C_p^{CO}	molar heat capacity of CO at constant pressure ($\text{J mol}^{-1} \text{ K}^{-1}$)
$C_p^{\text{O}_2}$	molar heat capacity of O_2 at constant pressure ($\text{J mol}^{-1} \text{ K}^{-1}$)
C_v	molar heat capacity of air at constant volume ($\text{J kg}^{-1} \text{ K}^{-1}$)
Cl	concentration-weighted pathlength for extinction (m^{-2})
D_f	aggregate fractal dimension
D_m	mobility diameter (m)
d_{ij}	distance between two adjacent primary particles (m)
d_p	primary particle diameter (m)
d_{32}	Sauter mean diameter (m)
E_e	irradiance (W m^{-2})
$E(m)$	refractive-index function for absorption
$F(m)$	refractive-index function for scattering

F	laser fluence (J m^{-2})
f_v	soot-volume fraction
G	geometry-dependent heat transfer factor
h	Planck constant ($6.626 \times 10^{-34} \text{ J s}$)
k	imaginary part of the complex index of refraction
K	constant used to account for non-idealities of gases involved in sublimation
K_λ	calibration constant used to correct for Σ_λ
k_{ann}	rate constant for annealing (s^{-1})
k_B	Boltzmann constant ($1.381 \times 10^{-23} \text{ J K}^{-1}$)
k_f	aggregate fractal prefactor
k_{ox}	overall rate constant for oxidation for $2\text{C} + \text{O}_2 \rightarrow 2\text{CO}$ ($\text{s}^{-1} \text{ m}^{-2}$)
L	mean free path (m)
m	complex index of refraction
M	particle mass (kg)
m_e	mass of an electron ($9.1095 \times 10^{-31} \text{ J s}^2 \text{ m}^{-2}$)
n	real part of the complex index of refraction
N_p	number of primary particles in an aggregate
N_A	Avogadro constant ($6.0221 \times 10^{23} \text{ mol}^{-1}$)

N_d	number of defect sites in the particle
p_0	ambient pressure (Pa)
p_v	saturation partial pressure of subliming carbon species (Pa)
q	modulus of the scattering vector (m^{-1})
$\dot{Q}_{\text{abs}}$	absorptive-heating rate for a single primary particle (W)
$\dot{Q}_{\text{agg}}$	conductive-cooling rate for an aggregate (W)
$\dot{Q}_{\text{ann}}$	heating rate from annealing for a single primary particle (W)
$\dot{Q}_{\text{cond}}$	conductive-cooling rate for a single primary particle (W)
$\dot{Q}_{\text{ox}}$	oxidative-heating rate for a single primary particle (W)
$\dot{Q}_{\text{rad}}$	radiative-cooling rate for a single primary particle (W)
$\dot{Q}_{\text{sub}}$	evaporative-cooling rate for a single primary particle (W)
$\dot{Q}_{\text{therm}}$	thermionic-cooling rate for a single primary particle (W)
R	universal gas constant ($8.3145 \text{ J mol}^{-1} \text{ K}^{-1}$)
R_g	radius of gyration of the aggregate (m)
S	LII signal per primary particle (V)
S_p	LII signal for a collection of particles (V)
t	time (s)

T	particle temperature (K)
T_0	ambient temperature (K)
T_f	final temperature of a molecular distribution that has exchanged energy with a surface (K)
$T_{\max}$	maximum particle temperature (K)
U_{int}	internal energy of the particle (J)
W_a	average molecular weight of air (0.02874 kg mol ⁻¹)
W_j	molecular weight of species C_j ($j \times 0.012011$ kg mol ⁻¹)
W_v	average molecular weight of subliming carbon species (kg mol ⁻¹)
w_b	width of the laser beam in the sample volume (m)
α_M	species-independent mass-accommodation coefficient of vaporized carbon clusters
α_T	thermal accommodation coefficient
γ	heat capacity ratio of air
H_{ann}	enthalpy of annealing (J mol ⁻¹)
H_{ox}	enthalpy of reaction for $C + \frac{1}{2}O_2 \rightarrow CO$, which is equivalent to the enthalpy of formation of CO (J mol ⁻¹)
H_v	average enthalpy of formation of subliming carbon species (J mol ⁻¹)
ε_λ	emissivity at wavelength λ

η	primary-particle shielding factor
κ_a	thermal conductivity of the bath gas ($\text{W m}^{-1} \text{K}^{-1}$)
λ	incandescence wavelength (m)
λ_L	laser wavelength (m)
ω	single-scattering albedo
	factor accounting for the solid angle detected and the detection efficiency of the detection system
ϕ	work function for a charged soot primary particle (J)
ρ_s	density of the primary particle (kg m^{-3})
σ_{abs}	absorption cross section (m^2)
σ_{ext}	extinction cross section (m^2)
σ_{sca}	scattering cross section (m^2)
σ_g	geometric standard deviation
Σ_λ	spectral response of the detection system

Summary of abbreviations and acronyms

AAC	aerodynamic aerosol classifier
AC-LII	auto-compensating LII
AMS	aerosol mass spectrometer
APM	aerosol particle mass (analyzer)
CA	crank angle
CCD	charge-coupled device
CLiiME	community LII modeling environment
CMD	count median diameter
CPC	condensation particle counter
CPMA	centrifugal particle mass analyzer
CRDS	cavity ring-down spectroscopy
CVS	constant volume sampling
CW	continuous wave
DMA	differential mobility analyzer
EC	elemental carbon
EGR	exhaust-gas recirculation
ELS	elastic light scattering

HAB	height above the burner
HDDV	heavy-duty Diesel vehicle
IR	infrared
LIDELS	laser-induced desorption with elastic light scattering
LIF	laser-induced fluorescence
LII	laser-induced incandescence
LIISim	LII simulation software
MD	molecular dynamics
NEXAFS	near-edge x-ray absorption fine-structure spectroscopy
nvPM	nonvolatile particulate matter
PAH	polycyclic aromatic hydrocarbon
PLIF	planar laser-induced fluorescence
PMS	particle-mass spectrometer
PMT	photomultiplier tube
RAYLIX	Rayleigh scattering, laser-induced incandescence, and extinction
SAXS	small-angle x-ray scattering
SEM	scanning electron microscopy
SMPS	scanning mobility particle sizer
SP2	single-particle soot photometer

TDC	top dead center
TEM	transmission electron microscopy
TiRe-LII	time-resolved LII
UV	ultraviolet
WAXS	wide-angle x-ray scattering
XPS	x-ray photoelectron spectroscopy

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Figure Captions

Figure 1: Experimental setup for time-resolved pulsed-LII-signal collection: Soot particles, generated in a flame, are irradiated with the output from an Nd:YAG laser at either 1064 or 532 nm. An aperture close to the laser selects the center of the beam. This aperture is relay-imaged to the detection region in the flame to generate a collimated beam with a top-hat spatial profile. Typical top-hat beam profiles are inset in the upper left-hand corner. The beam is monitored with a beam profiler and attenuated using a half-wave plate and thin-film polarizers; the graphs on each side of the profiles show the intensity profile through the center of the respective beams. The resulting incandescence is imaged onto a combination of fast photomultiplier tubes (PMTs) equipped with different bandpass filters. Notch filters (532 or 1064-nm, depending on laser wavelength) are also placed in front of the PMTs to block laser scatter.

Figure 2: Temporal profiles of the soot temperature (symbols) and pulsed-LII signal (solid lines) recorded for a laser wavelength of 1064 nm. The dotted lines represent the laser temporal profile. The particle temperatures were inferred from emission spectra, and the LII signal was recorded using a 681.8-nm bandpass filter. Measurements were made on mature soot in an atmospheric co-flow diffusion flame. Data are shown for fluences of (a) 0.1 J/cm^2 and (b) 1.45 J/cm^2 . The delay between the rise in temperature and LII signal is related to the detected spectral range and the strong nonlinearity of the signal as a function of the temperature. Adapted from Goulay *et al.* [117].

Figure 3: Pulsed-LII temporal profiles recorded at selected laser fluences. Measurements were made in an atmospheric flame using a 532-nm laser to heat the particles. The dashed curve represents the laser temporal profile, and the solid curves show LII signal using laser fluences of (1) 0.072 J/cm², (2) 0.14 J/cm², (3) 0.27 J/cm², and 0.72 J/cm². Reprinted from Ni *et al.* [116].

Figure 4: Fluence dependence of the peak-pulsed-LII signal and peak-particle temperature. (a) The maximum of the pulsed-LII temporal profile is shown as a function of fluence for laser wavelengths of 532 and 1064 nm. (b) The corresponding peak-particle temperatures are shown as a function of fluence. Data were recorded in a co-flow diffusion flame with a top-hat laser-beam profile. Adapted from Michelsen *et al.* [119].

Figure 5: CW-LII and ELS temporal profiles are shown for a single glassy-carbon particle injected into the cavity of a CW 1064-nm beam. The particle diameter is 600 nm. The laser-particle interaction profile is displayed for reference and was estimated by measuring scattering signal from a non-absorbing polystyrene sphere [131]. The timing between the laser profile and the LII signal is not known. Adapted from Moteki *et al.* [131, 518].

Figure 6: Experimental setup for CW-LII-signal collection: Soot particles are delivered to the Single-Particle Soot Photometer (SP2) at atmospheric pressure. A nozzle, positioned perpendicularly to the laser beam and detection axis, directs the aerosol to the center of the laser cavity where the beam waist is ~1.1 mm. The particles drift through the laser beam and are detected one at a time by two PMTs equipped with different broadband filters for

two-color LII and an avalanche photodiode for elastic scatter. Adapted from Schwarz *et al.* [125] with kind permission from John Wiley & Sons.

Figure 7: Illustration of processes influencing the temperature and mass of a particle during LII-signal collection: Particles are heated by absorption of laser radiation, and, to a lesser extent, by oxidation and annealing. Under most conditions, particles cool by conduction to the surrounding gas and sublimation, and, to a lesser extent, by thermionic emission and radiative emission. Particles lose mass by sublimation and oxidation. Signal is derived from radiative emission from the particle.

Figure 8: Ratio of heat-transfer rates using the approaches of Fuchs [91] and McCoy and Cha [207] for atmospheric pressure, an ambient gas temperature of 300 K, and a thermal-accommodation coefficient of 0.25. Results are shown as a function of primary-particle size and particle temperature. Reprinted from Kuhlmann *et al.* [167].

Figure 9: Example pulsed-LII energy-transfer rates. Model predictions of the temporal evolution of heating and cooling rates are shown for (a) a low-laser fluence and (b) a high-laser fluence. Calculations were performed for an ambient temperature of 1676 K and pressure of 1 bar, a primary-particle size of 33 nm, and a laser wavelength of 532 nm. Adapted from Lemaire and Mobtil [186].

Figure 10: Heat flux of particle cooling through conduction, evaporation, and radiation calculated for 25-nm soot particles in nitrogen at 300 K. The initial particle peak temperature depends on the pulsed-LII-laser fluence (see Fig. 4b). For particle peak temperatures below 4000 K in the low-fluence regime, cooling is dominated by conductive

cooling; above 4000 K, evaporation / sublimation plays an important role. Reprinted from Kock *et al.* [171].

Figure 11: Absorptive-heating and conductive- and evaporative-cooling rates for CW LII. The lines represent temporal profiles of heating and cooling rates for primary particles in an aggregate with varying exposed surface area, averaged over all primary particles in the aggregate. Calculations are shown for fresh mature particles with a fractal dimension of ~ 1.9 , mobility diameter of 126 nm, mass of 0.415 fg, and monodispersed primary-particle size of 13 nm. Adapted from Bambha and Michelsen [130].

Figure 12: Surface-temperature-dependent partial and combined accommodation coefficients for translational, rotational, and vibrational degrees of freedom of NO scattered from a graphite surface. Open symbols represent values derived from surface-scattering measurements, extrapolated above ~ 700 K [see [242] and references therein]. The solid circle is from Leroy *et al.* [237] for N_2 for which translational and rotational degrees of freedom were likely equilibrated. The solid line is a combination of the partial accommodation coefficients. Adapted from Michelsen [242].

Figure 13: Surface-temperature-dependent simulated partial and combined accommodation coefficients. Results for N_2 are shown for rotational accommodation α_r and for two degrees of freedom for translational accommodation, i.e., normal to the surface α_n and tangential to the surface α_t . The combined value shown as the black line neglects vibrational accommodation. Adapted from Daun [244].

Figure 14: Profile of the ratio of $E(m, 532 \text{ nm})/E(m, 1064 \text{ nm})$ (hexagons: temporal approach; squares: energetic approach) together with the soot-volume-fraction profile (diamonds) obtained from experiments in a low-pressure premixed flame. Reprinted from Clon *et al.* [261] with kind permission of Springer Science and Business Media.

Figure 15: Dependence of peak pulsed-LII signal and signal-decay rates on particle size and maturity. Results are shown for different heights above the burner in a premixed ethylene/air. The 1064-nm fluence dependence of the peak pulsed-LII signal is shown for flames with equivalence ratios of (a) 2.0 and (c) 1.77. Signal-decay rates are also shown for equivalence ratios of (b) 2.0 and (d) 1.77. Reprinted from Bladh *et al.* [284].

Figure 16: Fluence dependence of the absorption cross section. Concentration-weighted absorption cross sections are shown for (a) 532 nm and (b) 1064 nm. The symbols represent values derived from a combination of extinction and pulsed-LII measurements. The lines represent predicted increases in the absorption coefficient with a decrease in density and increase in particle size with increasing temperature caused by increasing fluence and laser heating. These predictions are based on values of the thermal-expansion coefficient for polycrystalline graphite (type 7087) and for expansion along the *c*-axis of single-crystal graphite. Values for integrated aggregate concentration over the path length Cl were set for best agreement with the absorption cross sections at the lowest fluences. Reprinted from Michelsen *et al.* [119].

Figure 17: Relative importance of distributions of aggregate size (N_p) and primary-particle diameter (d_p) for conduction in the transition regime (local gas temperature = 400 K). The

model is for low fluence (0.158 J/cm^2) pulsed LII at 1064 nm laser excitation. The calculations indicate that for these conditions both the polydispersity of aggregate size and of primary-particle diameter must be considered in interpreting pulsed-LII signals.

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Figure 18: Shielding factors in the free-molecular regime, for thermal accommodation coefficients of $\alpha_T = 0.37$ and $\alpha_T = 1$. The aggregates were created with $k_f = 2.3$ and $D_f = 1.8$, with the sizes $N_p = \{10, 20, 50, 100\}$, for $C_{ov} = 0, 0.125$ and 0.25 . Each point shows the mean shielding of 100 aggregates of the given type. Reprinted from Johnsson *et al.* [213] with kind permission of Springer Science and Business Media.

Figure 19: Pulsed-LII temporal profiles for aggregates with the same mean number of primary particles and different aggregate morphologies. Particles were heated to the same peak temperature with a laser fluence of 0.15 J/cm^2 at 1064 nm. Profiles were recorded for particles with an average fractal dimension of ~ 1.9 (solid line) and an average fractal dimension of ~ 2.4 (dotted line). Example TEM images for these particles are shown as insets. Adapted from Bambha *et al.* [14, 70].

Figure 20: CW-LII temporal profiles for aggregates with selected aggregate sizes and the same aggregate morphologies as shown in Fig. 20. Symbols represent average temporal profiles from 50 particles for each size/morphology. Profiles were recorded for particles with an average fractal dimension D_f of ~ 1.9 (solid symbols) and an average fractal

dimension of ~ 2.4 (open symbols) and selected aggregate-mobility sizes D_m . Adapted from Bambha and Michelsen [130].

Figure 21: CW-LII and ELS temporal profiles are shown for single-carbon particles injected into the cavity of a CW 1064-nm beam. Results are for (a) an uncoated glassy-carbon particle with a diameter of 600 nm (reproduced from Fig. 5 for reference and adapted from Moteki *et al.* [131, 518]) and (b) a colloidal-graphite particle coated with oleic acid with a core-particle diameter D_c of 110 nm and total particle diameter D_p of 500 nm. The same laser-particle interaction profile is displayed in each panel for reference and was estimated by measuring scattering signal from a non-absorbing polystyrene sphere [131]. The timing between the laser profile and the LII signal is not known. Adapted from Moteki and Kondo [131].

Figure 22: Fluence dependence of the peak pulsed-LII signal. The normalized peak of the LII temporal profile is plotted relative to laser fluence on a log scale for (a) low to moderate coating thickness and (b) heavy coating thickness, as indicated in the legend as percent coating by mass. All curves corresponding to the denuded particles are indicated by dotted lines. Adapted from Bambha *et al.* [70].

Figure 23: Sequence representing the LII images captured for increasing laser energies in a backward configuration. Pulsed-LII signals were collected using a Gaussian laser spatial profile at a laser wavelength of 1064 nm. Images demonstrate the increasing contribution of the outer regions of the laser beam with increasing pulse energy. The relative scale of the

LII signal is indicated by the grayscale bar on the right. Reprinted from Delhay *et al.* [355] with kind permission of Springer Science and Business Media.

Figure 24: Simulated pulsed-LII signal as a function for various laser-beam profiles.

Fluence curves from 18-ns gate starting 20 ns after the start of the laser pulse. Reprinted from Bladh *et al.* [173] with kind permission of Springer Science and Business Media.

Figure 25: Laser temporal profiles are shown for 300 single laser shots (from a frequency-doubled, injection-seeded Nd:YAG laser with an unstable-resonator-cavity design) with the seeder turned on (blue lines) and turned off (gray lines). Individual temporal profiles are shown with the seeder turned on (magenta line) and turned off (black line). Adapted from Goulay *et al.* [357].

Figure 26: Schematic diagram of the RAYLIX approach [162, 374]. A laser beam is split into two beams, one of which is delayed with respect to the other one. Both beams are formed into light sheets. The first, weaker sheet is used for Rayleigh scattering and extinction, which is measured through the fluorescence signals from a dye solution in cuvettes before and after the sheet passes through the flame. These signals are recorded in the outer regions of the camera capturing the scattered light. The second light sheet is employed for pulsed LII recorded by a second camera. Figure courtesy of R. Suntz.

Figure 27: Single-shot beam profiles of a laser sheet passing through (a) a turbulent air jet and (b)-(f) a turbulent non-premixed ethylene-air flame. The images were recorded at 2.2 m down-beam from the flame. Reprinted from Sun *et al.* [375] with kind permission of Springer Science and Business Media.

Figure 28: Comparison of primary-particle sizes derived from TEM and pulsed LII.

Primary-particle sizes derived from TEM images (gray bars) and fit with a lognormal distribution (black lines) are shown for selected HABs in an atmospheric ethylene-air co-flow diffusion flame. Primary-particle sizes derived from pulsed-LII temporal profiles are shown for analyses assuming a monodisperse-size distribution (green dashed lines) and accounting for an extreme of large (red dashed lines) and small (blue dashed lines) particles (see text). Reprinted from Cenker et al. [377] with kind permission of Springer Science and Business Media.

Figure 29: Size and mass distributions are compared for flame generated and size-selected soot particles. Soot was extracted from a co-flow diffusion flame and size selected at 125 nm. (a) The mobility-size distribution measured with an SMPS is shown for comparison. The modes are labeled with the mobility size at the peak. (b) The mass distribution from CW-LII signal using an SP2 instrument is compared with the mass distribution measured with a CPMA. The modes are labeled with the mass at the peak. Adapted from Bambha and Michelsen [130].

Figure 30: CW-LII and scattering temporal profiles from a particle demonstrating negative lag time. Broadband (magenta line) and narrowband (cyan line) LII-temporal profiles are shown with signal from the scattering channel (black line). The dotted yellow line represents the laser-particle interaction time. Adapted from Sedlacek *et al.* [407].

Figure 31: Sooting properties of a laminar premixed ethene/air flame measured with a combination of pulsed LII and elastic scattering. Primary particle diameters (top left) and

soot volume fractions (bottom left), radius of gyration R_g (top right) and number of primary particles per aggregate N_p (bottom right) Reprinted from Reimann *et al.* [399] with kind permission of Springer Science and Business Media.

Figure 32: Simultaneous images of PAH and soot-volume fraction. Images were taken in a pressurized ethylene-air swirl flame using UV laser excitation of PAH fluorescence and IR-initiated pulsed LII. Images are shown for (a) soot-volume fraction (corresponding color bar to the right of the panel), (b) PAH LIF intensity (corresponding color bar to the right of the panel), (c) PAH LIF intensity with soot interference removed (corresponding color bar to the left of the panel), and (d) combined soot-volume fraction and PAH intensity corresponding to the color bar on the right. Reprinted from Geigle *et al.* [519] with kind permission of Springer Science and Business Media.

Figure 33: Simultaneous detection of OH LIF (green) and soot pulsed LII (red) in a Diesel spray [436]. The numbers in the center refer to the crank-angle timing. Figure courtesy of J Dec.

Figure 34: Soot particle size (count median diameter, CMD) and width of the particle-size distribution σ_g from in-cylinder TiRe-LII measurements in a Diesel engine [171].

Figure 35: (a) Average diurnal traffic flow (vehicles min^{-1}) and fraction of the flow that was classified as heavy-duty Diesel (HDDV) during the 17 days of the study [199]. The HDDV fraction was based on vehicle counts. Yellow vertical bars denote the times when perpendicular transects were driven, which corresponded to the data used for emission-factor calculations. Error bars denote the standard deviation of the mean. (b) Ratio of

measured soot concentration (SP2: HS-LII) as a function of hour of day for three consecutive days during the study. Only data for distances less than 100 m from the highway during transects were used. Figure courtesy of J. Liggio.

Figure 36: Counting efficiency of SP2 instruments measuring aspirated fullerene agglomerates. Reprinted from Laborde *et al.* [376, 483].

Figure 37: Schematic diagram of the set-up for pulsed LII measurements in a carbon-black production reactor and trace of particle sizes measured upon variation of reactor parameters [484].

Table 1. Contribution of the energy-loss mechanisms.

	Pulsed LII (ns timescales)		CW LII (μ s timescales)	
	Main effects	Fluence range	Main effects	Time regime
<i>Heating mechanisms</i>				
Laser absorption	Peak LII, risetime	All fluences	LII timing, peak	All times
Oxidation	LII decay rate	Low fluences ¹	LII timing	Early ²
Annealing	Optical properties	All fluences	Optical properties	All times
<i>Cooling mechanisms</i>				
Conduction to air	LII decay rate	Low fluences ¹	LII timing, peak	Early ²
Sublimation	Peak LII, decay rate	High fluences ¹	Peak, decay rate	Variable ³
Radiation	Minor, unless vacuum	All fluences	Minor	All times
Thermionic emission	Minor	All fluences	Minor	All times

¹For mature soot at flame temperatures and atmospheric pressure, the low fluence range includes fluences $\leq \sim 0.15$ J/cm² at 1064 nm and $\leq \sim 0.08$ J/cm² at 532 nm, and the high fluence range includes fluences above these values [176, 186, 214, 218, 287]. These fluence ranges depend on ambient pressure and temperature and particle composition.

²Conductive cooling and oxidation are less important during sublimation or coating vaporization [130].

³The timing for sublimation depends on particle morphology, size, and composition and laser irradiance. Sublimation becomes important after the particle reaches the sublimation temperature, which often occurs several microseconds after the particle enters the laser beam but before the particle reaches the center of the beam [130].

Table 2. Interferences detected when performing LII of soot and the detection wavelength ranges devoid of these interferences [117, 337].

	532 nm	1064 nm
	PAH-LIF	
Fluence	0–3.5 J/cm ²	
Delay times ^a	< 20 ns	Interference not observed
Interference-free wavelength range	680–820 nm	
	C ₂ emission	
Fluence	>0.25 J/cm ²	>0.5 J/cm ²
Delay times ^a	<40 ns	
Interference-free wavelength range	400–456, 490–500, and 580–820 nm	
	C ₂ and C ₃ emission	
Fluence	>0.35 J/cm ²	>1.0 J/cm ²
Delay times	<40 ns	
Interference-free wavelength range	580–820 nm	

^aRelative to the maximum laser pulse intensity

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

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Figure 16

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Figure 37