Theoretical estimates of spherical and chromatic aberration in photoemission electron microscopy

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Abstract

We present theoretical estimates of the mean coefficients of spherical and chromatic aberration for low energy photoemission electron microscopy (PEEM). Using simple analytic models, we find that the aberration coefficients depend primarily on the difference between the photon energy and the photoemission threshold, as expected. However, the shape of the photoelectron spectral distribution impacts the coefficients by up to 30%. These estimates should allow more precise correction of aberration in PEEM in experimental situations where the aberration coefficients and precise electron energy distribution cannot be readily measured.

Keywords: photoemission electron microscopy, PEEM, electron optics, spherical aberration, chromatic aberration, aberration correction

Photoemission electron microscopy (PEEM) is an increasingly effective method for imaging surfaces and surface near-field processes in the ultraviolet, visible, and infrared spectral regions [1–3]. High peak intensity pulsed lasers allow practical imaging of nonlinear two- and three-photon photoelectron excitations on femtosecond time scales [4–7]. Moreover, with monochromatic and coherent light sources, PEEM permits the observation of optical diffraction and interference with high spatial resolution, and this in turn has opened the way for the quantitative study of photonic and plasmonic processes in the visible and

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infrared [8–14]. These developments make PEEM a powerful tool for the study of near-field optics, offering both excellent spatial resolution and ultrafast time resolution.

Spherical and chromatic aberration strongly limit spatial resolution in PEEM. Simultaneous correction of spherical and chromatic aberration is possible through the use of electrostatic mirrors [15–19]. However, in PEEM there is currently no direct method to fully characterize the aberration coefficients, which makes the design and operation of aberration correcting optics subject to uncertainty. This situation is qualitatively different from other electron microscopies, where the electron beam aberration can be quantitatively determined for a given sample to allow subsequent correction [18, 19]. In PEEM the aberration coefficients depend sensitively on the energy distribution of the photoelectrons, which can have significant width and structure that depends on the difference between emission threshold energy and photon energy. Here we present a method for the estimation of spherical and chromatic aberration coefficients in PEEM from the accelerating potential, cathode-to-anode distance, and emission energy distribution of the photoelectrons. Furthermore, we investigate the uncertainty in these estimates when the precise energy distribution shape is unknown using simple geometric models motivated by realistic spectra [20–31].

Spherical and chromatic aberrations in PEEM primarily originate in the accelerating field and objective lens [32–36]. Here we consider the aberrations of the accelerating field only. The aberration coefficients of subsequent lenses depend on the lay-out of the instrument and are comparatively insensitive to the emission energy maximum and distribution. However, a complete accounting of image aberration requires the magnifications and aberrations of the objective and other lenses. In particular, the spherical aberration of the objective lens can be as large as that of the accelerating field. For the purpose of this paper, the lens properties can be separately measured or computed, and then they can be added to the accelerating field aberrations following the method of Rempfer [33, 35, 37–39]. This method includes the effects of the accelerating field anode aperture.

We focus on the emission energy region near threshold, which is relevant to ultraviolet and multi-photon PEEM. The virtual specimen position of the accelerating field can be computed from simple kinematic arguments [33, 35]. Retaining only the leading order contribution in V_e/V_a , the virtual specimen is located a distance

$$z_a = 2\ell_a \left(1 - \sqrt{V_e/V_a} \cos \alpha_e\right) \quad (1)$$

behind the anode, where ℓ_a is the distance between the sample plane (cathode) and the anode at potential V_a , α_e is the photoelectron emission angle relative to the surface normal, and eV_e is the photoelectron emission energy. The paraxial virtual specimen position is

$$z_{a,0} = 2\ell_a \left(1 - \sqrt{V_e/V_a}\right). \quad (2)$$

The virtual specimen position varies with electron trajectory angle and energy. As a result, the image possesses both spherical and chromatic aberrations.

Chromatic aberration requires careful treatment, so we begin with the definition of the longitudinal chromatic aberration of the accelerating field,

$$\Delta_e z_{a,0} = \langle z_{a,0} \rangle - z_{a,0}(V_e), \quad (3)$$

where $\langle z_{a,0} \rangle$ is the mean position of the paraxial virtual specimen. The mean position is calculated from an energy-weighted average,

$$\langle z_{a,0} \rangle = \int_0^{V_{e,\max}} z_{a,0}(V_e) \rho(V_e) dV_e, \quad (4)$$

where the electron charge e has been dropped for convenience, $\rho(V_e)$ is the emission energy distribution, and integration is over the full emission energy spectrum $(0, V_{e,\max}]$. The energy distribution is equal-yield normalized such that

$$\int_0^{V_{e,\max}} \rho(V_e) dV_e = 1. \quad (5)$$

The lowest rank coefficient of chromatic aberration can be defined by

$$z_{a,0} - \langle z_{a,0} \rangle = C_{ca} \left(\frac{\langle V_e \rangle - V_e}{\langle V_e \rangle} \right) + \mathcal{O}(\alpha, \delta V_e^2), \quad (6)$$

where $\mathcal{O}(\alpha, \delta V_e^2)$ represents higher order dependence with angle and energy.

Next we consider the meaning of $\langle V_e \rangle$ in Eq. (6) in order to derive a simple expression for C_{ca} . Typically, $e \langle V_e \rangle$ is taken to be the mean emission energy, i.e., derived from $\bar{V}_e = \int V_e \rho(V_e) dV_e$. However, this interpretation presents some difficulty since $\langle z_{a,0} \rangle \neq z_{a,0}(\bar{V}_e)$, i.e., the mean position of the virtual specimen is not the same as the position for the mean energy. As a result, the left-hand and right-hand sides of Eq. (6) are equal at slightly different energies. To remedy this discrepancy, we define $\langle V_e \rangle$ as the energy of the mean position. Explicitly, if we equate Eqs. (2) and (4), we get the expression

$$\langle V_e \rangle = V_a (1 - \langle z_{a,0} \rangle / 2\ell_a)^2. \quad (7)$$

Using this definition, we arrive at the simple and familiar result for the lowest order coefficient of chromatic aberration [32],

$$\langle C_{ca} \rangle = -\ell_a \sqrt{V_a / \langle V_e \rangle}. \quad (8)$$

Calculations made with this result agree with those using a direct series expansion of Eq. (6) if and only if $\langle V_e \rangle$ is defined by Eq. (7). A similar approach also yields the mean coefficient of spherical aberration [32, 36],

$$\langle C_{sa} \rangle = \ell_a \sqrt{V_a / \langle V_e \rangle} = -\langle C_{ca} \rangle. \quad (9)$$

Since the expressions for virtual specimen position and the aberration coefficients vary with emission energy, the mean values $\langle z_{a,0} \rangle$, $\langle C_{sa} \rangle$, and $\langle C_{ca} \rangle$ in general depend on the distribution of emission energies. Typically, the distribution is assumed to be a simple step or truncated Gaussian function in electron optics. In this case, the maximum energy of the distribution $eV_{e,\max} \approx nh\nu - \phi$ largely determines the values of $\langle C_{sa} \rangle$ and $\langle C_{ca} \rangle$, where ϕ is the photoemission threshold energy, and $nh\nu$ is the energy of the n photons required to liberate an electron. The distribution of real electron emission energies is often complicated, especially for multi-photon emission. Spectra can exhibit one or more asymmetrically shaped peaks of varying widths depending on the photon energy, material and surface conditions [20–31]. Indeed, these deviations are usually considered

Figure 1: Simple equal-yield distributions to investigate the effects of asymmetry and width on the coefficients of spherical and chromatic aberration. The triangle distribution ρ_{Δ} (solid lines) varies the relative position ξ of the maximum. The symmetric trapezoid distribution $\rho_{\square}$ (dashed lines) varies the relative width δ of the central region. The two distributions are equal when $\xi = 0.5$ and $\delta = 0$ (solid black line).

desirable features that reveal the internal electronic structure of materials. In these cases, the shape of the distribution $\rho(V_e)$ is also important since C_{sa} and C_{ca} are nonlinear with factors of $\langle V_e \rangle$ in the denominator.

For our purposes, we make the broad simplification that the overall distribution can be described as an irregular triangle or trapezoid as shown in Fig. 1. First, the triangular model is used to investigate the effects of an asymmetric spectrum as a function of $\xi = V_{e,0}/V_{e,\max} \in (0,1)$, where $V_{e,0}$ is the energy at the maximum of the distribution. Similarly, the symmetric trapezoid model is used to investigate the effects of distribution width, where $\delta \in (0,1)$ is the relative width of the trapezoid top at maximum emission. Thus, the triangular distribution ρ_{Δ} with $\xi = 0.5$ is the same as the trapezoid distribution $\rho_{\square}$ with $\delta = 0$. The simplicity of these models offers analytic expressions for the coefficients of spherical and chromatic aberration of the accelerating field. In particular, the triangular distribution yields compact expressions for the mean

Figure 2: Mean coefficients of spherical aberration $\langle C_{sa} \rangle$ for triangle and symmetric trapezoid distributions as a function of the maximum emission energy on a log-log scale. The lines show contours in triangle asymmetry $\xi = 0.15$ (red), 0.5 (black), and 0.85 (blue). Variation in trapezoid width δ between 0 and 1 yields changes of $< 3\%$, a region covered by the width of the central black line.

coefficient of spherical aberration

$$\langle C_{sa} \rangle_{\triangle} = \frac{15}{8} \ell_a \sqrt{\frac{V_a}{V_{e,\max}}} \left(\frac{1 - \xi}{1 - \xi^{3/2}} \right), \quad (10)$$

Explicit definitions of the distribution functions and derivations of these expressions are given in the appendix.

Figure 2 shows the mean spherical aberration coefficient of these photoelectron spectral distributions. Chromatic aberration has the same value with opposite sign. As expected, the coefficient primarily scales inversely with the square root of the maximum emission energy. Variations in asymmetry result in smaller changes. Here, a triangle distribution with $\xi = 0.15$ increases the mean coefficient by 17% compared to a symmetric distribution, while $\xi = 0.85$ decreases the coefficient by 10%. At the extreme asymmetric distributions, $\xi = 0, 1$ leads to $\langle C_{sa} \rangle$ changes of +29%, -14%, respectively. These results are summarized in Table 1. Changes in distribution width have significantly less impact, with a maximum change of -1% to +3%. Therefore, for distribu-

Spectrum asymmetry ξ	0.0	0.15	0.5	0.85	1.0
Aberration change ΔC_{sa}	+29%	+17%	0	-10%	-14%

Table 1: Impact of spectrum asymmetry on the cathode coefficient of spherical aberration.

Figure 3: The effect of an aperture stop on the transmitted electrons. (a) The gray region shows the emission angle vs. energy distribution of transmitted electrons on a log-log scale. (b) Similarly, the aperture stop skews the equal-yield triangle distributions to smaller energies. $\xi = 0.15$ (red), 0.5 (black), and 0.85 (blue) are shown here.

tions of photoelectrons that are nearly symmetric, $\langle C_{sa} \rangle$ can be estimated to within about 10% from Fig. 2, and $\langle C_{ca} \rangle = -\langle C_{sa} \rangle$. For distributions that are suspected to be more asymmetric, it may be advantageous to measure the photoelectron emission spectrum to determine the mean aberration coefficients.

We also briefly consider the effects of an aperture stop that limits the angular width of the electron beam. While the primary effect is to significantly curtail the effects of spherical aberration, the emission spectrum is also impacted, decreasing the overall intensity and skewing the distribution to lower energies which have larger aberration coefficients, as shown in Fig. 3. However, our analysis of an aperture stop in the back focal plane of an objective lens shows that the associated changes in aberration coefficients are minor, on the order of 10%, especially small compared to the uncertainties of electron energy distribution and other imaging parameters.

As the calculations of $\langle C_{sa} \rangle$ and $\langle C_{ca} \rangle$ are straightforward, they can be carried out during the microscopy work. The main input parameters are the accelerating field potential V_a and length ℓ_a , and an estimate of the photoelectron spectral energy distribution, primarily characterized by the maximum emission energy. The total aberration estimate of the final image—using the combination

method of Rempfer [34, 35, 37]—requires the objective lens settings, i.e., magnification and aberration coefficients. In our microscope, we have established a routine that uses these inputs to estimate the total spherical and chromatic aberration coefficients for each new sample. These values are then displayed in real-time on a two-dimensional graph of $\langle C_{ca} \rangle$ vs. $\langle C_{sa} \rangle$, where they can be compared to the aberration coefficients of the electrostatic mirror corrector. The user can then change the mirror settings to improve upon the initial estimates and maximize resolution.

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Appendix

Here we provide mathematical definitions of the distribution functions and some derivations of formulas presented in the main article. Derivations are limited to the triangle distribution because these expressions are fairly compact. The original formulae were derived with the aid of Mathematica 9-10. Graphs using the trapezoid and aperture stop distributions were calculated directly using Mathematica.

Distribution definitions

In this appendix, we use a normalized energy variable,

$$\nu = V_e/V_{e,\max}.$$

The triangular energy distribution is

$$\rho_{\Delta} = \left(\frac{V_{e,\max}}{2} \right)^{-1} \begin{cases} \frac{\nu}{\xi}, & 0 < \nu < \xi; \\ \frac{1-\nu}{1-\xi}, & \xi \leq \nu \leq 1. \end{cases}$$

Similarly, the trapezoid distribution is

$$\rho_{\square} = \left(\frac{V_{e,\max}(\delta+1)}{2} \right)^{-1} \begin{cases} \frac{\nu}{(1-\delta)/2}, & 0 < \nu < (1-\delta)/2; \\ 1, & (1-\delta)/2 \leq \nu < (1+\delta)/2; \\ \frac{1-\nu}{1-(1+\delta)/2}, & (1+\delta)/2 \leq \nu \leq 1. \end{cases}$$

Mean emission energy of the triangle distribution

$$\begin{aligned}
\bar{V}_e|_{\Delta} &= \int_0^{V_{e,\max}} V_e \rho_{\Delta} dV_e \\
&= \int_0^{\xi} [\nu V_{e,\max}] \left[\frac{2\nu}{\xi} \right] d\nu \\
&\quad + \int_{\xi}^1 [\nu V_{e,\max}] \left[\frac{2(1-\nu)}{1-\xi} \right] d\nu \\
&= V_{e,\max} \left\{ \frac{2}{\xi} \int_0^{\xi} \nu^2 d\nu + \frac{2}{1-\xi} \int_{\xi}^1 \nu(1-\nu) d\nu \right\} \\
&= V_{e,\max} \left\{ \frac{2\xi^2}{3} + \frac{2}{1-\xi} \left[\frac{1}{6} - \frac{\xi^2}{2} + \frac{\xi^3}{3} \right] \right\} \\
&= V_{e,\max} \left\{ \frac{1+\xi}{3} \right\}
\end{aligned}$$

Mean paraxial virtual specimen position of the triangle distribution

$$\begin{aligned}
\langle z_{0,a} \rangle_{\Delta} &= \int_0^{V_{e,\max}} z_{0,a} \rho_{\Delta} dV_e \\
&= \int_0^{\xi} \left[2\ell_a \left(1 - \sqrt{\frac{\nu V_{e,\max}}{V_a}} \right) \right] \left[\frac{2\nu}{\xi} \right] d\nu \\
&\quad + \int_{\xi}^1 \left[2\ell_a \left(1 - \sqrt{\frac{\nu V_{e,\max}}{V_a}} \right) \right] \left[\frac{2(1-\nu)}{1-\xi} \right] d\nu \\
&= 4\ell_a \left\{ \frac{1}{\xi} \int_0^{\xi} \left[\nu - \nu^{3/2} \sqrt{\frac{V_{e,\max}}{V_a}} \right] d\nu \right. \\
&\quad \left. + \frac{1}{1-\xi} \int_{\xi}^1 \left[1 - \nu - \sqrt{\frac{V_{e,\max}}{V_a}} (\sqrt{\nu} - \nu^{3/2}) \right] d\nu \right\} \\
&= \frac{4\ell_a}{1-\xi} \left\{ \frac{\xi(1-\xi)}{10} \left[-5 + 4\sqrt{\xi} \sqrt{\frac{V_{e,\max}}{V_a}} \right] \right. \\
&\quad \left. + \left[\frac{(1-\xi)^2}{2} - \frac{2}{15} \sqrt{\frac{V_{e,\max}}{V_a}} (2 - 5\xi^{3/2} + 3\xi^{5/2}) \right] \right\} \\
&= 2\ell_a \left(1 - \frac{8}{15} \frac{1-\xi^{3/2}}{1-\xi} \sqrt{\frac{V_{e,\max}}{V_a}} \right).
\end{aligned}$$

Emission energy of the mean paraxial virtual specimen position of the triangle distribution

$$\begin{aligned}
\langle V_e \rangle_{\Delta} &= V_a \left(\frac{\langle z_{a,0} \rangle_{\Delta}}{2\ell_a} - 1 \right)^2 \\
&= V_a \left(\frac{8}{15} \frac{1 - \xi^{3/2}}{1 - \xi} \sqrt{\frac{V_{e,\max}}{V_a}} \right)^2 \\
&= V_{e,\max} \left(\frac{8}{15} \frac{1 - \xi^{3/2}}{1 - \xi} \right)^2.
\end{aligned}$$

Aberration of the triangle distribution

$$\begin{aligned}
\langle C_{sa} \rangle_{\Delta} &= -\langle C_{ca} \rangle_{\Delta} = \ell_a \sqrt{V_a / \langle V_e \rangle_{\Delta}} \\
&= \frac{15}{8} \frac{1 - \xi}{1 - \xi^{3/2}} \ell_a \sqrt{\frac{V_a}{V_{e,\max}}}.
\end{aligned}$$

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