Local enhancement effect in the photoluminescence intensity of Si quantum dots: single Medusa-type particles investigated by in situ microscope spectrometer

Hironori Tamamitsu^a and Ken-ichi Saitow^{a,b,*}

^aDepartment of Chemistry, Graduate School of Science, Hiroshima University, Higashi-Hiroshima 739-8526, Japan

^bNatural Science Center for Basic Research and Development (N-BARD), Hiroshima University, Higashi-Hiroshima 739-8526, Japan

Abstract

Photoluminescence intensity enhancement of a silicon quantum dot solution was investigated using *in situ* photoluminescence/dark-field microscope spectrometer. The intensity was enhanced by Medusa-type silver particle, composed of a sphere 760 nm in diameter and many nanochains. The enhancement factor (EF) was carefully estimated, based on various corrections and finite-difference time-domain (FDTD) calculations. The EF was changed from 33 to 42,000. Spectroscopic and FDTD analyses proved valuables to evaluate local enhancement effect. Wavelength dependences of EF and scattering spectrum indicated that the enhancement is attributed to the electric field localized on particle, given by not photoluminescence light but excitation light.

* Author to whom correspondence should be addressed.

Electronic mail: saitow@hiroshima-u.ac.jp

postal address: Hiroshima University, Higashi-Hiroshima 739-8526, Japan

phone numbers: +81-82-424-7487

1. Introduction

A semiconductor quantum dot (QD) has very specific properties such as a tunable band gap and luminescence wavelength,¹⁻³ a higher optical transition rate than bulk silicon,^{4,5} and efficient multiple exciton generation.⁶⁻⁸ These optical properties allow QDs to be utilized as materials in light-emitting-diodes (LEDs),⁹ lasers,¹⁰ biomarkers,¹¹ and photovoltaic cells.^{8,12} Silicon quantum dots (Si-QDs) are a crucial material in the development of silicon photonics, but increased luminescence intensity is required by many researchers and from an industrial perspective.

Surface plasmon resonance is a powerful method to increase the light intensity for Raman scattering and fluorescence intensities. The enhancement of photoluminescence (PL) intensity in Si nanomaterials has been reported by several research groups.¹³⁻²⁴ Biteen *et al.* first reported local-field-enhanced light emission from a silicon nanomaterial using a noble metal nanostructure, and they quantified the PL-intensity enhancement factor (EF) to be 40.¹⁸ Nychyporuk *et al.* reported the EF of Si-QDs to be 60 using a monolayer of silver (Ag) nano-islands; this EF of 60 is the largest EF obtained to date for Si-QDs.²⁴ According to our survey on the PL-intensity enhancement of Si nanomaterials, the average EFs from all publications is 7.¹³⁻²⁴ However, several important issues have not yet been investigated; most reported EFs have not been the EF in the vicinity of the enhancement material, but rather a bulk EF, involving PL signals from many QDs in non-enhanced regions. In addition, almost all of the EF effects of Si-QDs were studied at a single wavelength and the wavelength dependence of the EFs has not been reported. Thus, the local EF and the wavelength dependence are critical issues to achieve a clear understanding of PL enhancement in QDs.

We have fabricated photofunctional nanomaterials by pulsed laser ablation (PLA). Gold (Au) nanomaterials (nanonecklaces, fractal networks, microspheres)^{25,26}, RGB

light-emitting Si nanocrystals²⁷ and white-light-continuum emitting Si nanocrystals²⁸ have been synthesized. Here, we show the PL intensity enhancement of Si-QDs using *in situ* PL/dark-field microscope spectrometer and estimates the local enhancement factor. In addition, the PL signals from many non-enhanced QDs in solution are carefully treated by considering the various background signals and detection volume. The enhancement particle is specially prepared, such as a Ag or Au Medusa-type particle, as shown in Fig. 1, by synthesis using PLA in a supercritical fluid: the Medusa is one of the Gorgon sisters in Greek mythology. The Medusa-type particle consists of a sphere with a diameter of 700-800 nm and many nanochains. Both *in situ* PL/dark-field microscope spectrometer and finite-difference time-domain (FDTD) calculation provide a means to investigate the local enhancement effect of the Si-QD in the vicinity of a single Medusa-type particle. It was indicated that the EF is modified from 33 to 42,000 by changing the local enhancement volume. The mechanism of PL intensity enhancement was investigated by measuring the wavelength dependences of excitation and luminescence wavelengths and the scattering spectrum, under the condition of a single Medusa-type particle. As a result, the origin of enhancement was attributed to the electric field localized on the Medusa-type particle, produced principally by not the electric field of luminescence light but that of the excitation light.

2. Experimental

A solution of Si-QDs was synthesized by the PLA of a Si wafer immersed in 1-octyne solvent.²⁹ The light source for PLA was a Q-switched frequency-doubled Nd:YAG laser (532 nm) with an energy of 19 mJ/pulse, a repetition rate of 20 Hz, fluence of 0.8 J/cm², and an irradiation time of 60 min. After PLA, the Si-QD solution was passed through a membrane filter with a pore size of 100 nm. The average diameter of the Si-QDs was estimated to be 2.4±0.4 nm

from particle analysis with transmission electron microscopy (TEM) measurements²⁹. The Ag or Au Medusa-type particles were generated by PLA in supercritical CO₂, as described elsewhere^{25,26}. Briefly, a Ag or Au plate immersed in supercritical CO₂ was irradiated with the laser for 5 min at an isothermal condition of $T = 310$ K. The Nd:YAG laser was operated with an excitation wavelength of 532 nm, energy of 38 mJ/pulse in the case of Ag, or at 19 mJ/pulse in the case of Au, with a repetition rate of 20 Hz.

The PL spectra of the Si-QD solutions were measured by *in situ* PL/dark-field microscope spectrometer (Horiba Jobin Yvon, HR800) at four excitation wavelengths; 457.9, 488.0 and 514.5 nm using an Ar⁺ laser and at 632.8 nm with a He-Ne laser. The laser power in all cases was set to 2.3 μ W in front of the optical cell. An optical cell was developed for the *in situ* measurements of the enhancement in PL intensity due to a Medusa-type particle in solution. Figure 2(a) shows a schematic illustration of the optical configuration of the PL intensity measurement using the cell and PL/dark-field microscope spectrometer. The cell was designed to give a solution layer with a thickness of 190 μ m between the cover glass and glass slide. The cell body was made of stainless steel. The Medusa-type particles were attached to a glass slide (S9901, Matsunami Glass Ind. Ltd.) that was coated with an amino-functional group and immersed in the Si-QDs solution.

An objective lens (SLMPLN100x; Olympus) with a super long working distance (7.6 mm) was used to measure the PL spectra of the Si-QDs solution under the cover glass. To analyze the EFs due to a single Medusa-type particle, four spectra were measured with different configurations, as illustrated in Fig. 2(b): PL(i) sample PL spectrum measured using the Si-QDs solution and the Medusa-type particle, PL(ii) reference PL spectrum measured using the Si-QDs solution without the Medusa-type particle, BG(iii) background spectrum of (i) measured using 1-octyne solvent and the Medusa-type particle, and BG(iv) background spectrum of (ii)

measured using 1-octyne solvent without the Medusa-type particle. All obtained spectra were calibrated using the instrument function of the PL microscope spectrometer.

The scattering spectrum of a Medusa-type particle was measured in the dark field using the same microscope spectrometer, but with a slightly different optical configuration. The objective lens of the microscope in this case was equipped with a ring-shaped halogen lamp as the light source for the scattering measurement under dark field conditions. After the scattering spectrum was measured with this microscope, a background spectrum, i.e., the scattering spectrum in the absence of a Medusa-type particle, was also measured and subtracted from the sample scattering spectrum. To compensate for the instrumentation effect on the scattering spectrum, the scattering spectrum of a commercial diffuse-reflector was measured as a standard spectrum and used to correct the spectral shape of the Medusa-type particle scattering spectrum.

3. Results and discussion

Figure 3 shows the PL spectrum of the Si-QDs at the excitation wavelength of 633 nm. Red and black lines represent the PL spectra of the Si-QDs with (w/) and without (w/o) the Medusa-type Ag particle, respectively. The PL intensity of the Si-QDs was significantly enhanced by the Medusa-type particle. Here, the enhancement in the PL intensity is discussed using the local EF. The local EF (EF_L) is expressed by:

$$EF_L = \frac{(I_{PL(i)} - I_{BG(iii)}) - (I_{PL(ii)} - I_{BG(iv)})}{(I_{PL(ii)} - I_{BG(iv)})}, \quad (1)$$

where $I_{PL(i)}$, $I_{PL(ii)}$, $I_{BG(iii)}$, and $I_{BG(iv)}$ are the integrated intensities of spectra (i)-(iv) measured by the aforementioned configurations, respectively. This estimation, based on eq. (1), has the following four properties. Firstly, the PL signal of non-enhanced Si-QDs is removed by subtracting the reference PL signal from the sample PL signals, as expressed in the numerator of

Eq. (1). This subtraction suppresses overestimation of the EF from the PL intensity of the non-enhanced Si-QDs in solution. Secondly, the enhancement factor is obtained using the solution at around a Medusa-type particle, as shown in Fig. 2(a), i.e. detection volume ($96 \mu\text{m}^3$). Namely, the present EF is not given by collecting PL signals from many Si-QDs in the solution, such as in a volume of 1 mL, which would correspond to a bulk EF. Thus, the obtained EF results in the local value of EF_L . Thirdly, the small detection volume also acts as giving more correct EF, because the volume size changes seriously the magnitude of EF. Namely, when the volume is large, the value of $(I_{\text{PL(i)}} - I_{\text{BG(iii)}})$ in eq. (1) is nearly equal to that of $(I_{\text{PL(ii)}} - I_{\text{BG(iv)}})$. In this situation, EF becomes small value (*vide infra*). Lastly, the background signals are removed by subtracting the background data signal at each measurement, as shown by the red and black lines of Fig. 3(a). By taking account of all the four properties, it is elucidated that the present analysis gives a critical evaluation of the EF. Using Eq. (1), the maximum EF_L was estimated to be 33 (red solid line). $\text{EF}_L = 27$ is the average value obtained from three-time measurements (red dash-point line). The average value of all reported EFs for Si-QDs¹³⁻²⁴ is $\text{EF}=7$, which makes the present result, $\text{EF}_L=33$, a 5-fold increase.

Next, we investigated the electric field localized on the Medusa-type particle by finite-difference time-domain (FDTD) method with a commercial software (FullWAVE, Synopsys Inc). Using the calculation results, we obtained another local EF to quantify the effective enhancement factor, given by the localized electric field near the surface of the Medusa-type particle. In this simulation, the Medusa-type particle is modeled by assembling a spherical particle of Ag with a diameter of 760 nm and nanochains, composed of nanospheres with a diameter of 20 nm, as illustrated in Fig. 4(a). The surrounding medium is 1-octyne solvent. The incident light is linear polarized light and its wavelength is 630 nm. Figures 4 (b)-(f) shows

the calculated results of electric fields localized on the particle, and the magnitude of electric field is indicated by the color. Figures 4(b) and (c) show the electric fields on the Medusa-type particle and a reference spherical particle without the nanochains, respectively. It is found that the nanochains produce large electric field. To focus the nanochains, we calculated the electric field on the nanochains by changing the configuration. Figures 4(d), (e), and (f) show the electric field on the different configurations of nanochains to the linear polarization of incident light, corresponding to 0, 90, and 45 degrees, respectively. It was found that the 45 degree inclined configuration gives the largest enhancement in the three configurations.

Here, we can estimate the enhancement volume V_{enh} using the results of FDTD calculations, illustrated in Fig. 4. In the current study, the V_{enh} is defined as the volume, where the magnitude of electric field on the particle is twice greater than that of incident light. To estimate V_{enh} , we used the enhancement layer with the thicknesses of 8 nm from the surface of nanochains by approximating the magnitude of electric field as an averaged value for the three configurations, as shown in Fig. 4 (d)-(f). In the case of the reference spherical particle, Fig. 4(c), the enhancement layer was estimated as 30 nm. Using these enhancement layers, the values of V_{enh} for the Medusa-type and the reference spherical particles were estimated as $0.076 \mu\text{m}^3$ and $0.059 \mu\text{m}^3$, respectively, by conducting the integral computations of the layer volumes from the surfaces of particles. Consequently, we quantified the effective local EF (EF_{EL}) in the vicinity of surface of particles as follows,

$$\text{EF}_{\text{EL}} = \frac{(I_{\text{PL(i)}} - I_{\text{BG(iii)}}) - (I_{\text{PL(ii)}} - I_{\text{BG(iv)}})}{(I_{\text{PL(ii)}} - I_{\text{BG(iv)}}) \times \frac{V_{\text{enh}}}{V}}, \quad (2)$$

where $I_{\text{PL(i)}}$, $I_{\text{PL(ii)}}$, $I_{\text{BG(iii)}}$, and $I_{\text{BG(iv)}}$ are the integrated intensities of spectra (i)-(iv) measured by the configurations shown in Figs. (2), respectively, V_{enh} is the enhancement volume quantified by FDTD method, and V is the detection volume ($96 \mu\text{m}^3$) by *in situ* PL/dark field microscope

spectrometer, as shown in Fig. 2(a). Equation (2) raises an important issue; almost all published analyses for EFs, involving Eq. (1), have been conducted using the approximation $V_{\text{enh}} = V$. However, the magnitude of V_{enh} is significantly different from that of V . Figure 3(b) shows the result analyzed from Eq. (2). The PL intensity of the Si-QDs was enhanced up to 42,000-fold (the maximum data, red solid line) and 34,000-fold (three-time average, red dash-point line). These results indicate that EF_{EL} is 1300 times greater than that of EF_{L} ; therefore, a large difference exists between the two values, although EF_{L} is obtained by the use of *in situ* microscope spectrometer under the condition of a single Medusa-type particle. In other words, this result indicates that a bulk EF results in a significantly lower estimation than a local EF (*vide supra*). For instance, the bulk EFs have been obtained using signals from a large volume, e.g. $V \cong 0.1\text{-}1\text{ mL}$, which would result in a significantly different value than that using V_{enh} . Thus, it can be concluded that the concept of local EF and enhancement volume is a critical issue for estimating EF.

Finally, we discuss the mechanism of PL intensity enhancement of Si-QDs, based on the measurements of EF_{EL} as functions of the excitation and PL emission wavelengths and the scattering spectra under the condition of a single Ag and Au Medusa-type particles. These measurements enabled us to investigate the enhancement effect from the viewpoint of electric field, because the enhancement of electric field at around the metal nanoparticle can be characterized by the increases of absorption and/or emission rates.³⁰ Figures 5 (a) and (b) show the dependence of excitation wavelength of EF_{EL} by the Medusa-type Ag and Au particles as solid squares, respectively. As described in eq. (2), the value of EF_{EL} at each excitation wavelength was obtained using the integrated intensity of PL spectrum measured at each excitation wavelength. As shown in Figs. 5 (a) and (b), EF_{EL} for the Ag Medusa-type particle is greater than that for the Au Medusa-type particle. The data for enhancement with both

Medusa-type particles show that EF_{EL} reaches a maximum at an excitation wavelength of 633 nm. To consider the spectral feature of EF_{EL} , the scattering spectra of Ag and Au Medusa-type particles were measured and the results are shown in Fig. 5(a) and (b) as solid lines. Both profiles show good agreement between the EF_{EL} spectra and the scattering spectra. The scattering intensity is proportional to the square of the electric field of a scattering medium;³¹ therefore, the scattering spectrum indicates the magnitude of the electric field from the scattering medium as a function of wavelength. The scattering intensity of the Medusa-type particle becomes largest at a wavelength of 650 nm; therefore, the electric field on the Medusa-type particle is largest at a wavelength of 650 nm. According to the excitation spectra of EF_{EL} in Figs. 5(a) and (b), the value becomes largest at 633 nm. Thus, the large EF is attributed to a large electric field on the Medusa-type particle. As for the EF_{EL} as a function of luminescence wavelength, the EF_{EL} spectra are obtained by dividing the PL spectrum with the enhancement particle by that without the particle. Figure 5(c) and (d) show the EF_{EL} spectra, measured at the excitation wavelength of 633 nm, of Ag and Au Medusa-type particles as a function of luminescence wavelength, respectively. The spectral intensities of both Ag and Au increase monotonically as the luminescence wavelength becomes longer region. Namely, the spectral shapes of Figs. 5(c) and (d) are not in agreement with those of scattering spectra, as shown in Figs. 5(a) and (b), respectively. Accordingly, it was revealed that the excitation process contributes principally the PL intensity enhancement of Si-QDs. The large EF of PL intensity of Si-QDs was attributed to the localized electric field on the Medusa-type particle, produced by excitation process.

4. Conclusions

The electric field enhancement of Ag and Au Medusa-type particles on the PL of a

Si-QD solution was investigated using *in situ* photoluminescence/dark-field microscope spectrometer. The local enhancement effect was analyzed with a background free condition under the observation of a single Medusa-type particle and by calculating the localized electric field calculated on the particles with the FDTD method. The EF was evaluated as EFs of two local regions, and it was elucidated that the concept of local EF (EF_L and EF_{EL}) is a critical issue for characterization of the enhancement effect for QDs. The local EF was measured as a function of excitation and luminescence wavelengths. The localized electric field enhancement on the Medusa-type metal particle, generated by not the luminescence but the excitation process, was responsible for the wavelength dependence of the EF magnitude.

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Figure captions

Figure 1. Gold Medusa-type particle prepared using pulsed laser ablation in supercritical CO₂. (a) Scanning electron microscope image and (b) schematic illustration of a gold Medusa-type particle.

Figure 2. (a) Schematic illustration of the optical configuration for PL measurement using PL/dark-field microscope spectrometer and the detection volume, V . (b) Schematic illustrations of four spectral measurements: PL(i) sample PL spectrum measured using the Si-QD solution and Medusa-type particle, PL(ii) reference PL spectrum measured using the Si-QD solution without Medusa-type particle, BG(iii) background (BG) spectrum of PL(i) measured using 1-octyne solvent and Medusa-type particle, and BG(iv) background (BG) spectrum of (ii) measured using 1-octyne solvent without Medusa-type particle. The circles in PL(i) and PL(ii) represent Si-QDs dispersed in the solution.

Figure 3. (a) PL spectra of the Si-QD solutions. The inset shows spectra (i)-(iv) with a log plot of the vertical axis, where (i)-(iv) correspond to the optical configurations PL(i)-BG(iv) in Fig. 2(b). (b) PL spectra of the Si-QD solutions corrected according to the enhanced volume (V_{enh}). The inset shows the data with a log plot of the vertical axis. All spectra measured at an excitation of 632.8 nm using a He-Ne laser. w/ and w/o denote with and without Medusa-type particle, respectively.

Figure 4. FDTD calculations to estimate the enhancement volume for the local electric field in the vicinity of a Medusa-type particle. Surrounding medium is 1-octyne solvent. The incident

light is linear polarized light and its wavelength is 630 nm. The magnitude of calculated electric field is shown by the color. (a) Schematic illustration of the Medusa-type particle modeled by assembling a spherical particle of Ag with a diameter of 760 nm and nanochains, composed of nanospheres with a diameter of 20 nm. The electric fields on (b) the Medusa-type particle and (c) a reference spherical particle without the nanochains. The electric fields on the different configurations of nanochains to the linear polarization of incident light, corresponding to (d) 0, (e) 90, and (f) 45 degrees, respectively.

Figure 5. Spectra of the effective local EF (solid squares) as a function of excitation wavelength and scattering spectra (solid lines) of (a) single Ag and (b) single Au Medusa-type particles. Spectra of the effective local EF as a function of luminescence wavelength of (c) single Ag and (d) single Au Medusa-type particles.

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5