Spectral and spatial tailoring of the luminescence by metallic nanoparticles

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In this review, we have presented several techniques to tune the luminescence both spectrally and spatially by coupling the emission centers to metallic nanostructures, with an emphasis on our works. Metallic nanostructures include single, couples of, and arrays of nanoparticles. Especially the periodic array of the nanoparticles not only can modify the emission spectra, but also select the emission directionality out of the structure. These techniques may find application in the fields of illumination and sensing.

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1. Introduction

Fluorescence is one of the fundamental optical processes, and we make use of it in daily life, for the screen backlights and in- and out-door illuminations to name a few. Fluorescent materials have been intensively studied and developed, and to date many phosphors with quantum efficiency close to unity are available. Aside from the development of better materials, there have been studies to modulate the emission properties by modifying the macroscopic environment around the emitter, e.g., putting the emitter inside a waveguide and a cavity. One landmark is the achievement of laser emission, where the emitter in the state of population inversion radiates into the mode selected by the cavity. Here we review on how to tune the fluorescence by putting the emitter close to the metallic nanostructure in order to couple the emission to the plasmonic modes. A metallic nanoparticle, the simplest example of the metallic nanostructure, shows very large polarizability due to localized surface plasmon resonance (LSPR), which is the coherent oscillation of the free electrons in the nanoparticle driven by an external electromagnetic field. If an optical emitter is located in the proximity of a metallic nanoparticle, it can decay by exciting LSPRs. The emitter experiences a modified local density of optical states (LDOS) due to the presence of the nanoparticle, which influences its radiative and non-radiative decay rates. This important characteristic has led to the term optical antenna or nanoantenna when referring to metallic nanoparticles in analogy to radio-wave antennas. Several types of optical antennas have been reported, e.g., single nanoparticles, nanorods, dimer and bowtie antennas, Yagi-Uda antennas, and arrays of nanoparticles.

This review is organized as follows. In Section 2, extinction properties of a single metallic nanoparticle are examined and compared to those of the dielectric counterparts. In Section 3, spectral tuning of the stimulated emission from the dye molecules in close proximity to metallic nanoparticles is demonstrated as an example of the modification of fluorescence coupled to plasmonic modes. In Sections 4, extinction properties of the periodic array of metallic nanoparticles are examined, in which hybrid modes of plasmonic and optical modes are introduced. In Section 5, the emission control is demonstrated both spectrally and spatially by coupling the fluorescence to the hybrid modes in a system consisting of the periodic array of metallic nanoparticles and the layer of optical emitters.

2. Extinction properties of metallic nanoparticles

Metallic nanoparticles are very strong scatterers compared to the dielectric nanoparticles having the same dimensions, especially for the frequency around the LSPR. In Fig. 1, the extinction properties of a silver (Ag) nanoparticle are calculated in the visible, and compared with those for a nanoparticle of titanium dioxide (TiO₂), one of the oxides having the highest refractive index (n) in the visible. In the simulation, a sphere with the diameter of 50 nm is placed in a matrix having n = 1.5, and the scattering and absorption efficiencies, i.e., the ratios of scattering...
and absorption cross sections to the geometric cross section, are calculated based on Mie theory using a software Mieplot. The scattering efficiency for the Ag nanoparticle shows a peak at around the wavelength $\lambda$ of 436 nm, which is ascribed to the dipole mode of LSPR of the Ag nanoparticle. The scattering efficiency at this peak is as large as 14, meaning that the energy of the light illuminated on the area 14 times larger than the geometric cross section of the nanoparticle is concentrated on the nanoparticle. This creates a large local energy density on the surface of the nanoparticle. In contrast, the scattering cross section of the TiO$_2$ nanoparticle is less than unity for the range of calculation, because the diameter of the particle is much smaller than the wavelength of illuminated light. A comparison of the scattering efficiency between TiO$_2$ and Ag nanoparticles clarifies that an Ag nanoparticle scatters light far more efficiently.

3. Spectral control of photoluminescence by metallic nanoparticles

Several studies have clarified that the spontaneous emission can be facilitated by resonantly coupling it to the LSPRs. Experimental observation includes the increase in decay rate, as well as the modification in the spectral shape of the emission. In addition to the spontaneous emission, it is also possible to tune the stimulated emission by using plasmonic structures. We have demonstrated a very simple way of tuning the wavelength of amplification of spontaneous emission (ASE) using a system consisting of Ag nanoparticles and a dye-doped polymer layer. ASE is a process in which a spontaneously emitted photon is amplified by stimulated emission as it travels through a medium with gain. Due to the coupling of ASE to the LSPRs, the peak wavelength of the ASE is tuned by the wavelength of the LSPR.

Ag nanoparticles having well-controlled sizes were synthesized using a seed-mediated growth method described in Ref. 26. We have prepared two solutions, named as the seed and growth solutions, and mixed them to grow the seed particles to the size of interest. By adjusting the amount of seed solution to that of the growth solution, we varied the final size of the Ag nanoparticles, which results in a control of the wavelength of LSPR. The nanoparticles prepared were deposited on the silica glass by using a layer by layer method, which enables a dense and homogeneous deposition. Figure 2(a) shows the typical extinction spectra of Ag nanoparticles deposited on a silica substrate. The spectra are normalized to their maxima, and they are labeled as Samples-A, B, and C referring to the samples that contain Ag nanoparticles of different mean diameters. Among them, Sample-A consisted of the smallest nanoparticles and Sample-C the largest. The peak wavelengths of the LSPR are $\lambda = 540$ nm (Sample-A), 574 nm (Sample-B), and 655 nm (Sample-C), respectively. The inset of Fig. 2(a) shows a transmission electron microscopy (TEM) image of a typical nanoparticle prepared. The nanoparticles have a size of several tens of nanometer and a prism shape with a thickness of $\sim 5$ nm, which are comparable to the size and the shape reported previously.

The Ag nanoparticles deposited on the substrates were coated with an 80 nm thick layer of polyvinyl alcohol (PVA) containing rhodamine 6 G (R6G, 40 mM) by spin-coating. Then the samples were pumped with a second harmonic of a mode-locked Nd$^{3+}$-doped yttrium aluminum garnet laser (Nd$^3$YAG, $\lambda = 532$ nm, 2 Hz repetition rate, 25 ps pulse duration) for the emission spectra measurements. The excitation beam was focused by a lens (focal length $= 150$ mm) and incident normally to the surface of the layer. The diameter of the excitation spot was $\sim 0.5$ mm. The spot size of 0.5 mm and the repetition rate of 2 Hz were chosen to minimize the degradation of dye molecules during the experiment. The emission was collected by a fiber placed at 45° normal to the surface of the layer, and detected by a CCD coupled to a monochromator. Figure 2(b) shows the emission spectra of Sample-A. Below the threshold, the sample shows a broad emission centered at $\lambda = 565$ nm (presented by a dotted curve), which is a typical emission spectrum of R6G dye molecules. In contrast, a sharp peak appears on the broad emission above the threshold (solid curve). The inset shows the peak intensity (solid circles) and the full width at half maximum (FWHM, squares) of the emission as a function of the pump energy. At the pump

\[ \text{Fig. 2. Extinction spectra of the Ag nanoparticles (nanoparticles) deposited on silica glass substrates by using a layer-by-layer method.} \]

The mean particle size in Sample-A, B, and C is different from each other. Inset shows a TEM image for a typical Ag nanoparticle prepared. (b) Emission spectra of Sample-A below (denoted by a red dotted curve) and above (a black solid curve) the threshold. The inset shows the output intensity of the emission peak as a function of pump energy for Sample-A (solid circles) and a reference without nanoparticles (open circles). Full-width at half maximum (FWHM) of the emission spectra of Sample-A is plotted in the right ordinate (solid squares). (c) Emission spectra above the threshold for Sample-A (a black solid curve), Sample-B (red dashed), and Sample-C (blue dotted). The spectrum of a reference without nanoparticles is also shown by a gray dashed curve. (d) Relation between the wavelength of the ASE and that of the LSPR for the samples investigated.
energy of 31 μJ, a sharp peak with FWHM less than 5 nm appears and the peak intensity increases abruptly, indicating an onset of ASE. Also plotted is the peak emission intensity of a reference sample without any nanoparticles (open circles), showing a linear dependence on the pump energy. No narrow peak appears in the spectra for this reference sample. Figure 2(c) compares the emission spectra for Samples-A, B, and C above the threshold, together with a spectrum for a reference sample without any nanoparticles. While the reference shows a broad emission centered at 565 nm, a sharp peak appears in the spectra of Samples-A, B, and C. These peaks are observed at different spectral positions, although they all show the same emission spectra of dye molecules below the threshold. We prepared a series of samples containing Ag nanoparticles with different mean sizes that show the LSPR peaks ranging from 530 to 670 nm, and evaluated the emission properties. All the samples containing Ag nanoparticles exhibit ASE. Figure 2(d) summarizes the relation between the wavelength of the LSPR and that of the ASE. A systematic change in the ASE peak with respect to the LSPR wavelength has been observed.

In the present samples, Ag nanoparticles modify both excitation and emission processes. In the excitation, the pump light at λ = 532 nm excites not only the dyes but also the LSPR, which enhances the absorption of dye molecules due to the increase of local energy density, and helps the system to be the state of population inversion. In the emission process, Ag nanoparticles affect the LDOS of the dyes: The dye molecules near the Ag nanoparticle emit light preferentially at a frequency that is resonant to the LSPR. This modification is a local effect, and the dye molecules that are away from the nanoparticles are not affected. Considering the thickness of the polymer layer (~80 nm), the number of the dye molecules located in close proximity to the Ag nanoparticles is far less than the number of the molecules away from them. Consequently, we observe an unmodified spontaneous emission below the threshold. In contrast, above the threshold, ASE occurs at a wavelength that is shifted depending on the wavelength of LSPRs [Fig. 2(c)]. It is noted that the decay rate of the excited state of the dye increases when it couples to LSPRs. In such a case, photons coupled to LSPRs can be a stimulus that triggers stimulated emission in the dye-containing polymer layer that is in the state of population inversion. Therefore, above the threshold, we observe an ASE that is initiated by the photons that couple to the LSPR. Thus, the peak shift in emission spectra is correlated to the shift in the LSPR. When the core–shell nanoparticles are used instead of Ag nanoparticles, the shift in ASE wavelength is suppressed because of the absence of coupling between the LSPR and the dye molecules.

4. Plasmonic-photonic hybrid modes supported in the periodic array of metallic nanoparticles

While single metallic nanoparticles can sustain LSPRs, a periodic array of them may exhibit collective resonances resulting from the radiative coupling of LSPRs. Of particular interest are periodic arrays of metallic nanoparticles with a lattice constant comparable to the wavelength of light (Fig. 3). In these arrays, the radiative coupling can be enhanced by surface or guided modes. Two distinct mechanisms enhancing the radiative coupling of LSPRs have been reported in literature. The first one consists of the hybridization of guided modes in thin dielectric layers with LSPRs of nanoparticles in the proximity of the guiding layer, as shown in Fig. 3(a). For these hybridized modes, which we will refer to as quasi-guided modes, a wave-guiding structure is necessary. The second mechanism, shown in Fig. 3(b), relies on Rayleigh anomalies (RAs) or diffracted orders in the plane of the array, which lead to the hybridized resonances known as surface lattice resonances (SLRs). SLRs are favored when the array of nanoparticles is embedded in a homogeneous dielectric and for large nanoparticles. Both quasi-guided modes and SLRs accompany a large enhancement of electromagnetic wave not only in the nanoparticles but also between them, which is beneficial to emission enhancement of large volume of emitters.

Although both quasi-guided modes and SLRs have been extensively studied, they have never been simultaneously reported in a single sample because of the different conditions to support them: Quasi-guided modes need a waveguide that has a higher refractive index than the surrounding, while SLRs are favored when the medium surrounding the array is homogeneous. We have fabricated periodic arrays on top of a thin layer of emitting material acting as a light-emitting waveguide, and measured variable-angle light extinction and emission. The simultaneous observation of both modes has been demonstrated for the first time by reducing the mismatch in refractive index between the media surrounding the periodic array.

Our sample consisted of a Ce³⁺ doped yttrium aluminum garnet (YAG:Ce) thin layer on which an array of Ag nanorods are arranged (see Fig. 4). YAG:Ce is known as an important luminescent material currently used in white light-emitting diodes. Its broad emission in the visible centered at λ = 580 nm is useful to study its coupling to the dispersive plasmonic-photonic modes in a wide spectral range. The YAG:Ce layer was fabricated by using an epoxide-catalyzed sol–gel method. The layer was planarized by silicon nitride (Si₃N₄) by plasma-assisted chemical vapor deposition, and an array of Ag nanorods was fabricated on the layer by using substrate conformal imprint lithography. The array consisted of rectangular rods of 20 nm height with long (short) axis of 300 nm (70 nm) along the x (y) direction arranged in a lattice with constants aₓ = 500 nm and aᵧ = 200 nm, as shown in Fig. 4(b). The array was covered by a conformal layer of Si₃N₄ with a thickness of 20 nm to protect the Ag nanorods from oxidation.
5. Spectral and spatial control of the emission by periodic array of metallic nanoparticles

Figure 5(a) shows the zeroth-order extinction spectrum of the array plotted as a function of the wavelength and the angle of incidence \( \theta_{\text{inc}} \). The zeroth-order transmittance \( T \) was obtained by normalizing the transmission through the samples to that of the same samples but without the array of nanorods, and the extinction was defined as \( 1 - T \). The polarization of the incident light was set along the \( y \) axis, probing the short axis of the nanorods. The wavevector component of the incident plane wave parallel to the surface along the \( x \) axis was varied by rotating the sample to change \( \theta_{\text{inc}} \) in the \( x-z \) plane, as schematically shown in Fig. 5(a). In Fig. 5(a), several extinction bands are seen, which can be assigned to different modes. The broad and dispersionless one centered at around \( \lambda = 550 \text{ nm} \) is due to excitation of LSPR. Sharp bands of high extinction are visible across the LSPR, which are due to the excitation of quasi-guided modes in the YAG:Ce layer. The dispersion relation of the TE\(_0\) guided mode is calculated for a three layer system without an antenna array, and denoted by dotted lines.41) In this calculation, we consider a slab YAG:Ce waveguide with the thickness of 200 nm and \( n = 1.56 \), sandwiched in between an upper and lower medium of infinite thickness and a refractive index of SiO\(_2\) glass (\( n = 1.46 \)) and air (\( n = 1.0 \)). The dispersion is folded into the first Brillouin zone of the periodic array by taking into account the pitch in the \( x \) direction (\( ax = 500 \text{ nm} \)), i.e., the edge of the first Brillouin zone (\( \pi/a_x \)) is at the wavenumber = 6.28 mrad nm\(^{-1}\). SLRs appear at longer wavelengths than the quasi-guided mode. RAs of light waves diffracted by the array are plotted as solid curves, calculated assuming that the array is embedded in a homogeneous medium, with effective refractive index \( n = 1.48 \). The SLRs are located at \( \lambda = 742 \text{ nm} \) for normal incidence, with one resonance assigned to the \((-1, 0)\) RA and shifting to longer wavelengths, and the other the \((+1, 0)\) RA and shifting to shorter wavelengths with increasing \( \theta_{\text{inc}} \). The dispersion of SLRs and TE\(_0\) mode is very similar, as expected from their similarity in spatial distribution in Fig. 3.

In the present sample, simultaneous excitation of quasi-guided modes and SLRs are allowed because the conformal Si\(_3\)N\(_4\) layer reduces the refractive index mismatch surrounding the array. For comparison, we prepared the sample having the same structure as the presented one but without the Si\(_3\)N\(_4\) layer on top on the array. The extinction spectra show only the quasi-guided modes and LSPRs, and SLR is not supported because of the large index mismatch.35)

In Figs. 5(b) and 5(c), we plot the photoluminescence (PL) spectra as a function of the wavelength and the angle of the emission from the normal to the surface, \( \theta_{\text{em}} \). We collected the light polarized along the \( y \) direction. The inset shows a schematic illustration of the experiment. (c) Photoluminescence spectra for the reference YAG:Ce layer without a nanorod array.
by a fiber coupled spectrometer mounted on a rotation arm that was rotatable in the x-z plane around the excitation spot. Emission from the samples with and without an array is plotted in Figs. 5(b) and 5(c), respectively. Surprisingly enough, the spectral shape is completely modified by the array and becomes angle-dependent. The PL follows the dispersion of the extinction shown in Fig. 5(a), i.e., relative maxima in the emission intensity are achieved at the wavelengths and angles of the SLRs, the quasi-guided modes, and the LSPRs. This similarity indicates a reversed relation between the extinction and PL processes. In the extinction process, light illuminated from outside is coupled into the layer, whereas in the PL process fluorescence created inside the layer is coupled out of the sample. Especially, a large PL enhancement along the SLR and quasi-guided modes comes from a large spatial overlap between these modes and the layer of emitters, i.e., they are not localized in the nanorods, but extended over the layer of YAG:Ce.

The relation between the extinction and PL is further examined in Fig. 6, which presents the (a) 1–T, and (b) PL spectra, obtained at different angles. In Fig. 6(a), we plot extinction at the incident angles $\theta_{\text{inc}} = 0$ (solid curve), 4 (dashed), and 7° (dotted). For all the angles, one can see a broad resonance centered at $\lambda = 550$ nm, which is due to the excitation of LSPRs in the individual rods. We also see the (+1, 0) SLR at a longer wavelength with an asymmetric, Fano-like, lineshape. This resonance blueshifts as the angle increases. For the spectrum at $\theta_{\text{inc}} = 4\,^\circ$, we also see a resonance at $\lambda = 776$ nm, which corresponds to the ($-1$, 0) SLR. Figure 6(b) displays the PL spectra collected at $\theta_{\text{inc}} = 0$ (solid curve), 4 (dashed), and 7° (dotted). The spectrum of a reference YAG:Ce layer without the periodic array at $\theta_{\text{inc}} = 0$° is also plotted as a filled area, showing the typical broadband emission of YAG:Ce peaking at 575 nm. The emission from the periodic array sample is more intense compared to the reference, besides the additional PL enhancement at the resonances.

We have confirmed that this effect of periodic array is robust, i.e., the spectral and spatial emission control is not limited to a specific set of periodic array and optical emitters. We demonstrated other combination, an array of aluminum dots and a polymer film containing fluorescence dye molecules. By matching the period of the array and the wavelength of fluorescence, much larger enhancement, as large as 60, can be achieved.

6. Conclusions

In this review, we discussed the modifications of fluorescence properties of the emitters coupled to metallic nanostructures. It was emphasized that metallic nanoparticles are strong scatterers compared to dielectric counterparts around the frequency of LSPR, which give them a power to work as optical nanoantennas. As examples of modified fluorescence, our works on the wavelength-tuning of ASE coupled to Ag nanoparticles and on the wavelength and direction control of PL of YAG:Ce layers coupled to the periodic array of Ag nanoparticles were highlighted.

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