Direct observation of amplified spontaneous emission of surface plasmon polaritons at metal/dielectric interfaces

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We report on direct observation of amplified spontaneous emission (ASE) of surface plasmon polaritons (SPPs) at the interface of a silver film and a gain medium. Based on a typical Kretschmann configuration incorporated with Rhodamine 6G molecules, the growing ASE spectra of SPPs have been clearly identified by carefully conducting a pump-dependent angle-resolved spectral measurement. Spectral narrowing effects induced by the SPP amplification are also demonstrated. The observed phenomena are helpful in understanding the fundamental interactions between SPPs and gain medium, which could enable wide applications on plasmonic sources and sensors. © *2011 American Institute of Physics*. [doi:10.1063/1.3605599]

Plasmonic structures can impose strong enhancement and confinement on electromagnetic field, which find applications ranging from sensing, sub-wavelength resolution imaging, plasmonic circuitry, etc.[1] However, the intrinsic absorption of metal fundamentally limits the performances of these plasmonic systems, for example, the sensitivity of surface plasmon resonance on external perturbations.[2] Integrating the metallic structures with an optical gain medium has been proposed as a possible solution to this problem in versatile schemes,[3–7] based on which surface plasmon amplification has been successfully demonstrated.[8–15] However, the direct observation of the amplification effect of surface plasmon polaritons (SPPs) by the gain medium is still challenging and has aroused various ambiguities and controversies. The reason lies in the difficulties of direct recognition of the spontaneous emission (SE), amplified spontaneous emission (ASE), and lasing-like stimulated emission (LSE) of SPPs, as well as the photonic emission from the gain medium.

Here we report on the direct experimental evidences of ASE of SPPs (ASE_{SPP}) at the interface of a silver film and a polymethyl methacrylate (PMMA) film doped with Rhodamine 6G (Rh6G) dye molecules. By carefully conducting a pump-dependent angle-resolved spectral measurement, the SE and ASE of SPPs, as well as the emission of Rh6G have been successfully recognized. The results provide a clear physical picture on the SPPs amplified by the gain medium. It turns out that the phenomenon reported in Ref. 12 may be actually an ASE_{SPP} instead of the LSE of SPPs.

Our experimental setup is based on a typical Kretschmann configuration.[12,16,17] As shown in Fig. 1(a), a 55-nm-thick silver film is deposited on a prism (SF57, $n_{prism} = 1.846$ at wavelength 589.3 nm), on which we first deposit a 5-nm-thick SiO_2 layer and then spin-coat a PMMA film (about 1.3 μm thick, $n_{PMMA} \approx 1.49$) doped with Rh6G at a concentration of 0.015 mol/l. The thin SiO_2 layer is utilized to avoid the fluorescence quenching induced by direct touching of the molecules with the metal layer[18] and thus get a higher SPP excitation efficiency.[19] To match the absorption peak of Rh6G, a pulsed laser with wavelength $\lambda = 532$ nm and pulse duration ~5 ns is used to excite the Rh6G molecules from the PMMA side.[20] The diameter of the laser spot is about 1 mm and its polarization is normal to the x-z plane (such a polarization can minimize the scattering of the pump laser into SPPs). Since the heat dissipation in PMMA has a characteristic time of 0.5 s,[21,22] we set the repetition rate of the pulse at 0.5 Hz to minimize the photodestruction.[21] After the pump pulse strikes at the PMMA/Rh6G film, the SPP modes are excited via two ways: the fluorescence of Rh6G can be scattered into SPPs by the roughness of the surface, while the excited Rh6G molecules near the metal surface can be directly coupled into SPP modes with a high efficiency via a nonradiative decay process.[16,23] Once SPPs are excited, they propagate along the interface of silver and PMMA/Rh6G and

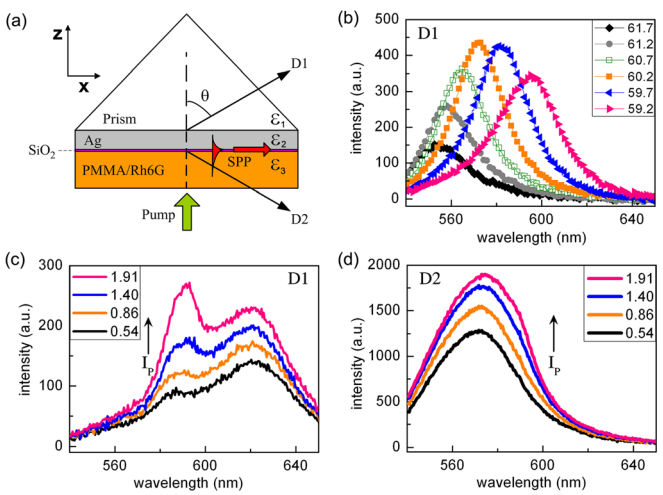

FIG. 1. (Color online) (a) Schematic diagram of the experimental setup. A ns laser pump is used to excite Rh6G molecules and SPPs. The SPPs are decoupled by the prism, and corresponding emission is measured through detection path D1. Meanwhile, the emission of Rh6G is measured from the film side through detection path D2. (b) Measured SE spectra of the SPPs decoupled at different angles θ (in degree). $I_P = 0.54$ mJ/cm^2. (c) Direct evidence of ASE_{SPP} decoupled at $\theta = 58.4°$. Spectra are measured with different I_P as noted (unit: mJ/cm^2). $\lambda_{ASE} = 592.87$ nm. (d) Measured emission spectra of Rh6G recorded at D2 under the same I_P corresponding to (c). The difference between (c) and (d) clearly demonstrates that the ASE is from SPPs other than Rh6G.

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are simultaneously decoupled into radiative waves by the prism at an angle that satisfies $\sqrt{\varepsilon_1}\sin\theta = \sqrt{\varepsilon_2\varepsilon_3/(\varepsilon_2+\varepsilon_3)}$, where ε_i ($i = 1,2,3$) is the permittivity of the ith medium as marked in Fig. 1(a). The angle-dependent decoupling of SPPs can be utilized to resolve the emission of SPPs from the emission of Rh6G. As shown in Fig. 1(a), we use two detection paths, D1 and D2, to realize this function. In D1, an optical fiber (400 μm in diameter) connected to a spectrometer (Ocean Optics, USB2000) is used to collect the SPP emission at the prism side with different detection angles (θ) and pump intensities (I_P). While in D2, the emission from Rh6G that is not coupled to SPPs is detected from the film side.

Figure 1(b) shows the SE spectra of the SPPs decoupled at different angles measured with D1 scheme. The spectra peaks vary with the decoupling angles, which can be seen as the emission of Rh6G modulated by a SPP exporting function,[12] i.e., only those SPPs with frequencies satisfying $\sqrt{\varepsilon_1}\sin\theta = \sqrt{\varepsilon_2\varepsilon_3/(\varepsilon_2+\varepsilon_3)}$ could emit into the prism at a specific angle.[12,16,17] The angular dependence of the spectra indicates the dispersion relations of SPPs.[12,16,17] However, with the increase of I_P, these emission curves change dramatically. Figure 1(c) shows the SPP emission spectra at a fixed angle $\theta = 58.4°$ under different I_P. It can be seen that a shoulder at about 592.87 nm can be recognized and grows much faster than the other part of the spectra when I_P increases. This spectral evolution is mostly like the ASE properties that are involved with gain amplification.[18,24] In our system, there are only two possible amplification mechanisms, i.e., the amplification of Rh6G emission from the PMMA/Rh6G film[18] and the amplification of SPPs propagating along the metal/dielectric interface.[12] In such a case, to identify the type of ASE in Fig. 1(c), we record the Rh6G emission spectra at the D2 path under the same I_P. As illustrated in Fig. 1(d), although the SE intensity of Rh6G grows with the increased I_P, no significant change occurs in the spectral shape. Therefore, it can be concluded that the ASEs observed in Fig. 1(c) are from the amplified SPPs. It also indicates that in our configurations the threshold of ASE_{SPP} is much lower than that of ASE of Rh6G, which agrees well with the descriptions in Ref. 12. This results from two aspects: on one hand, the thickness of PMMA/Rh6G film ($\sim$1 μm) is only $\sim$1% of the propagation length of SPPs ($\sim$100 μm) and on the other hand, the field enhancement induced by SPPs leads to reinforce the interaction between SPPs and dye molecules. Such a low threshold operation of ASE is very useful for the optimization of optical gain in plasmonic systems.

As another evidence of ASE_{SPP}, the emission spectra of SPP decoupled at different angles are graphed in Fig. 2, where I_P is fixed at 1.91 mJ/cm^2. Two peaks can be recognized in these curves: one is dependent on the detection angles while the other one is position-independent. As mentioned above, the one having angular dependence is the SE of SPPs,[12] the wavelength of which has to meet the criterion of SPP decoupling conditions. The other peak, i.e., the ASE peak, is from SPPs that are amplified by the gain from dye molecules. The position of the ASE_{SPP} peak is determined by the strength of the interaction between SPPs and dye molecules, i.e., the ASE_{SPP} peak occurs at the wavelength where the interaction is maximized. This interaction is very sensitive to various physical parameters including the gain characteristics

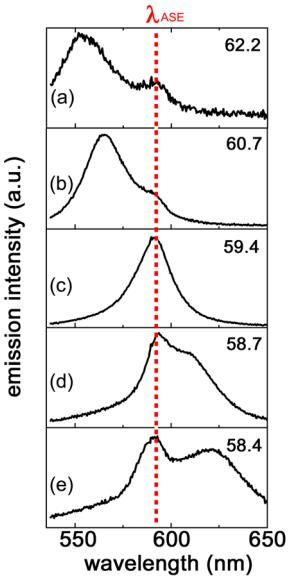

FIG. 2. (Color online) Measured emission spectra of the SPPs decoupled at different angles θ as noted. $I_P = 1.91$ mJ/cm^2. As indicated by the dashed line, the ASE wavelength (592.87 nm) is independent on the detection angles.

and realistic concentration of the dye molecules, the thickness and roughness of the metallic film, as well as the refractive index of the matrix. In our case, the ASE_{SPP} peak shows that the interaction in our sample is the strongest at wavelength 592.87 nm. Once determined by the interaction strength of SPPs and dye molecules, the ASE peak position is independent on the decoupling angle, although its peak intensity with respect to the corresponding SE background changes dramatically with the detection angle (Fig. 2). In such a case, if the ASE peak and the SE background are overlapped at other observation angles, as shown in Fig. 2(c), one is hard to directly identify the ASE spectra in experiments. That is why we can clearly observe the growth of ASE_{SPP} spectra in Fig. 1(c) at a specific angle, while the previous studies could not identify direct amplification of SPPs in spectra.[12,13]

During the increase in the pump intensity, besides the growth in ASE intensity [inset of Fig. 3(a)], we have observed clear spectral narrowing effects. As shown in Fig. 3(a), the full width at half maximum of the ASE_{SPP} spectra is decreased from 33.51 to 24.64 nm when I_P increases from 0.54 to 1.91 mJ/cm^2. This 26% in spectrum narrowing is a typical characteristic of amplified SPPs and agrees well with previous reports.[12,13] Moreover, the change in ASE intensity (ΔI_{ASE}) as a function of the increase in pump intensity (ΔI_P), as shown in Fig. 3(b), indicates a nearly linear relationship when measured at different angles θ. The two observations, again, confirm the ASE properties of the SPPs.

It should be mentioned that in previous studies people sometimes concluded their observations as LSE of SPPs based on such evidences as the spectral narrowing and pump-related threshold effects.[12] However, these two effects are not sufficient in identifying LSE because ASE generally possesses the similar spectral narrowing and pump-related threshold effects as those of the LSE.[13,24] One fundamental difference in the formation of ASE and LSE is that LSE needs feedback mechanisms while ASE does not. Therefore,

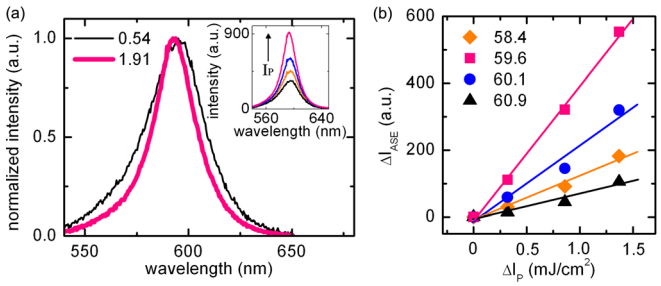

FIG. 3. (Color online) Normalized emission spectra of SPPs decoupled at $\theta = 59.2°$, where the spectra are peaked at the ASE wavelength ($\lambda_{ASE} = 592.87$ nm). Inset: measured emission spectra of SPPs under different I_P. From bottom to top: $I_P = 0.54$, 0.86, 1.40, and 1.91 mJ/cm^2. $\theta = 59.2°$. (b) Change in ASE_{SPP} intensity (ΔI_{ASE}) as a function of the increase in pump intensity (ΔI_P) measured at different angles θ as noted. The data measured at different θ show nearly linear relationship (as guided by the solid lines).

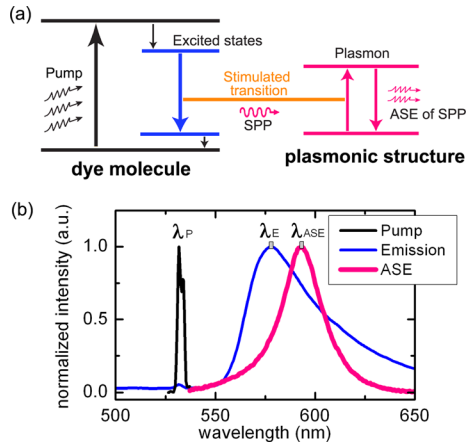

FIG. 4. (Color online) (a) Schematic diagram of the energy levels and transitions in the generation of ASE_{SPP}. (b) Measured spectra of the pump laser, the emission of Rh6G, and the ASE of SPPs.

in those systems where the feedback mechanisms are lacking or not clear, ASE other than LSE is more likely dominant in the observations.[12] In our studies, besides the spectral narrowing and pump-related effects, we have observed an emerging peak at specific wavelengths [Fig. 1(c)], which is a key feature of the ASE that was observed in various systems.[24,25] That is why we conclude our observations as ASE other than LSE. However, due to the noise at pump intensity below 0.54 mJ/cm^2 and the photodestruction of PMMA above 1.91 mJ/cm^2 during the continuous spectral measurement, we can only judge the threshold of the ASE as 0.54 mJ/cm^2 from the limited data, and no LSE of SPPs are observed even when the samples are destroyed by the laser.

With the information of above experiments, we are now clear on the understanding of the mechanism of the SPP amplification. At low pump intensities, only a small part of the molecules, which are incoherent to each other, can be excited and coupled to SPPs randomly, part of which are then decoupled by the prism out into air as the SE of SPPs. Further increase in the pump intensity could drive more molecules into the excited states and transfer their excitation energy to the plasmon excitation through resonant coupling transitions, as sketched in Fig. 4(a).[3,26] These transitions are stimulated by the plasmons already in the nanosystem, causing the buildup of a macroscopic number of SPPs.[3] Amplified SPPs are thus formed and decoupled by the prism in the form of ASE_{SPP}. Figure 4(b) shows the measured spectra of the pump laser, the emission of Rh6G in polymer film on a glass substrate, and the ASE of SPPs, respectively. The center wavelengths of the spectra match very well with our theoretical model [Fig. 4(a)].

In conclusion, our experiments have shown clear evidences of ASE_{SPP}, including the sharp growing of ASE_{SPP} peak with increasing pump intensity, the wavelength independence of the ASE_{SPP} peaks on the decoupling angles, and the SPP amplification induced spectral narrowing effects. Both the methods and results provide an important understanding in accurate description and efficient exploit of the interactions between metallic structures and gain media. Such knowledge is useful for studies on plasmonic structure designing, loss compensation, plasmonic sensing, and nanosource applications.

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