Amplified spontaneous emission of surface plasmon polaritons and limitations on the increase of their propagation length

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Amplified spontaneous emission of surface plasmon polaritons (SPPs) at the interface of a resonant gain medium has been observed. The amplification is accompanied by significant spectral narrowing and limits the gain available for compensation of SPP propagation losses. The effect is similar to the deteriorating influence of amplified spontaneous emission in laser resonators. © 2010 Optical Society of America

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Resonant interaction of surface plasmon polaritons (SPP) with molecules, ions, and semiconductor quantum dots near a metal interface attracts considerable attention, since its understanding may lead to a new nanophotonic technologies for sensing, optical data processing, and quantum information. One of the most significant applications may be the amplification of the SPP waves or lasing into SPP modes [1–7].

SPPs, being surface waves confined to a metal–dielectric interface, are subject to significant propagation loss and thus have relatively short propagation length [8]. With high demand for SPP applications in optical information processing, the question that has arisen is how to increase the propagation length. Several experimental demonstrations of various degrees of SPP amplification in different systems have been realized in the visible as well as at datacom wavelengths [3,4,6,9,10]. The search, however, still continues for the SPP amplification scheme that can be integrated into practical plasmonic circuitry [11]. In this context, the investigations and understanding of the mechanisms of SPP interaction with amplifying media is essential in order to optimize stimulated emission conditions.

Here we report on the observation of amplified spontaneous emission (ASE) of surface plasmon polaritons (ASESPPs) excited at the interface with a resonant amplifying medium. Generally, ASE occurs during light propagation in the gain medium with inverse population and is sometimes called traveling wave lasing or superfluorescence [12]. Significant spectral narrowing of the SPP emission line is observed with the ASE threshold around 5 W/cm^2. The appearance of ASESPP is accompanied by a reduction of the amplification of the signal SPPs and limits the increase of the signal SPP propagation length to about 30% before ASE becomes dominant at higher excitation powers. The observed effects are analogous to the behavior of unoptimized lasers, where ASE limits the performance by depopulating the excited states. These observations are important for the development of SPP amplifiers and coherent emitters of SPPs (SPASERs) and the understanding of the SPP interaction with quantum systems.

In these studies, silica/Au/polymer structures were used (Fig. 1). Au films (100 nm thickness) were deposited onto silica substrates using magnetron sputtering. Nine pairs of slit arrays with different separation distances (20–250 μm) were fabricated using a focused ion beam (FIB) milling machine. The parameters of the gratings (300 nm groove width, 1450 nm period for the coupling grating and 2130 nm for the outcoupling grating) were chosen to ensure efficient excitation of SPPs at the Au/polymer interface using the 633 nm wavelength of the probe excitation at an angle of incidence of 53° and to decouple the light of around 1200 nm wavelength from the decoupling grating at an angle of 64°.

The polymethyl methacrylate (PMMA) films of below 1 μm thickness were spin coated onto the Au structures. The polymer films were doped with PbS quantum dots (QDs) and used as an amplifying medium. The QDs concentration in the polymer was 5 wt.%. The quantum dots were synthesized in-house as described in [13].

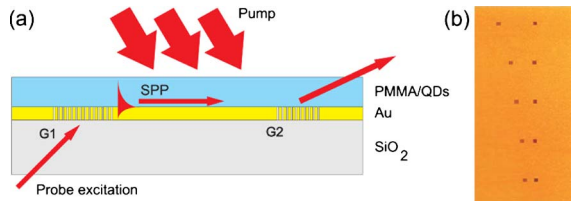

Fig. 1. (Color online) (a) Experimental setup for the optical measurements: a He–Ne laser light is coupled to SPPs on the G1 grating and excites signal SPPs via QD fluorescence. The amplifying medium is continuously pumped with a second He–Ne laser. The signal from the decoupling grating (G2) is collected into an optical fiber connected to the spectrometer. (b) Optical images of the in/out-coupling grating pairs with different distances between them.

The optical setup is illustrated in Fig. 1. The sample is mounted in a rotation stage. A 633 nm He–Ne laser (3 mW) is directed at the in-coupling grating (G1) and excites the QD photoluminescence at around 1160 nm that is coupled to the SPPs at the Au/polymer interface and is used as a weak (signal) SPP wave in the experiments. Since the SPP propagation length at the excitation frequency is below 2 μm on metal/polymer interface, the signal SPP source is localized in the vicinity of the grating. The Au thickness is large enough to prevent coupling between the SPP modes on the film interfaces. Moreover, despite the similar refractive indexes of the polymer and the substrate, the effective refractive index of the SPP mode on the interface between Au and thin polymer layer is significantly different from that of the SPP on the interface between Au and macroscopically thick substrate. The measured signal SPP propagation length, in the absence of pumping, is about 110 μm, which is between the SPP propagation length at an Au/air interface (150 μm) and Au/semi-infinite polymer (45 μm) interface at this frequency. The SPP nature of the signal detected from the out-coupling grating is confirmed by polarization and angular dependence measurements. The signal from the out-coupling grating (G2) is collected by a 20× NIR objective lens and sent to a fiber input of a spectrometer with a liquid-nitrogen-cooled InGaAs charge-coupled device (CCD). The pump He–Ne laser operating at the 633 nm wavelength (3.5 mW max power) is focused onto an area of about 100 μm diameter between the grating pairs. The power of the pump light can be controlled with a neutral density filter.

Figure 2 presents the pump intensity dependencies of the SPP spectra. When QDs in the sub-wavelength-thick polymer layer are excited with the pump beam, their emission is transferred to SPPs via near-field dipole-coupling to surface modes of the Au film [5]. The excited SPPs then propagate along the metal/polymer interface and are detected after out coupling from grating G2. (Please note that the signal SPP beam was switched off during these measurements.)

The spectral width of the SPP emission spectra becomes significantly narrower [Fig. 2(b)] with increasing pump intensity, compared to the low-power excited SPP spectra. The drop in the spectral width can be derived from the linear theory of the amplified SPPs below the stimulated emission threshold [6,7]. The gain narrowing stops at about 10 W/cm^2. This corresponds to the regime of the so-called completely narrowed ASE. The ASE threshold can be determined from the middle point between the fluorescence and the intensity corresponding to the completely gain-narrowed ASE [14]. This gives a very low threshold value for ASESPP of about 5 W/cm^2. The observed SPP intensity dependence on the pump power follows a typical trend for ASE with almost exponential growth at low pump intensities followed by a saturation tendency when the completely narrowed ASE has developed for higher pump intensities.

The coherent nature of the interaction between the signal (weak) SPP beam and gain medium is illustrated in Fig. 3, clearly showing two different regimes. At weak pump intensities, a net gain in the signal SPP mode is observed. At higher pump intensities, the gain in the signal SPP mode is virtually absent and ASESPPs dominate the signal. A strong ASESPP leads to a reduction of the population inversion of the gain medium and competition with stimulated emission into signal SPPs. The occurrence of ASESPP in all propagation directions prohibits reaching the gain threshold and transformation of

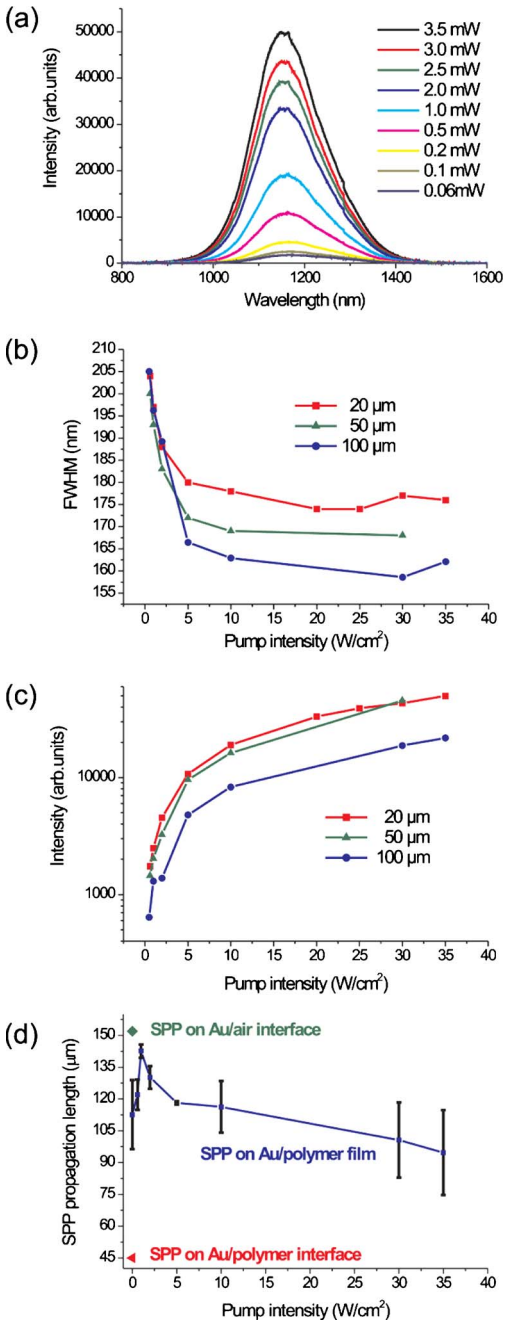

Fig. 2. (Color online) (a) SPP emission spectra for different pump intensities. (b) FWHM and (c) SPP intensity dependencies on the pump intensity for 3 pairs of in/out-coupling gratings. (d) the dependence of the propagation length of the signal SPPs on the pump intensity. The SPP propagation lengths on Au/semi-infinite polymer and Au/air interfaces are also shown.

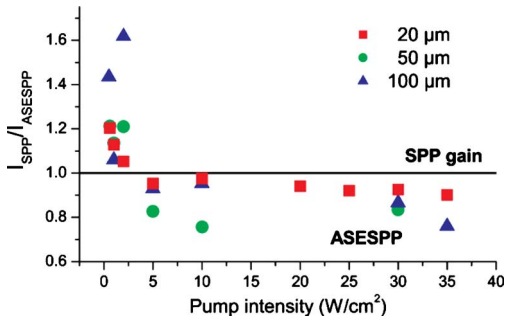

Fig. 3. (Color online) Pump intensity dependence of the normalized gain coefficient for different in/out-coupling grating pairs.

the probe SPPs into stationary lossless SPPs predicted at and above the lasing threshold [7].

Using the SPP intensity dependencies on the pump power for several sets of the in/out-coupling gratings with different distances between them, the dependence of the signal SPP propagation length on the pump intensity can be reconstructed [Fig. 2(d)]. The propagation length increases at low pump intensities reaching the maximum of approximately a 30% increase at about 1 W/cm^2. This corresponds to the intrinsic gain coefficient $g(I_p)=L_{SPP}^{-1}(0)-L_{SPP}^{-1}(I_p) \sim 17$ cm^{-1} (here, $L_{SPP}(0)$ and $L_{SPP}(I_p)$ are the SPP propagation length without pump and with pump, respectively). Similar gain values were measured for dielectric-loaded SPP waveguides doped with PbS quantum dots [10]. With a further increase in the pump intensity the propagation length of the signal SPPs becomes smaller due to competition of stimulated SPP emission and ASESPP and tends to the stationary level, where it is in the same range as the SPP propagation length in the absence of the pump. The pump intensities where the net gain is lost corresponds to the same intensities where the completely narrowed ASESPP regime is reached. Note that the theory of the amplified SPP propagation disregarding the ASESPP dynamics predicts stronger narrowing of the SPP spectra for longer propagation distances [7], the trend observed in Fig. 2(b).

In conclusion, we have observed ASE of SPPs on the interface of a resonant gain medium. The ASESPP has a low threshold intensity of about 5 W/cm^2. ASESPP limits the gain available for loss compensation in the signal SPP beam, and only about a 30% increase in the propagation length has been observed at low pump intensities before ASESPP becomes dominant. A similar effect is known in laser physics as gain depletion due to ASE [12]. The increased role of ASE may be due to the increased spontaneous emission rate in QDs near the metal film caused by the available SPP states. The Auger recombination in the quantum dots possible at high pump intensities may also affect the depopulation of the excited states and, thus, result in the decrease of the gain, but its relative contribution is unlikely to be significant for the pump intensities used in this work. Practical approaches to the enhancement of the SPP propagation length should, therefore, consider the ways to suppress unwanted amplified spontaneous emission.

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