Observation of Stimulated Emission of Surface Plasmon Polaritons

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ABSTRACT

We report a direct experimental evidence of stimulated emission of surface plasmon polaritons (SPPs) at telecom wavelengths (1532 nm) with erbium doped glass as a gain medium. We observe an increase in the propagation length of signal surface plasmons when erbium ions are excited optically using pump SPP. The design, fabrication, and characterization of SPP waveguides, thin gold metal strips, embedded in erbium (Er) doped phosphate glass is presented. Such systems can be suitable as integrated devices coupling electronic and photonic data transmissions as well as SPP amplifiers and SPP lasers.

The unique features of surface plasmons, such as enhanced and spatially confined electromagnetic fields at metal−dielectric interfaces, have been exploited in various fields ranging from nanophotonics[1,2] to metamaterials[3] and biosensing.[4−7] Despite the great promise of surface plasmons, many applications remained impractical due to high losses resulting from the damping of electromagnetic fields in metals. Active plasmonics, which describes the interaction between an active medium and surface plasmons, offers an opportunity to expand surface plasmon based applications.[1−15] Recent studies have focused extensively on surface plasmon amplification[16−23] to overcome the losses associated with surface plasmons. Several designs have been suggested to compensate the SPP losses by introducing gain in the dielectric surrounding the metal; the gain media included quantum well heterostructures,[16−18] quantum dot inclusions,[19] and dye molecules.[20] The gain medium in these configurations provides the energy to compensate for the SPP loss by the process of stimulated emission, thus increasing the SPP propagation length. Large magnitude of this effect can result in a complete compensation of loss and may even lead to SPP lasing. Very recently, lasing has been demonstrated in metallic-coated nanocavities using semiconductor heterostructures;[24] however, the lasing mode is not purely plasmonic.

A key milestone to realize a SPP laser is to establish stimulated emission of SPPs. Preliminary experiments combining active medium and SPPs have studied stimulated emission of SPPs by measuring the change in the reflectance.[23] However, the indirect nature of this observation as well as the small measured change ($\sim 10^{-5}$ in differential reflectance corresponding to gain of 0.8 cm^{-1}) offer rather limited evidence of SPP enhancement. In this letter, we present a direct evidence of stimulated emission of SPP by measuring the change in the propagation length in a SPP waveguide. We report such measurements in both continuous and pulsed modes to prove that the enhancement contribution from spontaneous emission is negligible, and it arises from stimulated emission of SPP. This experimental study offers a possibility of exploring and realizing SPP-based applications such as plasmonics-based signal processing.

To demonstrate stimulated emission of SPPs, we use plasmon waveguides, finite width thin gold strips, surrounded by erbium-doped phosphate glass and that operate at telecom wavelengths. Such a symmetric configuration supports a long-range surface plasmon mode, which has a symmetric transverse electric field across the width and thickness of the metal strip.[25,26] Using pump SPPs, active erbium ions in phosphate host glass are excited to a higher energy state. The excited ions decay by spontaneous emission process into all the available modes. A thin metal strip embedded in a dielectric material can support multiple plasmonic spatial modes;[25] most of these modes are highly confined and have short propagation length. Only a small fraction of energy goes to the long-range SPP mode by spontaneous emission. However, energy transfer can be channeled to long-range SPP mode by the stimulated emission process using signal SPP (Figure 1a). A requirement for stimulated emission, the buildup time of the stimulated emission needs to be shorter than the excited-state lifetime, is easily met due to the very long lifetime (~ 7.9 ms) of the excited erbium ions. In our

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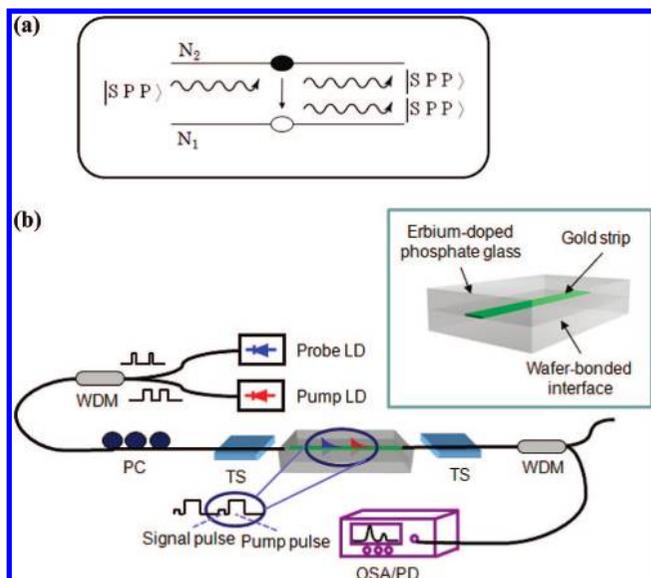
Figure 1. (a) Stimulated emission process of surface plasmon polaritons in a gain medium. (b) Experimental configuration for inducing stimulated emission of surface plasmon polaritons. A signal SPP (1532 nm) and pump SPP (1480 nm) codirectionally propagate along the metal strip embedded in Er-doped gain medium. The signal and pump beams from laser diodes (LD) are combined by a fiber wavelength-division multiplexer (WDM). For input fiber section, a polarization controller (PC) is used together with a regular single mode fiber for maintaining the state of polarization. Precision translation stages (TS) are used for aligning the input and the output fiber with the waveguides. In pulsed mode, the widths of the signal and pump pulses are 150 and 500 μs with a 1 ms period, respectively, and the time delay between the pump and signal pulses is 30 μs. Er^{3+} ions are excited to the upper energy state by the pump SPP, and stimulated emission is induced by the signal SPP. The output is monitored either by an optical spectrum analyzer (OSA) or a photodetector (PD).

work, experiments are designed such a way that both the pump and the signal are SPPs interacting with erbium ions. The long-range mode supported by metal strip SPP waveguides was shown to have attenuation as low as 1.4 dB/cm depending on the width and the thickness of the metal strips, cladding materials, and fabrication methods,[10,27] whereas a gain of 4.1 dB/cm using Er-doped glass with 5.3 $\times$ 10^{20} cm^{-3} Er^{3+} concentration was experimentally obtained.[28] This comparable gain with erbium-doped phosphate glass and SPP propagation loss of metal strip waveguides implies that lossless propagation of SPP or even lasing could be possible in configurations of SPP waveguides surrounded by Er-doped glass.

Our experimental setup, as shown in Figure 1b, is developed to accommodate the optical pumping of Er-doped glass by using long-range SPP mode guided by the same metal strip. We use a laser diode with nominal wavelength of 1480 nm as the pump and a different laser diode at 1532 nmsignal pu as the signal. The pump and signal beams are combined by a fiber multiplexer and delivered to the SPP waveguides through a regular single mode fiber with a polarization controller. The mode overlap between the signal and pump SPP is close to unity, maximizing the pump−signal interaction. To distinguish the stimulated emission from spontaneous emission effects, we study the pump−signal interaction both in continuous and pulsed mode. In the latter, the laser diodes are modulated such that there is no overlap between the pump and the signal. The signal pulse is transmitted right after the pump pulse has propagated and excited the active ions.

To fabricate SPP waveguides, gold metal strips, we use standard top-down fabrication techniques: chemical−mechanical polishing, photolithography, etching, deposition, and lift-off. Erbium-doped phosphate glass wafers (MM-2 laser glass, Kigre Inc.) with 4.2% Er_2O_3 and 1% Yb_2O_3 are polished using chemical−mechanical process with CeO_2 slurry, which results in flat surfaces with rms of surface roughness around 0.5 nm. After a thorough cleaning process, SPP waveguides of different widths are transferred into the glass wafer substrates by photolithography and dry-etching process. The resulting depth of the trenches is around 22 nm. A thin film of pure gold ($\sim$20 nm) with a flash of chromium ($\sim$2 nm) is then evaporated to fill the trenches. The lift-off process results in SPP waveguides on Er-doped glass wafer, which is diced into chips. These chips are thoroughly cleaned and bonded to blank chips of similar size of Er-doped glass using a sodium phosphate buffer solution, which was centrifuged, filtered, and maintained at a pH value of 7. The chip pairs are then placed in a vacuum desiccator, and the bonding strength of the chips is further strengthened by heat treatment. The bonded chips are further diced into different lengths cutting across the metal waveguides. The final configuration results in SPP waveguides with erbium doped glass gain medium surrounding them as illustrated in Figure 1b. Figure 2a shows scanning electron microscope (SEM) image of the bonded sample's cross section. The substrate and the superstrate are well bonded, with a thin interlayer formed during low temperature bonding process.

To find the distinct features of SPP amplification by stimulated emission, we perform both pulsed and continuous mode measurements with signal and pump lasers. To this end, the input fiber is precisely aligned to SPP waveguides, and the output from the waveguides is imaged onto an IR CCD using a 40$\times$ objective. Figure 2b shows the observed and calculated (inset) mode profile. The SPP nature of the propagating mode is verified by measuring the polarization dependence and sensitivity to misalignments of the input fiber. The transmission of the SPP mode through the waveguide is measured by an optical spectrum analyzer and a photodetector. The output fiber is carefully aligned with the SPP waveguide so that maximum output power and strong polarization extinction ratio of >20 dB are obtained, which ensures the collection of only the guided SPP mode.

We measure the total insertion loss for the signal SPP mode without any pump, and it is found to be 39 dB for an 8 μm wide and 8 mm long SPP waveguide. The total insertion loss includes passive waveguide loss, coupling loss between the waveguide and the fibers, and absorption loss by active Er ions in the absence of pumping. The estimated passive waveguide loss by excluding the coupling and Er absorption losses is 27.5 dB. This passive waveguide loss includes both intrinsic loss in the metal and scattering loss. Scattering loss is expected due to imperfect bonded interface,
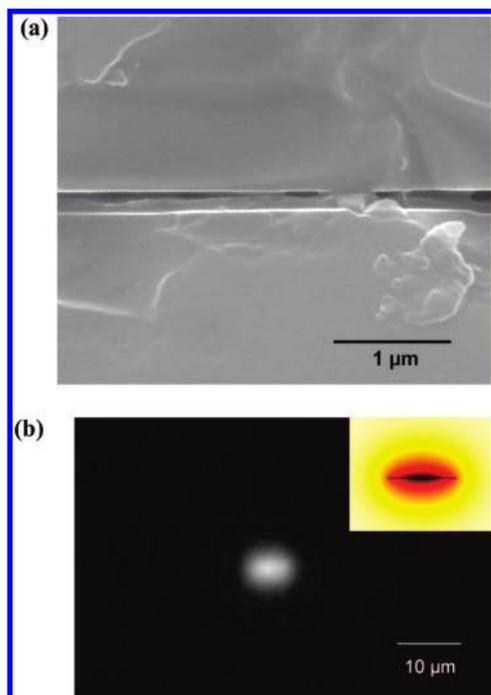
Figure 2. (a) Cross sectional scanning electron microscope image of the bonded Er-doped glass chips. The chips are bonded with a thin bonding interlayer formed during low temperature bonding process and heat treatment. (b) Mode profile image of signal SPP acquired by an IR CCD camera. The inset shows calculated mode (power distribution) of signal SPP. The thickness of the metal strip is 22 nm and the width is 8 μm.

which breaks locally the symmetry of the dielectric environment surrounding the SPP waveguide, causing increase of radiation and coupling loss.

We next probe our system in the small signal limit where the gain is not depleted by the signal. We pump the Er ions with SPP mode using the same fiber-end coupling as the signal. The signal SPP stimulates the transition of the erbium ions from the upper $^4I_{13/2}$ manifold to the ground $^4I_{15/2}$ manifold, resulting in a stimulated emission, thereby enhancing the signal SPP. To study the dynamics of the stimulated emission of SPPs, we modulate the injection current of the signal and pump lasers, resulting in pump and signal square pulses, and measure the signal enhancements as a function of the pump power. We use signal and the pump pulse durations of 150 and 500 μs with 1 ms period and no overlap in time domain. The time delay between the pump and signal pulses (30 μs) is much shorter than the lifetime, $\tau = 7.9$ ms, of the excited-state of erbium ions, which ensures that the population of the excited state is virtually maintained to interact with the signal SPP pulse. The measured signal pulses in time domain by the photodetector with increasing pump power are displayed in Figure 3, and the signal enhancement versus average pump power is plotted in the inset.

A key characteristic in Figure 3 is that the ground level of the signal remains almost unchanged even at the maximum average power of 92 mW. This unambiguously confirms that the contribution of amplified spontaneous emission is negligible in the observed signal enhancements. We find that
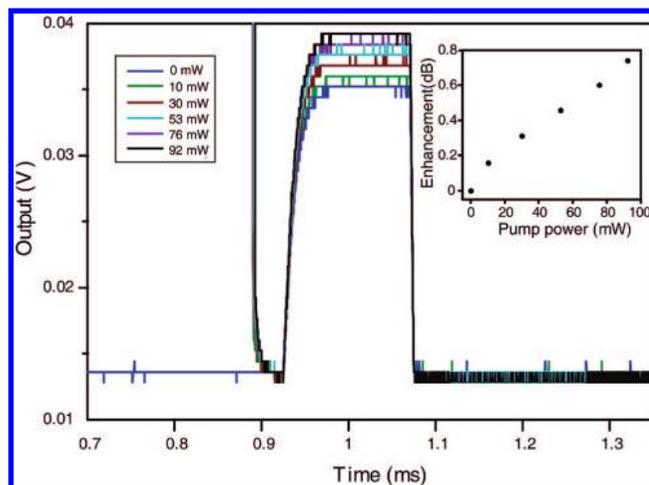
Figure 3. Enhancement of the signal in pulsed mode for different pump powers. The pulse widths of the signal and the pump are 150 and 500 μs with a 1 ms period, respectively. The vertical lines on the left of the signal pulses are the falling edge of the pump pulse. The signal enhances with increasing pump power; however, the ground level of the signal does not change, indicating that the contribution of amplified spontaneous emission is negligible and the enhancements are due to stimulated emission process. The inset shows observed signal enhancements against pump power.
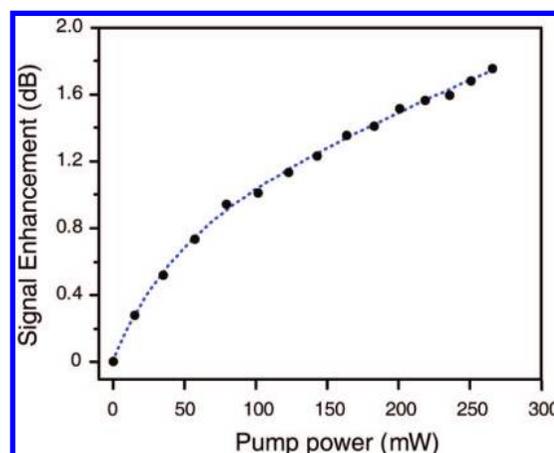
Figure 4. Enhancement of the signal as a function of pump power in continuous mode. At the maximum pump power of 266 mW, the signal is ~50% enhanced. The dotted line indicates a guide for eyes. The rapid decay of the pump SPP along the waveguide limits the total enhancement of the signal SPP.

the maximum signal enhancement in pulse mode is 0.74 dB (18%). In continuous wave mode, however, a maximum signal enhancement of ~50% (1.73 dB) was recorded at a higher pump power of 266 mW (see Figure 4). Nevertheless, the maximum enhancement is still less than the absorption of Er-doped glass at 1532 nm, which indicates that the stimulated emission rate of signal SPP is smaller than the stimulated absorption rate.

While the rapid decay of the pump power along the waveguide limits the enhancement to relatively small values, other factors such as thermal effects and imperfections in the bonded interface may also affect the enhancement. Thermal effects arise as optical pump energy is delivered to erbium ions and as pump energy is dissipated into heat due to ohmic losses in metal. These thermal effects are dual: First,

the optical gain deterioration due to change in occupation probability density of each manifold according to Boltzmann distribution and the probability of occupation for levels at the top of each manifold increases with increasing temperature.[29] Second, metal loss increases due to increased electron scattering rates;[30] theoretical estimation shows that the change in passive propagation loss due to increased metal loss, and the gain deterioration effects are only partially responsible for the gain limitation at high pump powers. In addition to the thermal effects, the optical quality and the possible nonuniformity of the bonded interface along the SPP waveguide provides varying local signal enhancements. On the basis of the measurements of different SPP waveguides, we suspect that the bonding interface plays a crucial role in reducing signal enhancements and the quantification of this effect on signal enhancement, however, is elusive. Although the signal enhancements appear to be small, Figure 4 clearly illustrates that we are far from saturation. There is enough room for further enhancements by reducing the propagation loss down to a few dB/cm levels and thus ensuring slower decay of the pump and signal power.

In conclusion, we demonstrated a direct evidence of stimulated emission of surface plasmon polaritons using pump and signal SPP. Our system is well-suited to study the dynamics of SPP amplification. Further, the coupling of a single emitter or an ensemble of emitters in the excited state to SPP mode can be studied, which opens a route to new experiments in weak and strong coupling regimes. An improved system providing a symmetric dielectric environment surrounding the SPP waveguide, for example, polymers containing active rare-earth ions, could be used to overcome the metal losses and achieve SPP lasers with optical feedback mirrors or gratings even at ambient temperature. Such systems like those described in this letter can be used for integrated optical devices.

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