SPECIAL ISSUE: Optical Gain Materials towards Enhanced Light-Matter Interactions

Loss compensation of surface plasmon polaritons in organic/metal nanowire heterostructures toward photonic logic processing

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ABSTRACT Surface plasmon polaritons (SPPs) are crucial for the development of next generation information and communication technologies. However, the ohmic losses inherent to all plasmonic devices seriously limit their practical application in on-chip photonic communications. Here, loss compensation of SPPs and their application in photonic logic processing was demonstrated in rationally designed organic/silver nanowire heterostructures. The heterostructures were synthesized by inserting silver nanowires (AgNWs) into crystalline organic microwires, which served as a microscale optical gain medium. These heterostructures with large organic/metal interfacial areas ensured the efficient energy transfer from excitons to SPPs. Gain for subwavelength SPPs in the heterostructure was achieved through stimulated emission of strongly confined SPPs. Furthermore, cascade gain was performed to realize basic nanoscale photonic devices, such as Boolean logic units. The results would pave an alternative avenue to incorporating SPP-enhanced devices into hybrid photonic circuitry.

Keywords: surface plasmon, nanowire heterostructure, organic nanowire, loss compensation

INTRODUCTION

Surface plasmon polaritons (SPPs), the coherent electron oscillations at metal-dielectric interfaces, can highly concentrate and channel light at subwavelength scale [1–4], making them ideal candidates for nanoscale optical and optoelectronic devices that hold great potential in nanophotonics [5–10], imaging [11,12] and high-throughput sensing [13]. Nevertheless, the ohmic losses

inherent to all plasmonic devices seriously limit their practical application in on-chip photonic communications [14–17]. Introducing optical gain into the dielectric material adjacent to the metal nanostructures has been identified as an effective means to compensate for the loss [18–21]. To date, the loss compensation of SPP has been demonstrated with various gain systems, such as lanthanide ion-doped glasses [22], quantum dot (QD)-doped films [23,24] and dye solutions [25]. However, these hybrid systems suffer from either large device sizes or sophisticated fabrication techniques [26], making them unsuitable for highly integrated photonic circuits and systems.

Organic crystals with abundant intermolecular interactions allow for numerous low-dimensional nanostructures [27,28], such as nanowires and nanobelts [29], which permit the integration with plasmonic nanostructures without the limit of strict lattice matching and sophisticated epitaxial technology [30]. Especially, the embedded organic/metal hybrid systems can not only ensure the output of strongly confined SPPs at subwavelength scale [31], but also benefit the exciton-plasmon coupling for SPP compensation due to their large organic/metal interfacial areas [32,33]. More importantly, organic assemblies exhibit abundant excited-state processes and high stimulated emission cross section, making it possible to provide excellent optical gain for loss compensation of subwavelength SPPs [34]. The compensation accompanied with the nonlinear increase of SPP signals would offer an effective approach to realize imperative photonic devices, such as nanoscale logic

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elements [18,19,35].

Herein, we propose a strategy to realize loss compensation of SPP signals in organic/silver nanowire (AgNW) heterostructures and their application in photonic logic processing. The heterostructures were constructed by inserting AgNWs into crystalline organic microwires, which served as an effective optical gain medium at microscale. These heterostructures with large organic/metal interfacial areas permit the efficient energy transfer from excitons to SPPs. The loss compensation of SPPs was demonstrated in the heterostructures based on the stimulated emission of strongly confined SPPs. Furthermore, cascade gain was performed in an individual heterostructure with two excitation spots, which enabled us to achieve basic nanoscale Boolean logic unit. These results offer a novel understanding of the assembly mechanism of hybrid materials, and provide a valuable guidance to incorporating SPP-enhanced components into nanoscale hybrid photonic circuitry.

EXPERIMENTAL SECTION

Materials

1-(4-Bromo-2-hydroxy-phenyl)-3-[4-(1-ethyl-propyl)-phenyl]-propenone (BHEP) was designed and synthesized through the classical Claisen-Schmidt condensation reaction [36]. The AgNWs were synthesized by reducing AgNO_3 with ethylene glycol (EG) in the presence of polyvinylpyrrolidone (PVP). Silver nitrate (AgNO_3), PVP and EG were purchased from Aldrich Chemical Co., and used without further purification.

Preparation of BHEP/AgNW heterostructures

The AgNW solution (20 μL , 2 mg mL^{-1}) in ethanol was dropped on a quartz plate. After the complete volatilization of ethanol, 400 μL of BHEP solution (1 mg mL^{-1}) in cyclohexane was subsequently dropped on the same plate. With the gradual evaporation of the solvent, BHEP molecules nucleate and self-assembled into one-dimensional (1D) microwires, and thus grew around the AgNWs. As a result, the BHEP/AgNW heterostructures were obtained on the quartz plate after the complete volatilization of cyclohexane. It is worth noting that the yield of the organic/Ag heterostructure with two excitation spots is lower than that of the sample with only one excitation spot.

Characterization

The morphology of the BHEP/AgNW heterostructure was examined with scanning electron microscopy (SEM,

FEI Nova NanoSEM450) and transmission electron microscopy (TEM, JEOL JEM-2010). Photoluminescence (PL) lifetime images were taken with fluorescence-lifetime imaging microscopy (FLIM) by scanning the samples with a 405 nm picosecond pulse laser. The FLIM (PicoQuant) was composed of picosecond pulsed diode laser (PDL800-D), fiber coupling unit (FCU II), laser scanning microscope (Olympus FV-1000), four-channel detector router (PHR 800), and photomultiplier detector assembly (PMA Series). The absorption and fluorescence spectra were measured on a UV-visible spectrometer (Perkin-Elmer Lambda 35) and a fluorescent spectrometer (Hitachi F-7000), respectively. Bright-field optical images and fluorescence microscopy images were taken from an inverted fluorescence microscope (Nikon Ti-U), by exciting the samples with a mercury lamp. The 532 nm CW laser as the probe laser beam was focused on the tip of AgNW to excite the SPPs. The focused pulse laser beam (400 nm, 150 fs, 1000 Hz) was used to pump the BHEP wires through neutral density filter and half-wave plates. The spatially resolved spectra were measured with a monochromator (Princeton Instrument Acton SP 2300i) connected with an EMCCD (Princeton Instrument ProEM 1600B).

RESULTS AND DISCUSSION

The strategy for loss compensation of SPP in an organic/metal heterostructure is schematically illustrated in Fig. 1a. SPPs launched by a continuous wave (CW) probe laser propagate through the organic/metal junction and scatter into the free space from the other distal end. Upon pulsed illumination, the ordered molecular aggregates around the plasmonic waveguide would permit the energy transfer from excitons to the propagating SPPs, which leads to stimulated emission of SPPs and thus the loss compensation. Here, BHEP (Fig. 1b) was designed and synthesized as the model compound for the study on loss compensation of SPPs due to the following reasons [36]. (i) Compared with the reported compounds used in organic/metal heterostructures [32], BHEP exhibits typical excited-state intramolecular proton transfer process as an efficient four energy-level structure, which would benefit the optical gain and amplification due to the guaranteed population inversion without obvious self-absorption (Fig. S1) [34]. (ii) The Br and O atoms of BHEP have a potential coordination effect with metals and would thus benefit the growth of crystals around the plasmonic waveguide [37]. In addition, the hydrogen-bond interaction ($\text{O}\cdots\text{H}$) facilitates the crystal growth along a specific direction to form a 1D nanostructure

Figure 1 Design and fabrication of the BHEP/AgNW heterostructures. (a) Schematic illustration of an organic/metal heterostructure for SPP loss compensation. (b) Synthetic route of BHEP. (c) Bright-field microscopy image of the organic/metal nanowire heterostructures. Scale bar is 10 μm . Inset: magnified SEM image of the junction in a typical heterostructure. Scale bar is 2 μm . (d) TEM image of the heterostructure. Scale bar is 500 nm. Insets: selected area electron diffraction (SAED) patterns of BHEP and AgNWs. (e) Schematic diagram for the growth process of the organic/metal nanowire heterostructures. The corresponding SEM images are also shown. Scale bars are 2 μm .

(Fig. S2), making it possible to achieve hybrid nanowire heterostructure with small device footprint. As for the plasmonic materials, chemically synthesized AgNWs with diameters of ~ 180 nm were utilized to support the SPPs due to their atomically smooth surfaces and low propagation loss (Fig. S3). All the AgNWs used in this work were not coated with the surfactants.

The heterostructures were prepared by embedding AgNWs in BHEP crystals during crystal growth in the liquid phase (Fig. S4). In a typical preparation, AgNWs dispersed in ethanol (2 mg mL^{-1}) were first drop-cast onto a quartz wafer, and then 0.5 mL of BHEP solution (cyclohexane, 1 mg mL^{-1}) was dispersed on the same substrate. With the evaporation of the solvent, the molecules self-assembled into 1D microwires, and grew around the AgNWs. As a result, BHEP/AgNW hetero-

structures were obtained and dispersed on the substrates for the subsequent characterizations (Fig. 1c). The SEM image (Fig. 1c, inset) indicates that the AgNWs are partially embedded in the organic microstructures, which can ensure a stable exciton-SPP coupling insensitive to mechanical vibrations [30,38]. The adopted diameter of AgNWs is ~ 180 nm, and the corresponding field energy portion of the plasmon mode on the surface is nearly 80% [39], which is suitable for coupling with the gain molecules nearby. Meanwhile, the thickness of the molecular layer is supposed to be slightly higher than the AgNW diameters, which would make AgNWs surrounded by abundant gain molecules and thus benefit the SPP loss compensation. The organic single-crystalline nanowire at the junction (Fig. 1d) possesses low scattering density and smooth surfaces, thus making it possible to provide effi-

cient optical gain for loss compensation of subwavelength SPP signals.

To investigate the growth mechanism of the heterostructures, we studied the evolution of their morphology over growth time (Fig. 1e). At the initial stage, AgNWs were dispersed in the solvent due to the wetting of the solvent that can reduce the van der Waals interaction between the AgNWs and the substrate. During the evaporation of the solvent, the BHEP molecules nucleated and assembled into 1D crystals, and then the AgNWs were stuck on the organic microwires. The BHEP crystals nearby the AgNWs grew around the wire driven by avoiding the major disruption of lattice structure and reducing the overall interfacial energy, which result in the final embedded heterostructures. Following this growth process, we can adjust the width of the nanowire to modulate the gain region by changing the concentration of BHEP units. For instance, the width of nanowires can be tuned from ~ 2 to $8\ \mu\text{m}$ by changing the concentration of BHEP molecules from 1 to $4\ \text{mg mL}^{-1}$ (Fig. S5).

These embedded heterostructures provide much larger organic/metal interfacial area as compared with the junction constructed by simple point contact, which would be beneficial for the energy transfer from excitons

to SPPs in the AgNWs [32,40]. This merit was demonstrated by studying the decay of excitons in a typical heterostructure as shown in Fig. 2a. Here, the decay kinetics was characterized by FLIM, which can be used to record the fluorescence lifetimes of chromophores at each spatially resolvable element of a microscope image. The distribution of the lifetime manifests that the PL decay at the junction is obviously faster than that at the body of the organic wire (Fig. 2b, c and Fig. S6). The faster decay reveals that the larger contact area of the embedded structure would benefit the energy transfer from excitons to the SPPs. Under UV (330–380 nm) excitation, small bright red spots were observed at the AgNW distal ends (Fig. S7), which not only exhibit the typical feature of the SPP waveguide, but also confirm the efficient exciton-plasmon coupling at the embedded sections.

In the crystalline heterostructure, the highly ordered molecular aggregates would induce the gain anisotropy around AgNWs [32], facilitating the investigation of the relationship between the pump polarization and excitation efficiency for the realization of high optical gain. Here, a single heterostructure was excited with a linearly polarized pump laser (400 nm, 150 fs), and the parameter θ refers to the angle between the long axis of the BHEP

Figure 2 Optical characterizations of the BHEP/AgNW heterostructures. (a) Bright-field microscopy of an embedded nanowire heterostructure. Scale bar is $5\ \mu\text{m}$. (b) FLIM image of the heterostructure in (a). (c) Average lifetimes across the junction (marked with red line in (b)). (d) Bright-field and PL images of the heterostructures with different cross angles between the pump polarizations and long axis of the BHEP microwire. Scale bar is $5\ \mu\text{m}$. (e) Normalized PL intensities I_θ versus θ . I_θ is the SPP intensity detected at the BHEP/Ag junction. (f) The spatial relationship among BHEP molecular transition dipole moment (blue arrow) and the preferential grow direction (orange arrow) of the BPEA NW. (g) PL spectra collected from the junction with the pump laser polarization parallel to the BHEP wire. Inset: Bright-field and PL images of the heterostructure. Scale bar is $5\ \mu\text{m}$.

wire and the pump laser polarization (Fig. 2d). It was observed that when θ was 0° – 30° , the emission from the exciting point of BHEP NW was much stronger than that when θ was increased to 60° – 90° , which reveals that the pump polarizations would significantly influence the optical gain in the hybrid system. The corresponding emission intensity oscillates between the maximum and the minimum values with θ ranging in a period of 180° (Fig. 2e), which should be a direct consequence of the polarization-dependent excitation efficiency. As shown in Fig. 2f, the BHEP molecules stack nearly parallel to the b axis, tilted at an angle of 26° , and the transition dipole of BHEP is along the molecular long axis (Fig. S8). Therefore, when θ is 0° or 180° , namely, the pump laser polarization is parallel to the BHEP wire, the largest excitation efficiency can be obtained. As shown in Fig. 2g, with increasing pump pulse energy over a threshold (Fig. S9), sharp PL peaks in the gain region was observed and amplified, which indicated the lasing action in the heterostructure, showing great potential to offer the ef-

ficient optical gain for SPP loss compensation.

These heterostructures with efficient exciton-plasmon coupling and high optical gain provide an opportunity to investigate the loss compensation of SPPs. The gain measurements were carried out on a home-built far-field microphotoluminescence system (Fig. S10). As shown in Fig. 3a, SPPs were launched by focusing a 635 nm CW probe laser beam at the left tip of AgNW, and was supposed to be loss-compensated through stimulated emission by focusing the linearly polarized pump laser on the BHEP-AgNW junction [25]. SPP gain would be achieved if the output signal at the right NW tip in panel III (pump + probe) was enhanced compared with the value obtained from panel I (probe only) after correcting for the background spontaneous emission in panel II (pump only). As shown in Fig. 3b, a clear enhancement of the output signal was observed with optical pumping, i.e., $I_{\text{pump+probe}} - I_{\text{pump}} > I_{\text{probe}}$. To evaluate the internal optical gain, the output signal enhancement factor was defined as: $f = (I_{\text{pump+probe}} - I_{\text{pump}}) / I_{\text{probe}}$, where I refers to the peak value

Figure 3 Loss compensation of SPPs. (a) Bright-field and PL images of the heterostructure excited by a probe laser (I), a pump laser (II), and both the probe and pump lasers (III), respectively. Scale bar is 5 μm . (b) PL spectra collected from the right tip of the AgNWs under the three different excitation conditions shown in (a): (i) probe only; (ii) pump only; (iii) pump + probe. The inset shows that an output signal enhancement factor of $f = 1.5$ was obtained from the spectra at the probe wavelength. (c) Plots of SPP signal enhancement factor f vs. the pump intensity.

of the corresponding intensity at 635 nm [33]. The inset in Fig. 3b demonstrates a 1.5-fold enhancement of the SPP output at a pump power of $11.2 \mu\text{J cm}^{-2}$.

A clear threshold for SPP gain was observed as a function of pump irradiance (Fig. 3c), while maintaining the constant power of the probe laser and the polarization of the probe laser. The SPP signal enhancement factor of $f > 1$ was evident at a pump irradiance of $\sim 4 \mu\text{J cm}^{-2}$, and f increased linearly with pump irradiance above this threshold. The observation of both threshold and linear pump power dependence are signatures of stimulated SPP emission [18,41]. For larger pump powers ($> 12 \mu\text{J cm}^{-2}$), lasing in the organic crystal was observed and became the dominant gain process, which suppressed the further enhancement of SPP signals (Fig. S11). We can speculate that the SPP-enhanced process as well as the performances of nanophotonic devices could be improved by properly designing the hybrid structure [42,43].

Thanks to the flexibility of the liquid-phase fabrication method, self-assembled organic/Ag heterostructures permit the AgNW to be embedded into more than one organic wire [44], which may offer cascade gain process,

and prompt us to perform optical signal processing at the nanoscale [45]. For example, the AgNW was simultaneously embedded into two organic wires (Fig. 4a), resulting in two individual heterojunctions in a single system. The pulse laser was split into two beams to pump the two BHEP-AgNW junctions, respectively (Fig. S10). The corresponding PL microscopy images in Fig. 4b exhibit the four different combinations of two pump lasers as input signals. In this heterostructure, the excitation spot with optical pumping was defined as 1, while that without optical pumping was defined as 0. Then these four images represent four combinations of input values: (1, 1), (1, 0), (0, 1) and (0, 0), respectively, which is in accordance with the Boolean logic inputs. Such heterojunction forms a nanoscale logic gate with two input terminal (I1 and I2) and one output terminal (O). The value of the SPP output was obtained after correcting for the background spontaneous emission.

The function of the gate depends on the intensities of SPPs scattered at O port for different gain cases (Fig. 4c). Assuming that the maximum in Fig. 4d is 1 arbitrary unit (au), we find that $O(1, 1) = 1 \text{ au}$, O

Figure 4 Photonic logic processing based on loss compensation of SPPs. (a) Bright-field image of the heterostructure with two excitation spots. Scale bar is 5 μm . (b) PL microscopy images under the excitations of four different combinations of the two excitation spots. Here, 1 and 0 were defined respectively according to the optical gain in the junction. (c) The formed basic logic gate with two simultaneous inputs and one output signal. (d) The scattering spectra of SPPs at the O terminal under the excitations shown in (b) after correcting for the background spontaneous emission. T1 and T2 represent the thresholds for OR and AND gates, respectively. (e) Summary of optical logic operations for AND and OR gates.

$(0, 1) = O(1, 0) = 0.55$ au, and $O(0, 0) = 0$ au. By defining an intensity threshold (T) to separate ON and OFF states among these values, either OR or AND logic operations could be achieved in such hybrid structure. For example, for a threshold of 0.4 au (T_1 , the dotted line in Fig. 4d), $(I_1 = 1, I_2 = 1)$ results in $O = 1$, $(I_1 = 1, I_2 = 0)$ results in $O = 1$, and $(I_1 = 0, I_2 = 1)$ results in $O = 1$, showing the performance of an OR gate. While, when the higher threshold (such as $T_2 = 0.85$ au, the dashed line in Fig. 4d) was defined, only $(I_1 = 1, I_2 = 1)$ results in $O = 1$, and all other three input combinations lead to $O = 0$, which serves as an AND gate. The total outputs with different thresholds have been summarized in Fig. 4e, demonstrating an OR or AND gate. We believe that this concept can be further generalized and expanded to more complex SPP-enhanced components to serve as various nanoscale devices, such as splitters, dichroic mirrors, routers, and so on [46].

CONCLUSIONS

In summary, we report a type of embedded organic/silver nanowire heterostructure to realize loss compensation of subwavelength signals and their application in photonic logic processing. The heterostructures were constructed by inserting AgNWs into crystalline organic microwires, which functioned as a microscale optical gain medium. Due to the large organic/metal interfacial area, these heterostructures exhibited efficient exciton-plasmon coupling, and transferred the exciton energy to SPPs. Loss compensation of SPPs was demonstrated in the heterostructure based on stimulated emission of strongly confined SPPs. Furthermore, the individual heterostructure with two excitation spots possessed cascade gain, which enabled us to achieve basic Boolean logic unit at nanoscale. These results offer a novel understanding of the assembly mechanism of hybrid materials, and are essential for incorporating plasmonic amplifiers as practical components into high-capacity photonic circuits.

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Author contributions Zhao YS conceived the idea and supervised the project. Lv Y designed the experiments and prepared the materials. Lv Y and Xu FF performed the optical measurements. Lv Y, Xu FF and Wang K put forward the theoretical model and contributed to the theoretical calculations. Lv Y, Li YJ and Zhao YS analyzed the data. Lv Y, Li YJ, and Zhao YS wrote the manuscript. All authors discussed the results and commented on the manuscript.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

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有机/金属纳米线异质结中的SPPs损耗补偿用于光学逻辑运算

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摘要 表面等离子体激元(SPPs)对于下一代信息通信技术的发展至关重要。然而, SPPs器件固有的欧姆损耗限制了它们在片上光学通讯中的实际应用。本工作中, 我们在合理设计的有机/金属纳米线异质结中实现了SPPs的损耗补偿及其在光学逻辑运算中的应用。通过将银纳米线(AgNWs)插入结晶有机微丝中来合成异质结构, 并将其用作微米级光学增益介质。所制备的异质结具有较大的有机/金属界面面积, 能确保从激子到SPPs的高效能量转移。异质结构中亚波长SPPs的增益是通过强烈限制SPPs的受激发射实现的。此外, 我们还实现了SPPs的级联增益, 并将其用于构建微纳光子学器件, 例如Boolean逻辑单元。相关结果为具有SPPs增强功能的器件用于混合光子学回路提供了新的思路。