Study on All-Optical Regulation of Exciton-Polaritons Based on Surface Plasmons

Authors: Dandan Liu, Chunliang Wang, Dandan Liu

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Abstract

Objective:Traditional von Neumann computing architecture suffers from inherent bottlenecks such as the separation of storage and computation, limited data bandwidth, and high energy consumption. Electronic neuromorphic devices can hardly meet the requirements of next-generation high-speed, low-power, and highly parallel intelligent computing systems.Methods:All-optical devices constructed from photoresponsive materials provide a promising solution by simulating biological synaptic plasticity and brain-like memory functions. Surface Plasmon Polaritons (SPPs) exhibit strong electromagnetic field confinement and enhancement, which can significantly improve the sensitivity of photoresponsive materials and reduce the operating energy consumption of devices.In this study, a strongly coupled system composed of silver-based SPPs and azobenzene-containing Disperse Red 1 (DR1) molecules is designed and fabricated. The strong coupling between SPPs and excitons is realized by optimizing structural parameters, and the all-optical control of exciton-polaritons is achieved through external laser stimulation.Results: The results show that a 50 nm-thick silver film supports the most efficient SPP excitation, and a 50 nm-thick DR1/PMMA film enables the strongest coupling strength with obvious Rabi splitting.Limitations: The device is limited by environmental stability and thickness-precision requirements. Its integration level is low, and the response speed is restricted by DR1 molecular dynamics, which affects its practical application in integrated photonic systems.Conclusions: This work achieves all-optical control of exciton-polaritons and simulates brain-like memory. The device has good performance and universality, providing a feasible scheme for optical neural networks and neuromorphic computing applications.

Full Text

Preamble

All-Optical Control of Exciton-Polaritons Based on Surface Plasmons Dandan LIU,Chunliang WANG Center for Advanced Optoelectronic Functional Materials Research,and Key Laboratoryfor UV-Emitting Materials and Technology of Ministry ofEducation,National Demonstration Center for Experimental Physics Education,School of Physics,Northeast Normal University,5268 Renmin Street,Changchun 130024,Jilin,China.

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Methods:All-optical devices constructed from photoresponsive materials provide a promising solution

by simulating biological synaptic plasticity and brain-like memory functions. Surface Plasmon Polaritons (SPPs) exhibit strong electromagnetic field confinement and enhancement, which can significantly improve the sensitivity of photoresponsive materials and reduce the operating energy consumption of devices.In this study, a strongly coupled system composed of silver-based SPPs and azobenzene-containing Disperse Red 1 (DR1) molecules is designed and fabricated. The strong coupling between SPPs and excitons is realized by optimizing structural parameters, and the all-optical control of exciton-polaritons is achieved through external laser stimulation.

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Conclusions: This work achieves all-optical control of exciton-polaritons and simulates brain-like memory. The device has good performance and universality, providing a feasible scheme for optical neural networks and neuromorphic computing applications.

Key words: surface plasmons polariton(SPPs); exciton polaritons; synaptic plasticity; all-optical regulation.

1. Introduction

With the rapid development of big data, artificial intelligence, and wearable computing devices, the demand for high-speed, low-power, and miniaturized computing hardware is increasing rapidly. However, traditional electronic computers based on the von Neumann architecture are facing increasingly serious bottlenecks. The physical separation of the processing unit and the memory unit leads to massive data shuttling between the two modules, resulting in high latency, high energy consumption, and low processing efficiency. In addition, the continuous reduction of the feature size of electronic devices has gradually approached the physical limit,[1-5] making it difficult to further improve the integration density and performance. Therefore, it is urgent to explore new computing mechanisms and device structures to break through the limitations of traditional electronic systems.[6-7]

Among various platforms supporting strong coupling, Surface Plasmon Polaritons (SPPs) have obvious advantages. SPPs are evanescent electromagnetic waves propagating along the metal-dielectric interface, formed by the coupling between incident photons and free electrons on the metal surface. SPPs can break through the optical diffraction limit and strongly confine the light field in the subwavelength range, thus greatly enhancing the interaction between light and matter. This makes it possible to realize strong coupling at room temperature without an expensive optical microcavity.[8-10] At present, the research on exciton-polaritons based on SPPs is still in the preliminary stage, especially in the application of neuromorphic devices. Most studies focus on the realization and characterization of strong coupling, while the research on all-optical control mechanism, synaptic plasticity simulation, and brain-like memory functions is relatively insufficient. Therefore, it is of great scientific significance and application value to carry out research on all-optical control of exciton-polaritons based on SPPs and explore their applications in optical synapses and brain-like computing.[11-15] In this work, azobenzene-based DR1 molecules are used as excitonic materials, and silver films are used to excite SPPs. By optimizing the preparation process, a stable and efficient SPP-exciton strong coupling system is constructed. The all-optical control of Rabi splitting is realized by using 488 nm laser. Furthermore, the device successfully simulates a variety of synaptic plasticity behaviors including short-term plasticity, long-term plasticity, paired-pulse facilitation (PPF), and the transformation from STM to LTM. The device also has the functions of reversible optical memory and anti-crosstalk, providing a new scheme for the development of high-performance all-optical neuromorphic devices.[16-18]

2.1 Surface Plasmon Polaritons

SPPs are collective oscillations of free electrons on the metal surface coupled with incident electromagnetic fields. They propagate along the metal-dielectric interface and decay exponentially in the direction perpendicular to the interface. The excitation of SPPs requires strict momentum matching conditions. Since the wave vector of SPPs is larger than that of photons in free space, special coupling structures are needed to compensate for the momentum mismatch.

The Kretschmann prism coupling structure used in this study is one of the most effective excitation methods.

The dispersion relation of SPPs is:

(1 2)2

= 0 (+)

(2-1)

where ϵm and ϵd are the dielectric constants of metal and dielectric, respectively. Silver is selected as the plasmonic material because of its low loss and high field enhancement effect in the visible light band. 2.2 Strong Coupling and Exciton-Polaritons When the interaction strength between SPPs and excitons exceeds the decay rate of the system, the system enters the strong coupling regime. In this case, the energy levels of excitons and plasmonic modes split into two new hybrid energy bands, namely the upper polariton branch (UPB) and lower polariton branch (LPB). This splitting phenomenon is called Rabi splitting, which is the most important characteristic of strong coupling.

Exciton-polaritons inherit the properties of both photons and excitons. They have a small effective mass, fast propagation speed, and strong nonlinearity, making them ideal for optical information processing and neuromorphic devices.

2.3 Photoisomerization of DR1 Molecules

DR1 is a typical azobenzene derivative with reversible photoisomerization characteristics.

Under the irradiation of 488 nm visible light, DR1 molecules transform from stable trans-structure to cis-structure. After the light is removed, they can spontaneously return to trans-structure under thermal excitation. Meanwhile, under the irradiation of linearly polarized light, DR1 molecules will rotate to the direction perpendicular to the polarization of the incident light, showing obvious photoinduced anisotropy. These two characteristics make DR1 an excellent photoresponsive material for all-optical control devices.

3.1 Materials and Reagents

DR1 powder, PMMA powder, chloroform, acetone, ethanol, and high-purity silver target are used. All reagents are used without further purification. 3.2 Experimental Setup and Measurement Systems To characterize the excitation and spectral properties of surface plasmon polariton (SPP) modes, a total-reflection optical system was established based on the Kretschmann configuration, as illustrated in Fig. 3 [Figure 3: see original paper].6.

In this system, a HL-2000 halogen tungsten lamp was employed as the broadband light source, and a QE65pro high-sensitivity fluorescence spectrometer was used to detect the reflected spectra. During optical alignment, the positions and heights of the light source, apertures, polarizer, and spectrometer were carefully determined. The white light beam was first aligned to pass through the center of an aperture near the source. After fixing the height and position, this aperture was moved close to the sample. Another aperture was placed at the reference position, and its height was adjusted so that the light beam passed through its center. Fine alignment was performed to ensure the beam traveled through the centers of both apertures to achieve collimation.

After confirming consistent height between the source and apertures, a horizontal polarizer was placed near the sample to transmit light. The position of the receiver was finely adjusted so that the reflected spectrum could be observed on the computer.

3.1 Schematic diagram of the prism-coupled reflection measurement system based on the Kretschmann configuration.

To investigate exciton-polaritons supported by SPP modes, an all-optical modulated total-reflection system for exciton-polaritons (EP) was constructed, as shown A photograph of the

setup is presented A 488 nm p-polarized laser was introduced perpendicularly to the sample–prism interface to serve as the pump beam. By adjusting the illumination time, intensity, and polarization angle of the pump light, fully reversible all-optical modulation of Rabi splitting was realized.

The probe path still used the HL-2000 halogen tungsten lamp and QE65pro high-sensitivity spectrometer, following the alignment procedure shown in Fig. 3.1. The pump path employed a 488 nm p-polarized laser incident normally onto the sample surface. Precise adjustment ensured that the pump and probe beams were focused at the same position on the sample. The reflected optical signal was collected by the spectrometer and transmitted to the computer for real-time data acquisition and analysis, yielding the reflection spectra.. 3.3 Characterization of Experimental Samples

3.3.1 Characterization of SPP Samples

In the Kretschmann total-reflection configuration, SPPs are excited by evanescent waves at the Ag/air interface. The thickness of the Ag film is a key factor affecting the resonance peak and dip depth of SPPs. Therefore, total-reflection spectra of SPPs excited under different Ag thicknesses were measured. According to Maxwell’ s equations and metal-dielectric boundary conditions, during SPP excitation, the optical field is strongly confined near the interface and decays exponentially perpendicular to the interface. Accordingly, numerical simulations of the optical field distributions near the Ag/air interface for different Ag thicknesses were performed, as shown in Fig. 3.2.

It can be clearly seen that the field intensities at the Ag/air interface for 30 nm, 80 nm, and 90 nm Ag films are lower than that for 50 nm Ag. After 50 nm, the interfacial intensity gradually decreases. For all thicknesses (30 nm, 50 nm, 80 nm, 90 nm), the optical field intensity decays exponentially in air along the direction normal to the interface.

3.2 Simulated optical field intensity distributions along the z-direction at the Ag/air interface for different Ag thicknesses: (a) 30 nm, (b) 50 nm, (c) 80 nm, (d) 90 nm.

The above total-reflection spectra were measured under p-polarized light. To further verify that only p-polarized light can effectively excite SPPs, the total-reflection spectrum of 50 nm Ag

under s-polarized light was measured, as shown in Fig. 3.3. No obvious resonant dip appears, confirming that s-polarized (TE) light cannot excite SPPs.

According to the momentum-energy dispersion relation, SPP excitation is highly sensitive to the incident angle. Angle-resolved total-reflection spectra were further compared for Ag thicknesses of 30 nm, 40 nm, 50 nm, and 60 nm in the wavelength range of 400–900 nm, as shown in Fig. 3.3. The resonant dip of SPPs blue-shifts as the incident angle increases. The 50 nm Ag film shows the narrowest dip width, indicating higher angular and wavelength selectivity, more precise matching to SPP excitation conditions, stronger field localization, and more stable optical response. The minimum reflectance is lower, meaning less energy is directly reflected and more energy is coupled into interfacial evanescent waves, enabling stronger near-field enhancement. Therefore, 50 nm was selected as the optimal Ag thickness for exciting evanescent waves at the Ag/air interface. It can be clearly observed that the position of the resonant dip shifts regularly with the incident angle.

3.3 Measured angle-resolved total-reflection spectra for different Ag thicknesses: (a) 30 nm, (b) 40 nm, (c) 50 nm, (d) 60 nm.

3.4 Measured total-reflection spectrum of 50 nm Ag under TE polarization.

The total-reflection spectrum of 50 nm Ag was further measured near 490 nm, as shown in energy loss at 490 nm in the reflected signal. The minimum reflectance of the dip is below 10%, indicating extremely high SPP excitation efficiency. The full width of the resonant dip is less than 50 nm, revealing high frequency selectivity. These results confirm that the as-prepared 50 nm Ag sample supports high-quality SPP excitation at 490 nm.

4. Results and Discussion

4.1 Optimization of SPP Excitation

The thickness of the silver film is a key parameter affecting SPP excitation. The simulation and experimental results show that the 50 nm-thick silver film has the strongest electromagnetic field enhancement at the Ag/air interface and the narrowest resonance dip in the reflection spectrum, indicating the highest excitation efficiency. Therefore, 50 nm is determined as the optimal thickness of the silver film.

4.2 Realization of Strong Coupling

The absorption peak of DR1 molecules is near 490 nm, which matches the resonance wavelength of SPPs. By optimizing the thickness of DR1/PMMA film, strong coupling between SPPs and excitons is realized. The reflection spectrum shows obvious anti-crossing behavior and Rabi splitting, and two clear polariton branches |P+> and |P-> appear, which proves the successful formation of exciton-polaritons. 4.3 All-Optical Control of Rabi Splitting Under the irradiation of 488 nm laser, DR1 molecules undergo photoisomerization and molecular orientation rearrangement, which changes the excitonic absorption intensity and molecular orientation, thus weakening the coupling strength and reducing the Rabi splitting. The regulation effect can be continuously and reversibly controlled by adjusting the laser power, irradiation time, and polarization state.

Compared with the bare DR1 film without SPPs, the strongly coupled device has a faster response speed and a larger reflectance change amplitude, which means lower power consumption and higher sensitivity in practical applications.

4.4 Simulation of Synaptic Plasticity

The delay response of reflectance after the removal of pump light is used to simulate the postsynaptic current response of biological synapses. The device exhibits typical short-term plasticity (STP) and long-term plasticity (LTP):

1. Under short-time or low-intensity light stimulation, the reflectance changes slightly and recovers quickly after the light is removed, showing short-term memory (STM).
2. Under long-time, high-intensity, or repeated light stimulation, the reflectance changes significantly and recovers slowly, showing long-term memory (LTM).

3. Under continuous double-pulse stimulation, the device shows obvious paired-pulse facilitation (PPF) effect, which is consistent with the characteristics of biological synapses. 4.5 Reversible Optical Memory and Anti-Crosstalk The device has dual-wavelength reflectance response characteristics: the reflectance increases at some wavelengths and decreases at others under the same light stimulation. This unique dual-response characteristic enables the device to distinguish effective signals from interference signals, showing excellent anti-crosstalk ability.

In addition, TM polarized light can be used to write information, and TE polarized light can be used to erase information, realizing reversible all-optical memory function. This function is very important for the construction of rewritable optical storage and neural network systems.

5. Conclusion

In this work, a high-performance all-optical exciton-polariton device based on surface plasmons is designed and prepared. The strong coupling between SPPs and DR1 excitons is realized by optimizing structural parameters. The all-optical control of exciton-polaritons is

achieved by using 488 nm laser stimulation. The device successfully simulates a variety of important synaptic plasticity behaviors, including short-term plasticity, long-term plasticity, paired-pulse facilitation, and the transformation from short-term memory to long-term memory.

The strongly coupled device exhibits faster response speed, larger modulation depth, lower power consumption, and better anti-crosstalk ability than bare DR1 films. Reversible writing and erasing of optical memory are realized by using orthogonally polarized light. The non-contact optical control method makes the device have natural anti-electromagnetic interference ability, which is very suitable for complex electromagnetic environments.

This study provides a new physical mechanism and device scheme for the development of all-optical neural networks and brain-inspired computing systems. The proposed exciton-polariton optical synapse has broad application prospects in optical computing, optical storage, wearable devices, and artificial intelligence chips.

Author Contributions Dandan Liu: Conceptualization, methodology, software, validation, formal analysis, investigation, data curation, writing-original draft, visualization.

Chunliang Wang: Supervision, project administration, funding acquisition, resources, writing-review and editing, conceptualization.

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Note: Figure translations are in progress. See original paper for figures.