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Optical bistability in the plexcitonic Ag-CuCl nanowire

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ABSTRACT

Optical bistability (OB) of plexcitonic Ag–CuCl nanowires (ACNWs) is investigated based on the quasi-static theory. It is found that the strong couplings between the plasmon resonance of inner Ag nanowire and the exciton resonance of outer CuCl shell result in two new plexcitonic modes, which can induce the OBs and hence the two OB regions. Furthermore, the initial wavelengths of the OBs in the ACNW show slight red shifts compared to the resonance wavelengths of the plexcitonic modes. In addition, we find that the plexcitonic modes-induced OBs in the ACNW can be modulated by varying the thickness of outer shell.

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1. Introduction

Optical bistabilities (OBs) in metal nanoparticles and nanostructures have attracted great attention in recent years because of their potential applications in optical switching, optical memory element, and optical logic device [1-5]. In metal nanoparticles, the local fields are dramatically enhanced by surface plasmon resonances (SPRs), exciting remarkable OBs [6-10]. Many groups have reported OB behaviors in many kinds of metal composite nanoparticles. For example, Daneshfar et al. [6,7] investigated the OBs in spherical/cylindrical plasmonic nanoparticles, and found the OB depends on the geometry, size, material, and surrounding medium. In two-phase nonlinear composite materials, the OB behavior is sensitive to the shape and volume fractional of metallic particles [8]. Gao et al. [11,12] investigated OBs of nonlinearshell-coated metallic nanoparticles by means of a self-consistent mean field approximation, and found the OB behaviors critically rely on the geometry of the shell-coated nanoparticle, especially the fractional volume of the metallic core [13]. It was found that the radial dielectric anisotropy in the metallic spherical and cylindrical nanoshells can significantly influence the OB effects [14,15]. Furthermore, the nonlocality was proposed to realize the tunable OB behaviors in the nonlinear plasmonic core-shell cylinders, indicating the nonlocalityenhanced OB [16,17].

More recently, plexcitonic nanostructures, composed of metal nanostructures and exciton materials, have attracted considerable interest due to the applications in surface enhanced spectrum [18], light harvesting [19], photocatalysis [20], and sensor [21]. Surface plasmons (SPs) of metal nanostructures can couple with the exciton resonances of exciton

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systems when they are in close proximity [22–29]. The weak plasmonexciton couplings tend to enhance the absorption or emission of exciton systems [22]. The strong couplings between the SP and exciton modes will result in two new plexcitonic modes by the energy exchange, and hence the Rabi splitting occurs [22–26]. In addition, the strong plasmon–exciton couplings can induce the Fano resonance, induced transparency, and enhanced optical nonlinearity [23,26–29]. However, little work has been carried out on the OB behavior of the plexcitonic modes in metal–semiconductor hybrid nanostructures [30,31].

In this paper, we investigate the optical bistabilities (OBs) in plexcitonic Ag–CuCl nanowires (ACNWs). The far-field, near-field, and OB properties of the ACNW have been calculated by using the quasi-state theory. The strong couplings between the dipole plasmon resonance of inner Ag nanowire and the exciton resonance of outer CuCl shell result in two new plexcitonic modes. The OB behaviors induced by these two plexcitonic modes are studied in detail. In addition, we discuss the influence of the geometry on the OB behavior of the ACNW.

2. Theory model

Fig. 1 shows the schematic illustration of an ACNW composed of an inner Ag nanowire with radius r_1 and an outer CuCl shell with thickness $(r_2 - r_1)$. The dielectric function of inner Ag nanowire ε_1 has real and imaginary frequency-dependent components and can be expressed as [26]

$$\epsilon_1(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma_f} + \chi_{\infty},\tag{1}$$



Fig. 1. (color online) Schematic of an Ag-CuCl nanowire.



Fig. 2. (color online) Scattering spectrum and the corresponding local field enhancement factor of the ACNW. Here, $r_1 = 10$ nm and $r_2 = 13$ nm.

where the background susceptibility $\chi_{\infty} = 4$, the bulk plasma frequency $\hbar \omega_p = 8.9780 \text{ eV}$, and the bulk collision frequency $\hbar \gamma_f = 0.0212 \text{ eV}$. The dielectric function of outer CuCl shell ϵ_2 can be expressed as [32]

$$\varepsilon_2(\omega) = \varepsilon_{\infty} + A \frac{\gamma}{\omega_0 - \omega - i\gamma},\tag{2}$$

where $\varepsilon_{\infty} = 5.59$, $\omega_0 = 5.0169 \times 10^{15}$, $\gamma = 7.4757 \times 10^{10}$, and A = 632.

In our investigation, the diameter of the ACNW is much smaller than the incident wavelength. The incident field could be regarded as being spatially uniform over the extent of the particle. Thus, the optical responses of the ACNW can be calculated using the quasi-static approximation [33]. As an incident electric field $\vec{E}_0 = E_0 \vec{e}_x$ travels along *y*-axis and is scattered by the ACNW, the obtained local electric fields \vec{E}_L in the core can be expressed as [34]

$$\vec{E}_{\rm L} = \frac{4\epsilon_2\epsilon_3r_2^2}{\left(\epsilon_3 + \epsilon_2\right)\left(\epsilon_2 + \epsilon_1\right)r_2^2 + \left(\epsilon_1 - \epsilon_2\right)\left(\epsilon_2 - \epsilon_3\right)r_1^2}\vec{E}_0.$$
(3)

The obtained scattering coefficient is [35]

$$Q_{sca} = \frac{\sigma_{sca}}{\pi r_2^2} = \frac{k^3}{16\pi r_2^3 \varepsilon_0^2} |\alpha|^2,$$
(4)

$$\alpha = 2\pi\varepsilon_0 \frac{\left(\varepsilon_2 - \varepsilon_3\right)\left(\varepsilon_2 + \varepsilon_1\right)r_2^2 + \left(\varepsilon_1 - \varepsilon_2\right)\left(\varepsilon_2 + \varepsilon_3\right)r_1^2}{\left(\varepsilon_3 + \varepsilon_2\right)\left(\varepsilon_2 + \varepsilon_1\right)r_2^2 + \left(\varepsilon_1 - \varepsilon_2\right)\left(\varepsilon_2 - \varepsilon_3\right)r_1^2}r_2^2,\tag{5}$$

where *k* is the wave number and α is the electric polarizability.

It is well known that the enhanced local electric fields in metal nanostructures can excite OBs [19–24]. We consider the inner Ag nanowire with the nonlinear part and then study the OB behaviors of the

ACNW. In this case, the dielectric function of inner Ag nanowire should be modified as $\varepsilon_1\left(\vec{E}_N\right) = \varepsilon_1 + \chi^{(3)} \left|\vec{E}_N\right|^2$, in which $\chi^{(3)}$ is the third-order nonlinear susceptibility and \vec{E}_N corresponds to the local electric fields in the core for the nonlinear case [5,7]. Thus, Eq. (3) should be revised as given in Box I.

If we define $\chi^{(3)} \left| \vec{E}_{\rm N} \right|^2$ as y, Eq. (6) can be revised to a cubic equation.

$$^{3} + Ly^{2} + My = Nx,$$
 (7)

$$\begin{cases} x = \chi^{(3)} \left| \vec{E}_0 \right|^2 \\ L = \frac{2c_1c_2 + 2d_1d_2}{c_1^2 + d_1^2} \\ M = \frac{c_2^2 + d_2^2}{c_1^2 + d_1^2} \\ N = 16 \frac{|\epsilon_2|^2 |\epsilon_3|^2}{c_1^2 + d_1^2} \end{cases}$$
(8)

$$\begin{cases} c_1 = \operatorname{Re} \left[\epsilon_2 \left(1 + f^2 \right) + \epsilon_3 \left(1 - f^2 \right) \right] \\ c_2 = \operatorname{Re} \left[\epsilon_2 \left(\epsilon_1 + \epsilon_3 \right) \left(1 + f^2 \right) + \left(\epsilon_1 \epsilon_3 + \epsilon_2^2 \right) \left(1 - f^2 \right) \right] \\ d_1 = \operatorname{Im} \left[\epsilon_2 \left(1 + f^2 \right) + \epsilon_3 \left(1 - f^2 \right) \right] \\ d_2 = \operatorname{Im} \left[\epsilon_2 \left(\epsilon_1 + \epsilon_3 \right) \left(1 + f^2 \right) + \left(\epsilon_1 \epsilon_3 + \epsilon_2^2 \right) \left(1 - f^2 \right) \right] \end{cases}$$
(9)

Here, $f = r_1/r_2$. Based on Eq. (7), we could investigate the variations of the nonlinear local field $|\vec{E}_N|$ with the incident field $|\vec{E}_0|$. Throughout this paper, for simplicity, we assume the ACNW is embedded in vacuum. The third-order nonlinear susceptibility of Ag $\chi^{(3)}$ is fixed at about $4.4 \times 10^{-20} \text{ m}^2/\text{V}^2$, as reported in Refs. [7,15,36].

3. Results and discussion

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We first investigate the scattering and local field enhancement of the linear ACNW. In Fig. 2, the solid and dashed lines represent the scattering spectrum and corresponding local field enhancement factor $|E_{\rm L}/E_0|$ of the ACNW, respectively. Here, r_1 and r_2 are fixed at 10 and 13 nm, respectively. The strong couplings between the dipole plasmon resonance of inner Ag nanowire $\left(\omega_{Ag} \Big|_{1} \right)$ and the exciton resonance (ω_{e}) of outer CuCl shell result in two new plexcitonic modes: high-energy $\left(\omega_{Ag}^{e} \Big|_{1}^{h} \right)$ and low-energy mode $\left(\left. \omega_{Ag}^{e} \Big|_{1}^{l} \right)$ [26]. The two strong peaks at about 370.1 and 380.0 nm correspond to the $\omega_{Ag}^{e}\Big|_{1}^{h}$ and $\omega_{Ag}^{e}\Big|_{1}^{l}$, respectively. We also note a weak peak appears at about 375.2 nm that is due to the bare exciton transition in the CuCl shell [26]. The quasistatic theory predicts that the wavelengths of the peaks in the local field enhancement spectrum should match with those in the scattering spectrum [37]. The large local electric fields at two plexcitonic modes are promising for the existence of OB [6-10]. Because there are two peaks in the local field spectrum, we could expect more than one OB region in the nonlinear ACNW [38].

Fig. 3(a) shows the variations of $|E_N|$ in the nonlinear ACNW with $|\vec{E}_0|$ at different wavelengths. Here, $r_1 = 10$ nm and $r_2 = 13$ nm. The solid line represents the variation of $|\vec{E}_N|$ in the nonlinear ACNW at the wavelength of 374.0 nm, which locates in the $\omega_{Ag}^e|_1^h$ mode. With increasing $|\vec{E}_0|$ slowly from zero, $|\vec{E}_N|$ increases along the lower branch. When $|\vec{E}_0|$ exceeds the switch-up threshold (point A), $|\vec{E}_N|$ jumps discontinuously to the higher branch. At this point, the ACNW is driven into resonance and the local fields are sufficiently enhanced

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