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Optical bistability in metal/dielectric composite with interfacial layer

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Abstract

The optical bistability in a composite, composed of nonlinear metallic particles (spherical or cylindrical) with interfacial layers randomly embedded in a linear dielectric host, is investigated theoretically. It is shown that both interfacial property and the size of metallic particles can dramatically affect the optical bistable behavior, and the threshold value is more less in three dimension (3D) than in two dimension (2D) system under same physical condition.

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1. Introduction

Composite materials, consisting of small, nonlinear metallic particles (existing in the shape of sphere or cylinder) randomly embedded in a linear dielectric host, are well known for their complex responses to incident light fields [1,2]. Amusing phenomena, for example, optical bistability, etc., have been predicted theoretically [3,4] and observed experimentally [5]. Their promising applications include optical switches and transistors, pulse shapers, as well as memory elements. Previous theoretical work is of interest, but the effect of the interface separating metallic inclusion from dielectric host, and the interaction between the inclusions were neglected. (As a matter of fact, due to diffusion and roughness, etc., the interfaces between nonlinear inclusions and host medium are not very sharp, and thus there probably exist interfacial layers, physical property of which is different from that of inclusion or host [6,7].) Interfacial effect plays an important role in a variety of systems, and it can dramatically alter the systems' optical behavior. On the other hand, as far as many practical cases are concerned, a composite is often made of inclusion mixtures randomly embedded in a certain host, not in dilute limit. For this case, one must take into account the electrostatic interaction between the inclusions.

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In this paper, the effect of interfacial layer on the bistability is qualitatively studied, by introducing the interfacial factor I [8]. During such consideration, the composite should, in fact, be seen as a three-component system; we may treat it as a two-component system after the analysis of the induced dipole moment of spherical (or cylindrical) particle with an interfacial layer coated. We assume that the inclusions are not too densely packed; according to the Clausius–Mossotti approximation, only dipolar interaction between the inclusions should be taken into account. Also, we find that the inclusion dimension plays an important role in the bistable behavior as well. In addition, we shall study the effect of the size of the inclusion with the shape of sphere (or cylinder), on the bistability.

2. Formalism

2.1. Spherical inclusions

We now consider a single, small, nonlinear metallic sphere with dielectric function $\varepsilon_c(\boldsymbol{E}_c) = \varepsilon_c + \chi_c^{(3)}|\boldsymbol{E}_c|^2$ embedded in a uniform linear dielectric host of dielectric function ε_h. Here, ε_c is the linear part of $\varepsilon_c(\boldsymbol{E}_c)$, it is a complex number having a negative real part and a very small, positive imaginary part, $\chi_c^{(3)}$ is the third-order nonlinear susceptibility. In the limit of the quasistatic approximation, the electrical potential in the system can be written as follows:

$$\phi_c = -C\boldsymbol{E}_0 r\cos\theta, \quad r < a, \tag{1}$$

$$\phi_h = -\boldsymbol{E}_0(r - Br^{-2})\cos\theta, \quad r > a, \tag{2}$$

where $\boldsymbol{E}_0$ is a uniform external electric field, and a is the radius of small metallic particle. B and C can be determined under boundary conditions, and their expressions are

$$B = \frac{\varepsilon_c(\boldsymbol{E}_c) - \varepsilon_h}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h} r_c^3, \tag{3}$$

$$C = \frac{3\varepsilon_h}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h}. \tag{4}$$

The uniform electric field $\boldsymbol{E}_c$ inside the metallic particle is

$$\boldsymbol{E}_c = \frac{3\varepsilon_h \boldsymbol{E}_0}{\varepsilon_c + 2\varepsilon_h}. \tag{5}$$

The induced dipole moment of metallic particle can be given by

$$B\boldsymbol{E}_0 = \frac{\varepsilon_c(\boldsymbol{E}_c) - \varepsilon_h}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h} r_c^3 \boldsymbol{E}_0. \tag{6}$$

Let us add a concentric shell with dielectric function ε_s and thickness t to this metallic particle. Then the electrical potential in the system can be written as follows:

$$\phi_c = -C\boldsymbol{E}_0 r\cos\theta, \quad r < a, \tag{7}$$

$$\phi_s = -\boldsymbol{E}_0(Fr - Gr^{-2})\cos\theta, \quad a < r < a + t, \tag{8}$$

$$\phi_h = -\boldsymbol{E}_0(r - Br^{-2})\cos\theta, \quad r > a + t. \tag{9}$$

The induced dipole moment of coating metallic particle is given by

$$B\boldsymbol{E}_0 = \frac{\rho(2\varepsilon_s + \varepsilon_h)x + (\varepsilon_s - \varepsilon_h)}{2\rho(\varepsilon_s - \varepsilon_h)x + (\varepsilon_s + 2\varepsilon_h)}(a + t)^3 \boldsymbol{E}_0, \tag{10}$$

where $\rho \equiv (a/(a + t))^3$ and

$$x = \frac{\varepsilon_c(\boldsymbol{E}_c) - \varepsilon_s}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_s}. \tag{11}$$

Now, we consider the effect of interfacial layer through the limit, $t \to 0$, while $\varepsilon_s \to \infty$, the interfacial property is concentrated on a surface of zero thickness and only the quantity $t\varepsilon$ is of significance, we take [8]

$$I = \lim_{t \to 0, \varepsilon_s \to \infty} t\varepsilon_s \tag{12}$$

to characterize the interface between the metallic particles and the dielectric host. Here, I is just called the interfacial factor. Such description has been made to discuss the bound on the effective linear thermal conductivity of composite media with imperfect interfaces [8]. A similar discussion on the effective nonlinear optical response of such composite, was considered as well [9,10]. The interfacial layer is, in fact, the mixture of metal and dielectric; strictly speaking, since ε_s is a complex number, I is also a complex quantity. But, the real

part of the dielectric function of metallic particle is always a large negative number, whereas the imaginary part is a small, positive one. Thus, for simplicity, in the limit case, we may neglect the imaginary part. Now, I is only a real number. Crudely speaking, when I is taken as a negative (or positive) value, the interface exhibits metal-like (or dielectric-like); and $I = 0$ corresponds to the perfect interface. Assuming $\eta = \varepsilon_s/(\varepsilon_c(\boldsymbol{E}_c))$ and $\lambda = (a/(a+t))^3$, we obtain again an expression for the induced dipole moment of coating metallic particle,

$$B\boldsymbol{E}_0 = \frac{\tilde{\varepsilon}_c(\boldsymbol{E}_c) - \varepsilon_h}{\tilde{\varepsilon}_c(\boldsymbol{E}_c) + 2\varepsilon_h}(a+t)^3\boldsymbol{E}_0, \tag{13}$$

here, $\tilde{\varepsilon}_c(\boldsymbol{E}_c) = \mu\varepsilon_c(\boldsymbol{E}_c)$, with the factor μ being given by

$$\mu = \frac{(1+2\lambda)\eta + 2(1-\lambda)\eta^2}{(1-\lambda) + (2+\lambda)\eta}. \tag{14}$$

Taking the limit (i.e., Eq. (12)), we obtain

$$\mu = 1 + \frac{2I}{a\varepsilon_c(\boldsymbol{E}_c)}. \tag{15}$$

Under the above limit condition, comparing Eq. (6) with Eq. (13), we find that nonlinear metallic particle with an interfacial layer can be replaced by a solid particle with the radius a and the dielectric function

$$\tilde{\varepsilon}_c(\boldsymbol{E}_c) = \mu\varepsilon_c(\boldsymbol{E}_c) = \varepsilon_c(\boldsymbol{E}_c) + \frac{2I}{a}. \tag{16}$$

The uniform field inside the nonlinear particle is given by

$$\boldsymbol{E}_c = \frac{3\varepsilon_h\boldsymbol{E}_0}{\tilde{\varepsilon}_c(\boldsymbol{E}_c) + 2\varepsilon_h} = \frac{3\varepsilon_h\boldsymbol{E}_0}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h + 2I/a}. \tag{17}$$

Now, we consider a composite, which is made of a non-dilute mixture of nonlinear spherical metallic particles, randomly dispersed in a uniform linear dielectric host. And in the composite, there are interfacial layers between metallic particles and dielectric host. Due to the non-dilute case, the electric field seen by each of the inclusions is not the volume-averaged Maxwell field $\langle\boldsymbol{E}\rangle$ (here $\langle\cdots\rangle$ denotes volume average), which equals external field $\boldsymbol{E}_0$. Assume that the inclusions are not too densely packed, such that only dipolar interactions should be taken into account. Thus, the electric field seen by each of the inclusions should be the volume-averaged Lorentz local field $\langle\boldsymbol{E}_l\rangle$. Therefore, Eq. (17) receives the form

$$\boldsymbol{E}_c = \frac{3\varepsilon_h\langle\boldsymbol{E}_l\rangle}{\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h + 2I/a}. \tag{18}$$

Utilizing the Clausius–Mossotti approximation

$$\boldsymbol{E}_l = \boldsymbol{E} + \tfrac{4}{3}\pi\boldsymbol{P}, \tag{19}$$

hence,

$$\langle\boldsymbol{E}_l\rangle = \boldsymbol{E}_0 + \tfrac{4}{3}\pi\langle\boldsymbol{P}\rangle, \tag{20}$$

where $\langle\boldsymbol{P}\rangle = NB\langle\boldsymbol{E}_l\rangle$. Here, N is the number of metallic particles per unit volume. Taking notice of the volume fraction of metallic particles $f = \frac{4}{3}\pi Na^3$, we have

$$\boldsymbol{E}_c = \frac{3\varepsilon_h\boldsymbol{E}_0}{[\varepsilon_c(\boldsymbol{E}_c) + 2\varepsilon_h + 2I/a](1 - fB/a^3)}. \tag{21}$$

By using of Eqs. (21), (13) and (16), we get

$$y^3 + \alpha y^2 + \beta y + \gamma = 0, \tag{22}$$

where

$$y = \chi_c^{(3)}|\boldsymbol{E}_c|^2, \tag{23}$$

$$\alpha = \frac{2\varepsilon_h(2+f) + 2(\varepsilon_{c_1} + 2I/a)(1-f)}{1-f}, \tag{24}$$

$$\beta = \frac{[\varepsilon_h(2+f) + (\varepsilon_{c_1} + 2I/a)(1-f)]^2 + (1-f)^2\varepsilon_{c_2}^2}{(1-f)^2}, \tag{25}$$

$$\gamma = -\frac{9\varepsilon_h^2\chi_c^{(3)}|\boldsymbol{E}_0|^2}{(1-f)^2}. \tag{26}$$

Here, ε_{c_1} (or ε_{c_2}) is real (or imaginary) part of ε_c. Eq. (22) is a cubic equation in y, and signifies bistability.

2.2. Cylindrical inclusions

Let us consider a composite in which parallel, nonlinear cylindrical metallic particles with interfacial layers randomly dispersed in a linear dielectric host. External electric field $\boldsymbol{E}_0$ is applied,

perpendicular to the cylinder' axis. And there is an interfacial layer separating the cylinder and host medium. Following the section above, we obtain a cubic equation

$$y^3 + \alpha y^2 + \beta y + \gamma = 0, \tag{27}$$

where

$$y = \chi_c^{(3)} |\boldsymbol{E}_c|^2, \tag{28}$$

$$\alpha = \frac{2[(1-f)\varepsilon_{c_1} + (1+f)\varepsilon_h + I/a]}{1-f}, \tag{29}$$

$$\beta = \frac{[(1-f)\varepsilon_{c_1} + (1+f)\varepsilon_h + I/a]^2 + [(1-f)\varepsilon_{c_2}]^2}{(1-f)^2}, \tag{30}$$

$$\gamma = -\frac{4\varepsilon_h^2 \chi_c^{(3)} |\boldsymbol{E}_0|^2}{(1-f)^2}. \tag{31}$$

Here, the symbols represent the same physical meaning as the above. Consequently, the bistable behavior of 2D case can be obtained by solving Eq. (27).

3. Numerical results and discussion

We are now in a position to study the effects of the interfacial factor and the metallic-particle size on optical bistability behavior by solving Eq. (22) (or Eq. (27)) numerically. Choose Au/Al_2O_3 as numerical illustration. We take $\varepsilon_c = -13.67 + 1.04i$ (the incident frequency waves $\omega = 1.88$ ev) [11], $\chi_c^{(3)} \approx 2.8 \times 10^{-8}$, and $\varepsilon_h = 2.89$ [12]. In Fig. 1, the local field $|\boldsymbol{E}_c|^2$ inside spherical metallic particles is plotted as a function of the incident field $\boldsymbol{E}_0$ at $f = 0.03$ and $a = 3$ nm for three different interfacial layers $I = -1, 0, 2$. This plot exhibits bistable response clearly, and the part of the curves with negative slope are unstable. We find that the interfacial layer plays an important role in bistable behavior. It is shown that with the change of I from negative value to positive value, namely, with the transition of the interfacial layer from metallic property to dielectric property, the threshold values (the switch-up and the switch-down) of the bistability decrease. Compared with the case of no interfacial layer, the metal-like interfacial layer makes the threshold values increase, while the dielectric-like interfacial layer makes the threshold values decrease and damps out surface plasmon resonance of metallic component.

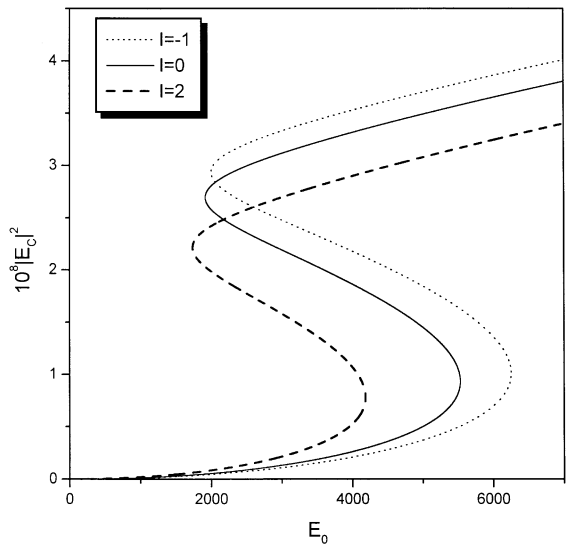

Fig. 1. Bistability curves for interfacial factors $I = -1, 0, 2$ with $\varepsilon_c = -13.67 + 1.04i$, $\chi_c^{(3)} \approx 2.8 \times 10^{-8}$, $\varepsilon_h = 2.89$ and the radius of small Au spheres $a = 3$ nm.

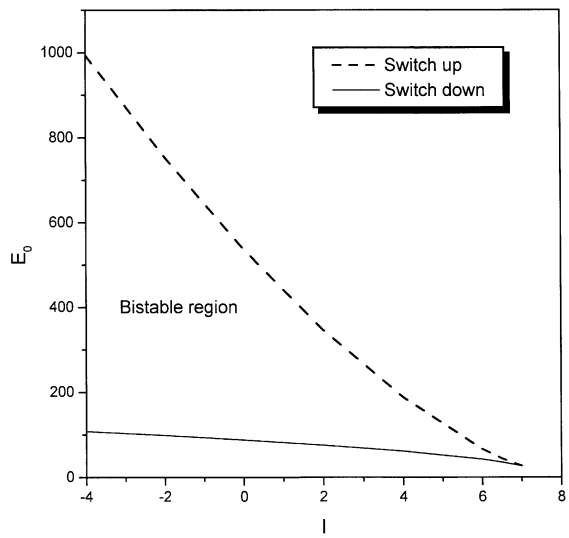

Fig. 2. Switching power versus interfacial factors I. Curve is for $\varepsilon_c = -13.67 + 1.04i$, $\chi_c^{(3)} \approx 2.8 \times 10^{-8}$, $\varepsilon_h = 2.89$ and the radius of small Au spheres $a = 3$ nm.

It is shown in Fig. 2 that, with increasing I, both the threshold values and bistable region are

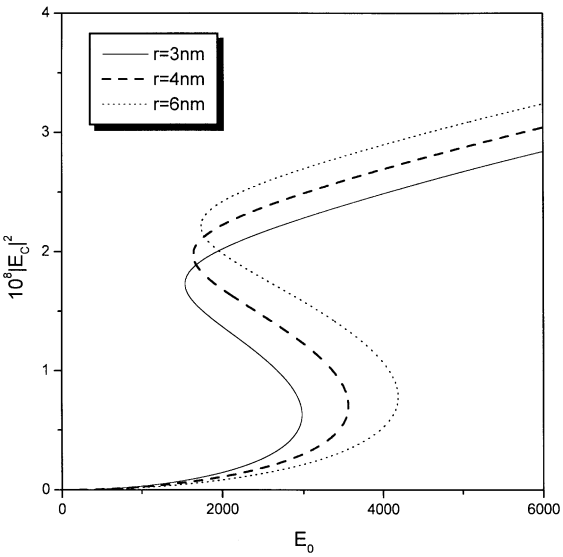

Fig. 3. Bistability curves for the radii of small Au spheres $a = 3$, 4 and 6 nm with $\varepsilon_c = -13.67 + 1.04i$, $\chi_c^{(3)} \approx 2.8 \times 10^{-8}$, $\varepsilon_h = 2.89$ and the interfacial factor $I = 4$.

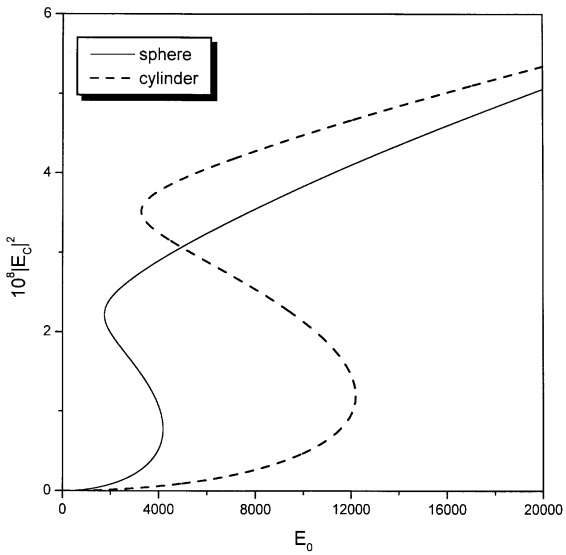

Fig. 4. Bistability curves for spherical inclusions and cylindrical inclusions with $\varepsilon_c = -13.67 + 1.04i$, $\chi_c^{(3)} \approx 2.8 \times 10^{-8}$, $\varepsilon_h = 2.89$, $I = 4$, and the radii of Au spheres and Au cylinders are $a = 3$ nm.

decreased, and the effect of the interfacial factor on the switch-up is more strong than on the switch-down. There exists a critical value of the interfacial factor (I_c); as the interfacial factor is more large than I_c (under present physical parameter, $I_c \approx 7$), the bistability disappears.

In Fig. 3, we plot the bistability curves for $a = 3$, 4 and 6 nm with $I = 4$. We find that the threshold values increase with the increasing radius a. From Fig. 1, we have known that the threshold values will increase for the case of metal-like interfacial layer, as, crudely speaking, means that metallic particle radius increase; hence, as shown in Fig. 3, we can also conclude that threshold values will increase. The result from Fig. 1 is in agreement with Fig. 3.

Fig. 4 shows that bistable behavior in 3D is quite different from that in 2D. For the case of the same physical parameters, the threshold values in 3D are more less than that in 2D, and the switch-up is much high in the latter than in the former. Thus it is also determined that the bistable behavior is dependent on the inclusion dimension.

4. Conclusions

In the present paper, to investigate the bistability of a certain composite containing spherical (or cylindrical) inclusions with interfacial layers, we regard the three-component system as a two-component one. Numerical results show that the interfacial layer and the size of the metallic particles can greatly affect optical bistable behavior, and that the dielectric-like interfacial layer is favorable to reduce the threshold values. Also, smaller the metallic particles chosen lower the threshold values obtained. We also find that the threshold values for 3D case are more low than those for 2D case. It is also determined that the optical bistable behavior is dependent on inclusions' dimension. During the investigation above, the interfacial factor I is only taken as a real number; strictly speaking, it should be a complex number, which may lead to absorption; however, if it is strong enough, we believe that it will prevent the appearance of bistable behavior. Our analysis is helpful to research on bistability devices of composite media. To obtain different bistable characteristics, some new techniques are needed for controlling interfacial property, inclusion's size and dimension.

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References

[1] Y. Hosoya, T. Suga, T. Yanagawa, K. Kurokawa, J. Appl. Phys. 81 (1997) 1475.
[2] R. Serna, J.M. Ballesteros, J. Solis, C.N. Afonso, D.H. Osborne, R.F. Hanglund Jr., A.K. Petford-Long, Thin Solid Films 318 (1998) 96.
[3] K.M. Lenng, Phys. Rev. A 33 (1986) 2461.
[4] D.J. Bergman, O. Levy, D. Stroud, Phys. Rev. B 49 (1994) 129.
[5] R. Neuendort, M. Ouinten, U. Kreibig, J. Chem. Phys. 104 (1996) 22.
[6] J.J. Wang, Z.Y. Li, J. Phys.: Condens. Matter 7 (1995) 6355.
[7] J.J. Wang, Z.Y. Li, Phys. Lett. A 199 (1995) 199.
[8] S. Torquato, M.D. Rintoul, Phys. Rev. Lett. 75 (1995) 4067.
[9] L. Gao, Z.Y. Li, J. Appl. Phys. 87 (2000) 1620.
[10] J.P. Huang, L. Gao, Z.Y. Li, Solid State Commun. 115 (2000) 347.
[11] P.B. Johnson, R.W. Christy, Phys. Rev. B 6 (1972) 4370.
[12] H.B. Liao, R.F. Xiao, J.S. Fu, Y. Kurokawa, J. Appl. Phys. 65 (1997) 673.