Optical Bistability by Surface Resonance Modes

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When a resonance mode is excited in a two-dimensional array of dielectric spheres, the local field intensity is enhanced inside the spheres. This resonance frequency decreases with the increase of the dielectric constant. By use of these two properties, a new mechanism is proposed to realize optical bistability at a low laser power.

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Observation of the optical bistability in semiconductors¹ has made the optical computing system a realistic object, and various architectures have been proposed.² Optical bistability comes from the nonlinear response of the material and is determined by the magnitude of the third-order susceptibility. Therefore, research has been focused on materials with large nonlinearity and short switching time.³ In this Letter, a new mechanism is proposed to reduce the laser power for the optical bistability.

We consider a two-dimensional array of dielectric spheres placed on a flat metal substrate. Our model corresponds not only to colloidal particles⁴ placed on a metal surface but also to microspheres fabricated on a flat surface.⁵ As the electromagnetic (EM) waves suffer multiple scatterings among spheres, the array of spheres show characteristic features of a dielectric slab with periodic modulation. Therefore, it can also be seen as a model for a crossed grating⁷ of dielectric materials coated on a metal surface. In a dielectric slab with dielectric constant larger than 1, there exist localized EM modes. Their dispersion relations lie outside the light cone and hence do not couple with incident waves. When the translational symmetry parallel to the surface is violated by a periodic modulation, however, the localized modes couple with the external field through umklapp processes. Thus, they can be excited by incident waves. We call these coupled modes resonance modes. Our calculation consists of two parts. The first part shows that, when excited, the resonance modes enhance the local field intensity inside the spheres by 2 orders of magnitude. The resonance frequency decreases (increases) with the increase (decrease) of the dielectric constant. In the second part we point out that, if these properties of the resonance modes are used, an optical bistability is realizable at a low laser power.

Maxwell's equations for the EM field of frequency ω give the integral equation for the electric field,

$$
\mathbf{E}(\mathbf{r}) = \mathbf{E}^0(\mathbf{r}) + \sum_{i} \int \tilde{\mathbf{G}}(\mathbf{r}, \mathbf{r}') \cdot v_i(\mathbf{r}') \mathbf{E}(\mathbf{r}') d^3 r', \qquad (1)
$$

where the tensor Green's function is defined by

$$
\tilde{\mathbf{G}}(\mathbf{r}, \mathbf{r}')_{i,j} = [\delta_{i,j} + (c/\omega)^2 \nabla_i \nabla_j] G(\mathbf{r}, \mathbf{r}').
$$
 (2)

 $E^{0}(r)$ is the incident plane-wave field and $G(\mathbf{r}, \mathbf{r}')$ is the usual Green's function. The potential $v_i(r) = [1 - \varepsilon_i(\omega)]$ \times (ω/c)² takes on finite values inside the spheres (i=1) and the substrate $(i=2)$. This expression shows that a material with a larger dielectric constant is equivalent to a more attractive potential, and explains the dielectricconstant dependence of the resonance frequency. Equation (1) is solved exactly for the following model. In the $x-y$ plane, the spheres of radius a are arrayed in a square lattice structure with lattice constant d. The distance from the center of the sphere to the substrate is Z_0 . Then the electric field above the sphere is given by 8

$$
E(r) = E^{0} exp(iq^{-} \cdot r) + R(q) E^{0} exp(iq^{+} \cdot r)
$$

+
$$
\sum_{G} F(G) exp(iq^{+}_{G} \cdot r),
$$
 (3)

where

$$
\mathbf{F}(\mathbf{G}) = (c/2\omega d^2 \gamma_{\mathbf{G}}) [\xi(\mathbf{q}_{\mathbf{G}}^+) + R(\mathbf{q}_{\mathbf{G}}) \xi(\mathbf{q}_{\mathbf{G}}^-)]/(l+1)\mathbf{a},
$$

$$
\mathbf{a} = V\mathbf{a}(i) = (i^{-1} - \Gamma - \tilde{\Gamma})^{-1} [\mathbf{a}(0) + \mathbf{a}(1)].
$$

The superscript $+$ or $-$ of the wave vector q means that its z component is positive (γ_q) or negative $(-\gamma_q)$, and $q_{\mathbf{G}}^+$ (=q⁺+G) is the wave vector shifted by the reciprocal lattice vector **G**. $R(q)$ is a reflectivity tensor of the substrate for an incident field of wave vector q^- . The matrices t, Γ , and $\tilde{\Gamma}$ describe the scattering by the sphere, a structure factor for the scattering between spheres, and a multiple scattering between a sphere and the substrate, respectively. The electric field inside the sphere is expanded by a vector spherical wave with coefficient $\alpha(i)$, and $\alpha(0) = \xi^+(\mathbf{q}^-)\mathbf{E}^0$ and $\alpha(1)$ $=\xi^+(q^+)R(q)E^0$ are the coefficients for the incident and reflected waves, respectively. The factor v relates the amplitude of the external field with that of the internal field. All these quantities are characterized by angular momentum l , magnetic quantum number m , and β (=M,N) which indicates the magnetic and electric components.

The result of the numerical calculation is given in Fig. 1 as a function of frequency which is scaled as $Z = d/\lambda$ with λ the incident wavelength. The parameters are chosen as $a = 0.2 \mu \text{m}$, $d = 0.5 \mu \text{m}$, $Z_0 = d/2$, $\varepsilon_1 = 3$, and the experimental data by Johnson and Christy⁹ are used

FIG. 1. Frequency dependence of the enhancement factor of the electric-field intensity averaged inside the sphere and the absorption spectrum for a two-dimensional array of dielectric spheres placed on a silver substrate.

for the dielectric constants of the silver substrate. The incidence angle is 20' and the incident wave vector lies in the x-z plane. Solid curves are for p polarization and the dotted curves are for s polarization. In the upper half, we show the electric field intensity averaged inside the sphere, I, scaled by the incident intensity I_0 . The absorption spectrum of the EM energy for a silver substrate is given in the lower half, which is calculated by integration of the z component of the Poynting's vector over the x-y plane. For the present parameters, $Z=1$ corresponds to $\hbar \omega = 2.48$ eV, which is smaller than the surface plasmon energy $[\varepsilon_2(\omega) = -1]$ of silver and the flat surface shows an almost complete reflection.

There are two resonance modes in our system, one localized in the dielectric array and the other localized in the metal substrate, i.e., a surface plasmon polariton (SPP). When the resonance modes are excited, evanescent waves from the arrayed spheres excite the SPP modes. The coupling is strong for p polarization as shown clearly in the absorption spectrum. Though modified by the interaction with SPP, the characteristic features of the resonance modes are the same as those of the dielectric array in vacuum, i.e., when excited, the internal intensity is enhanced by 2 orders of magnitude and the resonance frequency decreases when the dielectric constant is increased.

Next, let us introduce a nonlinearity into the dielectric constant of the spheres as

$$
\varepsilon_1(I) = \varepsilon_1(0) + 4\pi \chi^3 I,\tag{4}
$$

where the third-order susceptibility is chosen to be χ^3 = 10⁻⁵ esu. If we fix the frequency at slightly below the resonance, the optical bistability will be realized by the following mechanism. First, consider the intensity enhancement factor I/I_0 as a function of the dielectric constant. On an increase of the dielectric constant, the

FIG. 2. Specular reflectivity (bold lines) and the real part of the nonlinear dielectric constant (thin lines) as functions of the incident power. Bistability is indicated by the arrows. χ^3 $= 10^{-5}$ esu.

resonance frequency decreases and the enhancement factor at the fixed frequency first increases, attains its peak value of the order of 10^2 , and then starts to decrease. On the other hand, the dielectric constant is a function of the internal intensity I and increases linearly with it. Thus, in a certain region of I_0 , these two curves have three crossing points. This indicates the optical bistability. When the real part of χ^3 is negative, bistability is realizable for a frequency slightly above the resonance. The average intensity of the local field is a complicated function of the dielectric constant, and the self-consistent solution is calculated by iteration, i.e., the average intensity obtained by use of the linear dielectric constant $\varepsilon_1(0)$ is used to calculate the first iterated value of the dielectric constant. By use of this value, I calculate the average intensity, which is used for the second iteration of the dielectric constant. This process is repeated until self-consistency is attained. Then, reflectivity is calculated from Eq. (3) for the converged $\varepsilon_1(I)$. The calculation is performed for various values of the incident intensity. In a certain region of I_0 , two stable solutions are obtained. Figure 2 shows the diagram thus obtained for the optical bistability. The third p -polarized resonance mode is used, and the laser frequency is fixed at $Z = 0.87$. Thin curves show the nonlinear dielectric constant versus incident power and the bold curves show the specular reflectivity. The bistability is realized at a low laser power, more than 2 orders of magnitude smaller than that required by a Fabry-Perot resonator of the corresponding material parameters. This reduction of the laser power comes from the sensitive dependence of the resonance frequency on the nonlinear dielectric constant and the large enhancement of the local field. The eflect of the imaginary part of the dielectric constant is examined by putting $Im \varepsilon_1 = 0.01$ and 0.02, respectively. In the former case, the bistability is realized at a low excitation level. If we shift to a slightly smaller frequency Z , the bistability will be realized also in the latter case. Thus we can conclude that the use of the resonance modes of a thin dielectric layer is promising for the realization of the optical bistability at low laser intensities.

In this calculation, I assumed a plane-wave incident field. The effect of the limited cross section of the light beam was investigated by Kaplan and co-workers¹⁰ for the reflection from nonlinear interfaces. In our case, this eftect seems to be of considerable significance because a sharp resonance mode is used for the enhancement.

To realize optical bistability, Wysin, Simon, and $Deck¹¹$ proposed the use of an enhanced local field by the excitation of SPP in the Kretschmann configuration. The mechanism proposed in this Letter is quite different and is based on the enhancement by the excitation of the resonance mode of the nonlinear material itself together with the shift of the resonance frequency which is determined by the local field intensity. Therefore one can use the following features of the resonance modes. First, the resonance-mode frequencies are scaled approximately by the inverse of the lattice constant. Second, for a larger radius of the spheres, the density of resonance modes increases in the energy spectrum. Last, the resonance frequency depends on the incidence angle. Therefore, by controlling the lattice constant and sphere radius, and by changing the incidence angle, one is able to choose a convenient resonance mode. This point is useful for the experimental observation.

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