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Optical bistability driven by the light-induced forces between metal nanoparticles

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A motion-induced optical bistability is shown for a metal nanoparticle dimer that is plasmon coupled and bound with dispersion (colloidal) forces. The effect does not require any material nonlinearity.

The relative changes in the optical responses are important issue should be the light-induced motion of the particles and their optical response, which results in an optical nonlinearity. As we show, this bistability is the reason for the very sharp spectral dependence of the nonlinear absorption from penta-particle aggregates near a certain critical frequency that we revealed recently [17].

The effect manifests itself even for the simplest nanoaggregate—two silver nanoparticles (a dimer) of equal size [see Fig. 1(a)]. The incident light induces a dipole moment $\mathbf{d}_j$ on each particle, and these dipoles obey the well-known coupled-dipole equation (CDE),

$$\mathbf{d}_j = \alpha e \hbar \left( \mathbf{E}_0(r_j) + \frac{3(n \cdot \mathbf{d}_j) \mathbf{d} - \mathbf{d}_j \psi}{\varepsilon \hbar r^3} \right),$$

$$i,j = 1,2, \quad r = r_i - r_j, \quad r = |r|, \quad n = r/r, \quad (1)$$

where the light’s electric field is $\mathbf{E}(r,t) = \mathbf{E}_0(r) \exp(-i \omega t) + c.c., \alpha$ is the dipole polarizability of the particle, and $\varepsilon = 1.78$ is the host medium (ethanol) permittivity. We set $\phi = \psi = 1$, as the dimer is much smaller than the wavelength (see also [17]).

We use $\alpha = \alpha_0 (\varepsilon_m - \varepsilon_b)/(\varepsilon_m + 2\varepsilon_b)$, where the metal permittivity $\varepsilon_m = \varepsilon_n + i \varepsilon_m'$ is given by the Drude formula: $\varepsilon_m = \varepsilon_b - \omega_p^2/\omega(\omega + i \Gamma)$, in which we take $\omega_p = 9.3$ eV and $\Gamma = 0.058$ eV for the frequency and decay rate of the plasma oscillation, respectively, and $\varepsilon_b = 5.3$ to account for the interband electron transi-
tions. The spectrum of the collective plasmon resonance (at low intensity) of the studied aggregate is given in Fig. 1b, for the incident light polarized along the y axis (see also [19]).

The motion of the particles driven by the light-induced forces is described by Newton’s equations

\[ m \ddot{r}_i + \gamma \dot{r}_i = - \partial_i \left[ U_{EM}(r_1, r_2) + U_0(r_1, r_2) \right], \]

where \( \gamma \) is to account for a viscous friction. The overall potential energy consists of two parts. The first is the total potential energy of the dipoles in the incident field

\[ U_{EM} = - \sum_{i=1}^{2} \text{Re} \left[ (\mathbf{d}_i \cdot \mathbf{E}_0(r_i)) - e_h a |\mathbf{E}_0(r_i)|^2 \right], \]

describing either particles repulsion (which occurs at \( \omega = 2.9433 - 3.0015 \text{ eV} \), as computed) or attraction. Here and below we consider \( \mathbf{E}_0 = (0, E_y, 0) \). The other term, \( U_0 \), is responsible for the aggregation forces that bond two particles and, therefore, has no relation to the incident light. We will use the following simple form for \( U_0 \):

\[ U_0 = \eta (1/2 \xi^2 - 3/\xi), \quad \xi = (\Delta a), \quad \Delta = r - 2a, \]

which is adequate for a particle dimer with an equilibrium state. Such an equivalent potential is a result of the balance among dispersion, electrostatic, and steric forces for two metal nanoparticles in colloids [8].

The solution of the equation system [Eqs. (1) and (2)] is basically a dynamic problem. If we set up a temporal dependence of the incident light intensity, we then can find the time-varying particle coordinates, \( \mathbf{r}_i(t) \), and the complex amplitudes of the dipole moments, \( \mathbf{d}_i(t) \). These data allow the calculation of all of the optical characteristics of the aggregate, for instance, the absorbed power \( P(t) \):

\[ (2\omega/\varepsilon_h) |\mathbf{d}_i(t)|^2 \text{Im } \alpha/|\alpha|^2. \]

For excitation radiation with a Gaussian pulse shape and a pulse duration of \( \tau_p = 10^2 \gamma^{-1} \), the frequency dependence of the pulse-averaged nonlinearity of the absorption, \( (P_{NL}) = ((P(t) - P_0)/P_0) \), is shown in Fig. 2 (\( P_0 \) is the linear absorption, i.e., at low intensity). The optical nonlinearity is solely due to the light-induced motion of the nanoparticles, as all material optical constants have been chosen to be linear. The peak incident intensity \( I_0 \) amounts to 17.5 MW/cm\(^2\); this value (denoted as \( I \) below) is chosen to facilitate a comparison with [17,18].

The dependance is very sharp near some critical frequency; such a behavior is quite similar to that reported earlier for a more complex five-particle cluster [17]. Therefore, it is this peculiarity that we focus on in the present paper.

For this purpose, let us examine the nanoaggregate in the light field, with the goal being to determine the intensity-dependent points of the equilibrium interparticle distance. In other words, we consider a static mechanical problem with two states, with and without light (the light is cw in this case).

Actually both \( U_0 \) and \( U_{EM} \) depend only on the interparticle distance, \( r \). For the case of two nanoparticles, the CDE system allows us to find an analytical solution [19], so we can produce an explicit expression for the total potential energy of the irradiated aggregate,

\[ U_{tot}(r) = U_{EM}(r) + U_0(r) \]

\[ = e_h a^3 |\mathbf{E}_0|^2 \text{Re} \left( \frac{4 r^3 x}{2 \pi \alpha^3 - r^3} + 2 x \right) \]

\[ + \frac{I}{2(r - 2a)^2}, \]

where \( x = x' + i x'' = a/\alpha^3 \).

In Fig. 3, the dependence \( U_{tot}(r) \) is plotted for different light frequencies, with \( a^3 |\mathbf{E}_0|^2 = 0.715 \times 10^{-3} \text{ eV} \) (corresponding to the incident intensity \( I_0 = \tilde{I}/3 \)) and \( \eta = 0.1 \text{ eV} \). As we can see, for several frequencies the potential has two minima. In this case the metal nanoaggregate in a light field has two steady states, which means that the system exhibits bistability. The values of \( r \) corresponding to the potential extremes can be found as the roots of the following equation:

\[ (\partial/\partial r) U_{tot}(r) = 0. \]

In Fig. 4, the solutions of Eq. (6) for different light frequencies and intensities are shown. For \( I_0 = \tilde{I}/3 \), there are two bifurcation points, \( \omega = 3.0 \) and 3.1 eV, which define the range where the bistability exists.

In dynamics, the particles start from an initial interparticle distance and tend to reach one of the

![Fig. 2. Pulse-averaged normalized nonlinear absorption of the two-particle aggregate versus the light frequency.](image)

![Fig. 3. Total potential energy versus the distance r between the particles. The curve 5 corresponds to U0(r).](image)
Nevertheless, the bistability still exists because its permittivity. In our case, all permittivities are linear; bistability is caused by the nonlinearity of the material aggregated metal nanoparticles, where the bistability is indicated in Fig. 4 with the open circles. In this case, the corresponding equilibrium interparticle distances are system asymptotically comes to the steady state. The maximal roots in Fig. 4; which root the system tends toward depends on the initial conditions. Thus, for an initial spacing between the particles of \( \Delta_{eq} = (1/3)a \), corresponding to \( r(t=0) = (7/3)a \), the particles “choose” the lower root. The direct numeric integration of Eq. (2) gives a proof of this statement. For the steplike input intensity \( I_0(t \geq 0) = \bar{I}/3 \), the time evolution was computed for each light frequency until the system asymptotically comes to the steady state. The corresponding equilibrium interparticle distances are indicated in Fig. 4 with the open circles. In this case, the lower bifurcation point (i.e., \( \omega = 3.0 \text{ eV} \)) appears as the sharp difference in the nonlinear absorption spectrum (as is shown in Fig. 2). Note that other nonlinear optical effects caused by light-induced particle dynamics, such as, say, nonlinear scattering, should exhibit an analogous peculiarity in the frequency dependence.

On the other hand, if the particles start from a relatively large spacing [for instance, \( r(t=0) > 4a \)], the final state of the aggregate will be governed by the largest root of Eq. (6). In this case the corresponding bifurcation point (\( \omega = 3.1 \text{ eV} \)) would manifest itself in the optical response, while the other would be “invisible.”

The optical bistability described above is in close relation to that studied earlier in third-order nonlinear systems, like thin layer [20], single [21], and aggregated [22] metal nanoparticles, where the bistability is caused by the nonlinearity of the material permittivity. In our case, all permittivities are linear; nevertheless, the bistability still exists because its general origin is the intensity-dependent enhancement factor for the local field.

Along with the response shown in Fig. 2, the bistability of the system also implies a critical nonlinearity dependence on the initial interparticle spacing at frequencies near the bifurcation point. A technique can be suggested to optically distinguish two dimers that just slightly differ in their initial spacing via probing the nonlinear absorption (or scattering).

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