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Two optical bistability domains in composites of metal nanoparticles with nonlinear dielectric core

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ABSTRACT

It is shown that the local field in metal spherical particles with a dielectric core in an external varying electric field has two maxima at two different frequencies. The second maximum becomes more important with an increment in the metal fraction. Due to the nonlinear dielectric function of the core, the composite of these inclusions may have two optically induced bistability domains at different frequencies. At rather high metal fraction, two bistability domains merge and form one entire bistability domain. The parameters of these domains are studied numerically. The paper focuses on the second bistability domain, which has not been discussed in the literature so far. This domain exists in a comparatively narrow frequency range and its onset fields are lower than those of the first bistability domain. The lowest bistability onset fields are obtained in the entire domain. This peculiarity of the optical induced bistability in the metal composite with small dielectric cores can be attractive for possible applications.

1. Introduction

Optical induced bistability (OIB) is still of great interest since its theoretical discovery and experimental realization [1–3] because of its numerous possible applications. The most attractive systems for the study of OIB are composites consisting of metal covered dielectric particles with nonlinear dielectric functions embedded in a linear host matrix [4,5]. The point is that OIB is based on the nonlinear properties of the dielectric function of inclusions. The amplitude of the electric field of the incident intensive radiation is enhanced in the inclusion (local field) on the frequency close to the surface plasmon frequency of the metal. This enhancement makes the nonlinear part of the dielectric function (DF) of the core significant.

Usually, OIB is demonstrated with the help of *S*-like curves in the plane "applied field - local field (in an inclusion)" [4–6]. They show that three values of the local field correspond to one value of the applied field which implies the system to be unstable. These curves allow one to specify the onset and offset values of the applied fields of OIB at a particular frequency of the incident radiation.

In this paper, we show that the enhancement factor of the local field of metal nanoinclusion with a dielectric core has two maxima at different frequencies. The second maximum is important at comparatively high metal fractions. The composite with these inclusions has two bistability domains in different frequency ranges. In our previous paper [7], we proposed the analytic formulas specifying the OIB domain in the plane of "frequency of the incident radiation - modulus squared of the amplitude of incident radiation". This method can be considered as a complementary to the above mentioned method of *S*-like curves. With the help of this method, the present paper studies two domains of bistability in the composite of spherical metal inclusions with "small" nonlinear dielectric core focusing on the second bistability domain. With increment in the metal fraction, the two bistability domains merge into one with comparatively low onset fields. The second bistability domain and the peculiarities of such composites have not been discussed in the literature so far.

This paper is organized as follows. In Section 2, we analyze the enhancement factor of the local field in the metal nanoparticles with dielectric core for different metal fractions. Section 3 is dedicated to the numerical analysis of the two bistability domains in the composite with these inclusions. In Section 4, we summarize the main results of the paper.

2. Resonant frequencies and enhancement factor of local field in metal covered spherical inclusion

The distribution of the potential in a spherical metal nanoparticle with a dielectric core embedded in a dielectric matrix in an external constant electric field, can be written as follows [8]

$\Phi_1 = -E_h A r \cos\theta, \quad r \le r_1,$

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$$\Phi_2 = -E_h(Br - C/r^2)\cos\theta, \quad r_1 \le r \le r_2,$$
$$\Phi_h = -E_h(r - D/r^2)\cos\theta, \quad r \ge r_2. \tag{1}$$

Here Φ_1,Φ_2, and Φ_h are potentials in the dielectric core, metal, and the host matrix, respectively, E_h is the applied field, r and θ are the spherical coordinates of the observation point (the z-axis is chosen along the vector $\mathbf{E}_h$), r_1,r_2 are radiuses of the dielectric core and the metal shell, respectively. A,B,C,D are unknown coefficients.

From the continuity conditions of the potential and the displacement vector at the boundaries dielectric core–metal and metal–host matrix, we obtain a system of linear algebraic equations for A, B, C, D. The solution of this system can be presented as

$$A = \frac{9\varepsilon_2\varepsilon_h}{2p\Delta}, \tag{2}$$

$$B = \frac{3\varepsilon_h(\varepsilon_1 + 2\varepsilon_2)}{2p\Delta}, \tag{3}$$

$$C = \frac{3\varepsilon_h(\varepsilon_1 - \varepsilon_2)}{2p\Delta} r_1^3, \tag{4}$$

$$D = \left\{1 - \frac{3\varepsilon_h[(3-p)\varepsilon_2 + p\varepsilon_1]}{2p\Delta}\right\} r_2^3, \tag{5}$$

where

$$\Delta = \varepsilon_2^2 + q\varepsilon_2 + \varepsilon_1\varepsilon_h. \tag{6}$$

Here $q = (3/2p - 1)\varepsilon_1 + (3/p - 1)\varepsilon_h$, $p = 1 - (r_1/r_2)^3$ is a metal fraction in the inclusion, ε_1,ε_2, and ε_h are the dielectric functions (DFs) of the core, metal shell, and the host matrix, respectively. We note that Eq. (1) describes the electric field of the electromagnetic wave in the electrostatic approximation when the wave length of the incident radiation is much larger than a typical size of the inclusion [9]. In particular, Eq. (5) coincides with the polarizability of a coated sphere, which follows from the general expression of a coated ellipsoid [9].

Next we choose the DF of a metal in the inclusion to be in the Drude form

$$\varepsilon_2 = \varepsilon_\infty - \frac{1}{z(z + i\gamma)}. \tag{7}$$

Here we introduced dimensionless frequencies $z = \omega/\omega_p$ and $\gamma = v/\omega_p$ (ω is the incident radiation frequency while ω_p is the frequency of the metal shell, and v is the electron collision frequency). The DF of the inclusion core is

$$\varepsilon_1 = \varepsilon_{10} + \chi|\mathbf{E}|^2, \tag{8}$$

where ε_{10} is the linear part of DF, χ is the nonlinear Kerr coefficient, and $\mathbf{E}$ is the local field in the core.

Now we consider the enhancement factor of the local field in the inclusion. For weak incident fields $\chi|\mathbf{E}|^2 \ll \varepsilon_{10}$, the local field is presented as $\mathbf{E} = A\mathbf{E}_h$. In this relation A is given by Eq. (2) and is a complex quantity. Further, it would be convenient to deal with $|A|^2$, which is a real quantity. We call $|A|^2$ the enhancement factor and express it as

$$|A|^2 = \frac{81\varepsilon_h^2}{4p^2} \frac{\varepsilon_2'^2 + \varepsilon_2''^2}{[\varepsilon_2'^2 - \varepsilon_2''^2 + q\varepsilon_2' + \varepsilon_1\varepsilon_h]^2 + \varepsilon_2''^2(q + 2\varepsilon_2')^2}. \tag{9}$$

Here ε_2' and ε_2'' are the real and imaginary parts of ε_2 (7), respectively. For the sake of simplicity, we ignore the imaginary parts of ε_1 and ε_h.

Let us consider an ideal case when the decay of plasma vibration is extremely small ($\gamma \ll 1$). In this case, the second term in the denominator of Eq. (9) proportional to $\varepsilon_2''^2 \sim \gamma^2$ is negligible. The maximum of $|A|^2$ is reached when the first term in the denominator of Eq. (9) is zero. This condition gives a quadratic equation with respect to ε_2', which has two roots with two different resonant frequencies. For not very small γ and arbitrary p, the situation can be traced only numerically.

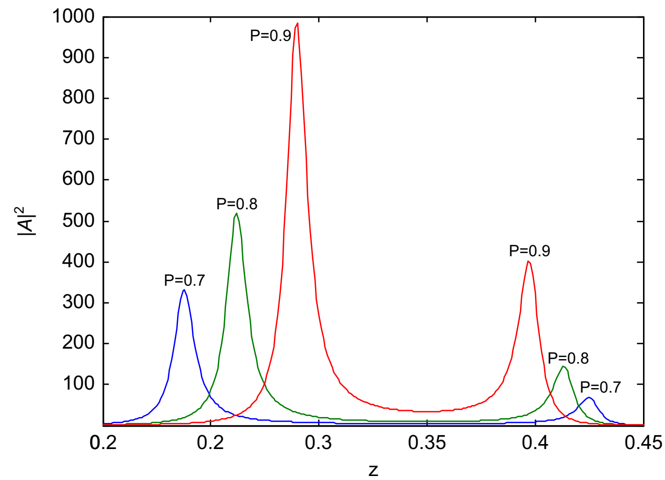

Fig. 1. Enhancement factor $|A|^2$ versus z of metal inclusion with a dielectric core at different p; $\varepsilon_\infty = 4.5$, $\varepsilon_1 = 6$, $\varepsilon_h = 2.25$; $\omega_p = 1.46 \times 10^{16}$ (frequency of the silver surface plasmons), $v = 1.68 \times 10^{14}$,$\gamma = 1.15 \times 10^{-2}$.

Fig. 1 shows $|A|^2$ versus z for different p. These curves are obtained with the help of Eq. (9) neglecting the nonlinear term in ε_1 given by Eq. (8). The numerical values of the DFs of the composite are taken from Ref. [7].

The most interesting feature of these graphs is the appearance of two peaks of the enhancement factor at two different frequencies. The positions and values of these maxima strongly depend on p (for fixed other parameters). For example, for $p < 0.4$, the second maximum is lower then the first one and very small. May be because of this the second maximum of $|A|^2$ was not specified earlier in the papers devoted to the study of OIB in coated particles [4,5,7]. For $p > 0.4$, it becomes more important and with further increase in p, both maxima become higher and move closer to each other.

It is necessary to note that p should not be very close to unity, so that ε_1 should be physically meaningful. However, "closeness" of p to unity does not mean that the radius of dielectric core r_1 is close to 0. The point is that r_1 is related to a radius of the inclusion r_2 as

$$r_1/r_2 = (1-p)^{1/3}. \tag{10}$$

For example, for inclusions with p=0.9 it gives $r_1 = 0.46r_2$ and even for p=0.999, we get $r_1 = 0.1r_2$.

One more parameter that strongly affects the enhancement factor is ε_∞. For example, using $\varepsilon_\infty = 1$ [10] with identical other dielectric constants, we obtain the second maximum of $|A|^2$ to be larger than the first one. At the same time, the width of the maxima get narrower and the corresponding resonant frequencies show blue shift. This is illustrated in Fig. 2.

Lastly, we emphasize that the second maximum in $|A|^2$ becomes important in inclusions with "large" fraction of metal that exceeds the fraction of the dielectric. In this paper, we are exploring the OIB in composites of metal inclusions with small dielectric cores which has not been done before. It is worth noting that appearance of two maxima in the enhancement factor of covered inclusions requires special conditions. It can be shown with the help of Eq. (9) that the metal inclusions covered by the dielectric, used in Ref. [11], have only one resonant frequency and one maximum of the enhancement factor.