Nonlinear optical and magneto-optical effects in non-spherical magnetic granular composite

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The magnetization-induced nonlinear optical and nonlinear magneto-optical properties in a magnetic metal-insulator composite are studied based on a tensor effective medium approximation with shape factor and Taylor-expansion method. There is a weakly nonlinear relation between electric displacement **D** and electric field **E** in the composite. The results of our studies on the effective dielectric tensor and the nonlinear susceptibility tensor in a magnetic nanocomposite are surveyed. It is shown that such a metal-insulator composite exhibits the enhancements of optical and magneto-optical nonlinearity. The frequencies at which the enhancements occur, and the amplitude of the enhancement factors depend on the concentration and shape of the magnetic grains.

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Nonlinear magneto-optical (NMO) response for magnetic nanoparticles is a new expanding field. Fundamental importance and potential technological applications greatly motivate experimental as well as theoretical studies. Experimental studies of nonlinear optical (NO) and NMO effects trace back to 1990s[1−3]. The transition of experimentalist's attention to the granular films with random distribution of nanoparticles opens a new chapter in NMO studies[4−7].

We consider magnetic metal-insulator systems which reveal unusual linear and NO and NMO effects. NO and NMO response can be observed at the frequency of incident light for a granular insulator-magnetic metal composite in which the constituent is described by a weakly cubic nonlinear relation between the electric displacement **D** and electric field **E**, $\mathbf{D}_i = \left(\varepsilon_i^{(0)} + \chi_i^{(3)}|\mathbf{E}_i|^2\right)\mathbf{E}_i$. Its linear dielectric function and nonlinear susceptibility are a complex, frequency-dependent tensor. The NMO response is determined by the off-diagonal as well as the diagonal elements of the tensor. For such a system one can obtain the NO and NMO enhancements[8].

In actual systems the granular constituents are usually not spherical in shape and even are shape distributed. The size and shape of the embedded metal particles can be determined with transmission electron microscopy (TEM). Before and after annealing, the reshaping of the inclusions takes place. How does the particle shape affect the NO and NMO response? The influence of particle shape modifying the optical and magneto-optical nonlinearity has not drawn much attention. Little theoretical work has been devoted to the particle shape effects on the NO and NMO properties. In this paper, by introducing the particle shape factor, we would like to deal with the effects of particle shape on the NO and NMO enhancements. Investigating our results, we show that the frequencies, at which the NO and NMO enhancements occur; and the magnitude of enhancements depend on the shape of the particles. Numerical calculations of a granular insulator-magnetic metal composite in the tensor effective media approximation (TEMA) and Taylor-expansion method make it possible to estimate the dependence of NO and NMO properties on the concentration and shape of magnetic particles. The desired NO and NMO properties can be obtained by suitable adjustment of the inclusion's shape.

We are discussing nonlinear magneto-optics in a granular metal-insulator composite at the frequency of incident light in which each component is described by a weakly cubic nonlinear relation between **D** and **E** of the form

$$\mathbf{D}_i = \left(\varepsilon_i^{(0)} + \chi_i^{(3)}|\mathbf{E}_i|^2\right)\mathbf{E}_i, \quad (i = 0, 1), \tag{1}$$

where $\varepsilon_i^{(0)}$ and $\chi_i^{(3)}$ are the tensor linear dielectric function and the third order nonlinear susceptibility of the ith component, i indcates metal or insulator. The magnitude of the nonlinear term $\chi_i^{(3)}|\mathbf{E}_i|^2$ is smaller than that of the linear part $\varepsilon_i^{(0)}$ (i.e. $\chi_i^{(3)}|\mathbf{E}_i|^2 \ll \varepsilon_i^{(0)}$). The linear dielectric tensor $\varepsilon_i^{(0)}$ and the third order nonlinear susceptibility $\chi_i^{(3)}$ are assumed to have the form[8,9]

$$\varepsilon_i^{(0)} = \begin{pmatrix} \varepsilon_i^{\mathrm{d}} & -\varepsilon_i^{\mathrm{od}} & 0 \\ \varepsilon_i^{\mathrm{od}} & \varepsilon_i^{\mathrm{d}} & 0 \\ 0 & 0 & \varepsilon_i^{\mathrm{d}} \end{pmatrix},$$

$$\chi_i^{(3)} = \begin{pmatrix} \chi_i^{\mathrm{d}} & -\chi_i^{\mathrm{od}} & 0 \\ \chi_i^{\mathrm{od}} & \chi_i^{\mathrm{d}} & 0 \\ 0 & 0 & \chi_i^{\mathrm{d}} \end{pmatrix}. \tag{2}$$

It is convenient to assume that all the diagonal elements remain equal. The off-diagonal elements are responsible for the magneto-optical (MO) response. The effective linear dielectric function $\varepsilon_{\mathrm{e}}^{(0)}$ and nonlinear response $\chi_{\mathrm{e}}^{(3)}$ of the whole system have the form

$$\langle \mathbf{D} \rangle = \left(\varepsilon_{\mathrm{e}}^{(0)} + \chi_{\mathrm{e}}^{(3)}\left\langle |\mathbf{E}|^2 \right\rangle\right)\langle \mathbf{E} \rangle, \tag{3}$$

where $\langle \mathbf{D} \rangle$ and $\langle \mathbf{E} \rangle$ are the spatial average of the electric displacement and the electric field, respectively.

Our goal is to study the effective linear and nonlinear response of the composite. In a binary composite, the diagonal and off-diagonal elements of the effective dielectric function can be written in the form of[8,10]

$$\varepsilon_{\rm e}^{\rm d} = F(\varepsilon_1^{\rm d}, \varepsilon_2^{\rm d}, f), \tag{4}$$

and

$$\varepsilon_{\rm e}^{\rm od} = \Psi(\varepsilon_1^{\rm d}, \varepsilon_2^{\rm d}, \varepsilon_1^{\rm od}, \varepsilon_2^{\rm od}, f), \tag{5}$$

where f is the volume fraction of ε_1 component and F, Ψ are the functions dependent on the geometry of the composite. In Eq. (1), the magnitude of the dielectric function is the sum of the linear part and the nonlinear one, i.e., $\varepsilon_i = \varepsilon_i^{(0)} + \chi_i^{(3)} \langle |\mathbf{E}_i|^2 \rangle$, and $\langle |\mathbf{E}_i|^2 \rangle$ is the mean square of the electric field in the ith component in the linear limit. In order to obtain the approximation for $\chi_{\rm e}^{(3)}$, we expand the functions F and Ψ in a Taylor series about $\varepsilon_{\rm e}^{\rm d(0)}$ and $\varepsilon_{\rm e}^{od(0)}$[10]

$$\varepsilon_{\rm e}^{\rm d} \approx F(\varepsilon_1^{\rm d(0)}, \varepsilon_2^{\rm d(0)}, f) + \sum_{i=1}^{2} F_i'(\varepsilon_1^{\rm d(0)}, \varepsilon_2^{\rm d(0)}, f)\chi_i^{\rm d} \langle |\mathbf{E}_i|^2 \rangle, \tag{6}$$

$$\begin{aligned}\varepsilon_{\rm e}^{\rm od} \approx{}& \Psi(\varepsilon_1^{\rm d(0)}, \varepsilon_2^{\rm d(0)}, \varepsilon_1^{\rm od(0)}, \varepsilon_2^{\rm od(0)}, f) \\ &+ \sum_{i=1}^{2} \Psi_i'(\varepsilon_1^{\rm d(0)}, \varepsilon_2^{\rm d(0)}, \varepsilon_1^{\rm od(0)}, \varepsilon_2^{\rm od(0)}, f)\chi_i^{\rm d} \langle |\mathbf{E}_i|^2 \rangle \\ &+ \sum_{i=1}^{2} \widetilde{\Psi_i'}(\varepsilon_1^{\rm d(0)}, \varepsilon_2^{\rm d(0)}, \varepsilon_1^{\rm od(0)}, \varepsilon_2^{\rm od(0)}, f)\chi_i^{\rm od} \langle |\mathbf{E}_i|^2 \rangle,\end{aligned} \tag{7}$$

where

$$\begin{cases} F_i' = \frac{\partial F}{\partial \varepsilon_i^{\rm d(0)}} \\ \Psi_i' = \frac{\partial \Psi}{\partial \varepsilon_i^{\rm d(0)}} \\ \widetilde{\Psi_i'} = \frac{\partial \Psi}{\partial \varepsilon_i^{\rm od(0)}} \end{cases}. \tag{8}$$

In the linear limit, we have the relation of[10]

$$\frac{f_i \langle |\mathbf{E}_i|^2 \rangle}{\mathbf{E}_0^2} = \frac{\partial \varepsilon_{\rm e}^{\rm d(0)}}{\partial \varepsilon_1^{\rm d(0)}} \equiv F_i'(\varepsilon_1^{\rm d(0)}, \varepsilon_1^{\rm d(0)}, f) = F_i'. \tag{9}$$

The diagonal and off-diagonal elements of the dielectric function of the composite can also be expressed as

$$\varepsilon_{\rm e}^{\rm d} = \varepsilon_{\rm e}^{\rm d(0)} + \chi_{\rm e}^{\rm d}\mathbf{E}_0^2, \qquad \varepsilon_{\rm e}^{\rm od} = \varepsilon_{\rm e}^{\rm od(0)} + \chi_{\rm e}^{\rm od}\mathbf{E}_0^2, \tag{10}$$

where $\mathbf{E}_0$ is the external field. Therefore, we have[8]

$$\chi_{\rm e}^{\rm d} = fK_1\chi_1^{\rm d} + (1-f)K_2\chi_2^{\rm d}, \tag{11}$$

and

$$\chi_{\rm e}^{\rm od} = fQ_1\chi_1^{\rm d} + (1-f)Q_2\chi_2^{\rm d} + fM_1\chi_1^{\rm od} + (1-f)M_2\chi_2^{\rm od}, \tag{12}$$

where

$$K_i = \frac{1}{f_i^2}F_i'|F_i'|, \tag{13}$$

$$Q_i = \frac{1}{f_i^2}\Psi_i'|F_i'|, \tag{14}$$

$$M_i = \frac{1}{f_i^2}\widetilde{\Psi_i'}|F_i'|, \tag{15}$$

$\chi_{\rm e}^{\rm d}$ describes the third NO response, and $\chi_{\rm e}^{\rm od}$ describes NMO response.

We consider two-component metal-insulator granular composite containing a volume fraction $(1-f)$ of insulator and fraction f of magnetic particles. For simplicity we assume that only the magnetic component is nonlinear. The magnetic component has a frequency-dependent dielectric tensor written in the form of Eq. (6). The dielectric function of the non-magnetic component is assumed to be an isotropic tensor,

$$\varepsilon_2 = \begin{pmatrix} \varepsilon_0 & 0 & 0 \\ 0 & \varepsilon_0 & 0 \\ 0 & 0 & \varepsilon_0 \end{pmatrix}. \tag{16}$$

As in a realistic composite system, the shape of inclusions is far from being spherical. For simplicity we consider such a metal-insulator granular composite where the inclusions are rotational ellipsoids. We also assume the inclusions are all identically aligned and their symmetry axes parallel to the z axis. By taking into account the particles' shape, the geometry-dependent TEMA satisfies the form[9]

$$f\frac{\varepsilon_1^{\rm d(0)} - \varepsilon_{\rm e}^{\rm d(0)}}{\varepsilon_{\rm e}^{\rm d(0)} + \frac{1}{2}(1-L)(\varepsilon_1^{\rm d(0)} - \varepsilon_{\rm e}^{\rm d(0)})} + (1-f)\frac{\varepsilon_0 - \varepsilon_{\rm e}^{\rm d(0)}}{\varepsilon_{\rm e}^{\rm d(0)} + \frac{1}{2}(1-L)(\varepsilon_0 - \varepsilon_{\rm e}^{\rm d(0)})} = 0, \tag{17}$$

and

$$f\frac{\varepsilon_{\rm e}^{\rm od(0)} - \varepsilon_1^{\rm od(0)}}{[\varepsilon_{\rm e}^{\rm d(0)} + \frac{1}{2}(1-L)(\varepsilon_1^{\rm d(0)} - \varepsilon_{\rm e}^{\rm d(0)})]^2} + (1-f)\frac{\varepsilon_{\rm e}^{\rm od(0)}}{[\varepsilon_{\rm e}^{\rm d(0)} + \frac{1}{2}(1-L)(\varepsilon_0 - \varepsilon_{\rm e}^{\rm d(0)})]^2} = 0, \tag{18}$$

where L is the depolarization factor of an ellipsoid whose principal axis is parallel to the z axis. For spheres, $L = \frac{1}{3}$. Thus, we obtain the effective dielectric tensor from above equations.

Using the geometry-dependent TEMA, we carry out calculations about the effect of magnetic particle shape on the NO and NMO response. The coefficients K, Q and M in Eqs. (13)–(15) are NO and NMO enhancement factors.

The frequency dependence of K, Q, and M are shown in Fig. 1. These nonlinearity enhancement factors have resonance peaks at some characteristic frequencies for different filling fractions of magnetic particles. Our calculations show that the characteristic frequencies are the

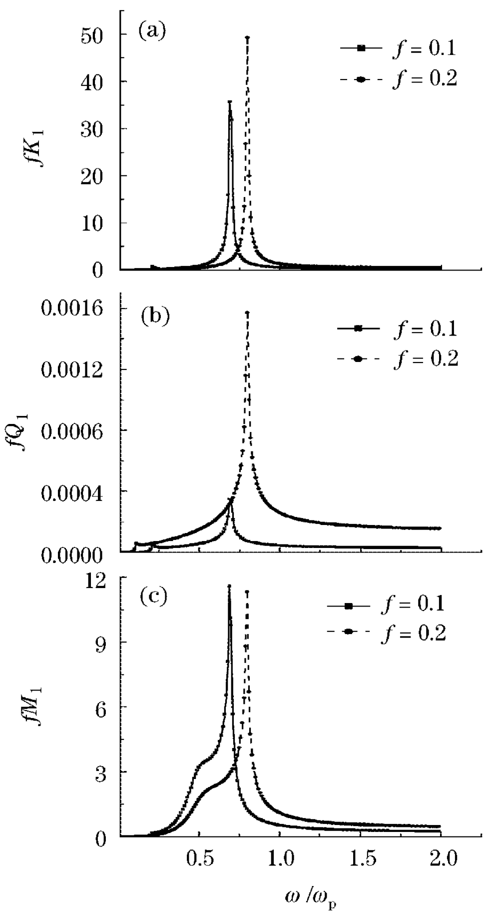

Fig. 1. NO and NMO enhancement factors fK_1, fQ_1, and fM_1 versus ω/ω_p for a composite at $L=\frac{1}{3}$ with $f=0.1$ and $f=0.2$. Optical and magneto-optical parameters were taken from Refs. [9,11].

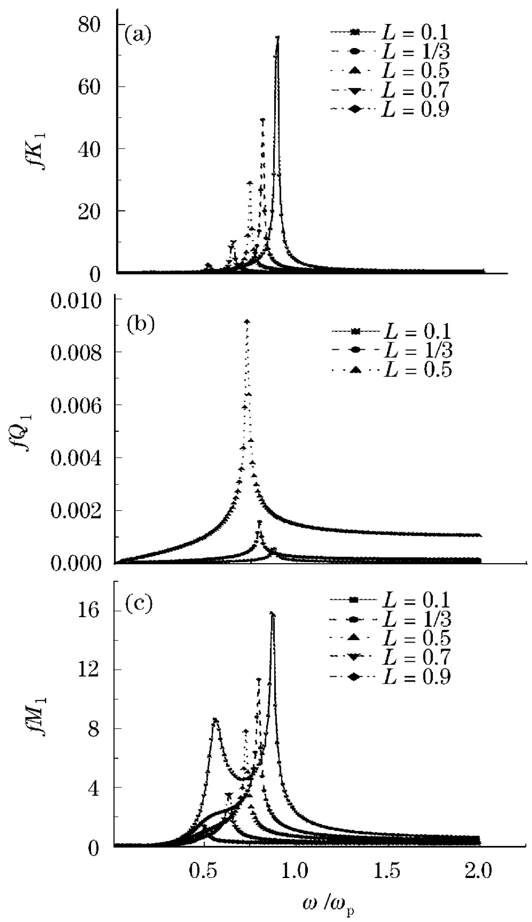

Fig. 2. NO and NMO enhancement factors fK_1, fQ_1, and fM_1 versus ω/ω_p at $f=0.2$ with various L for the same parameters as in Fig. 1.

same for both NO (K) and NMO (Q, M) peaks related to the surface plasmon resonance of the metal particles. Near the plasma frequency ω_p of metal, the magnitude of optical and magneto-optical nonlinearity reaches a maximum. At higher concentration of metal, the enhancement peaks exhibit blue-shift. The resonance optical peak (K) is enhanced with increasing the filling fraction of magnetic metal particles, so is Q. As to M, the magnitude of the peak decreases slowly with the increase of f.

The dependence of K, Q, and M on the shapes of the metallic particles is shown in Fig. 2 at a filling fraction $f=0.2$ for some different shape factors from $L=0.1$ (needlelike particles) to $L=0.9$ (platelike particles). The frequency at which the enhancement peaks occur depends on the particle shape. One can see that the needlelike particles lead to the strongest enhancement of K and M, but the weakest peak of Q. Of the particle shapes considered, the enhancement peaks shift to lower frequency with the increase of the shape factor L.

From Eq. (12) it is easy to observe that as long as one component exhibits nonlinear optics, a composite also presents NMO response even when both components have no magneto-optical nonlinearity, i.e., $\chi_i^{\mathrm{od}}=0$. Figure 3 shows the enhancement factors for optical and magneto-optical nonlinearity versus the filling fraction of magnetic metal particles f for different shape factors L. The enhancement peaks depend on the particle shape significantly. The greatest enhancement is observed for needlelike particles. With the shape factor increasing, the enhancement peaks occur at higher fraction.

In summary, fundamental importance and potential application of a new generation of magnetic tensors and storage media make NMO effects particularly attractive. In this paper, we discuss the effective NO and NMO properties in a magnetic metal-insulator granular composite. To gain an insight into how the particle shape influences the optical and magneto-optical nonlinearity, we make calculations of the NO and NMO factors based on a TEMA and Taylor-expansion method. By introducing the shape factor L of granular particles, we find that both the optical and magneto-optical nonlinearity are highly sensitive to the shape factors and thus may provide the freedom to maximize the optical and magneto-optical nonlinearity enhancements and to adjust the resonant frequencies where the enhancements occur.

Note that the present results are applicable based on the assumption that all the inclusions identically aligned have the same dielectric tensor and ellipsoid shape. Our work can be extended to study the realistic

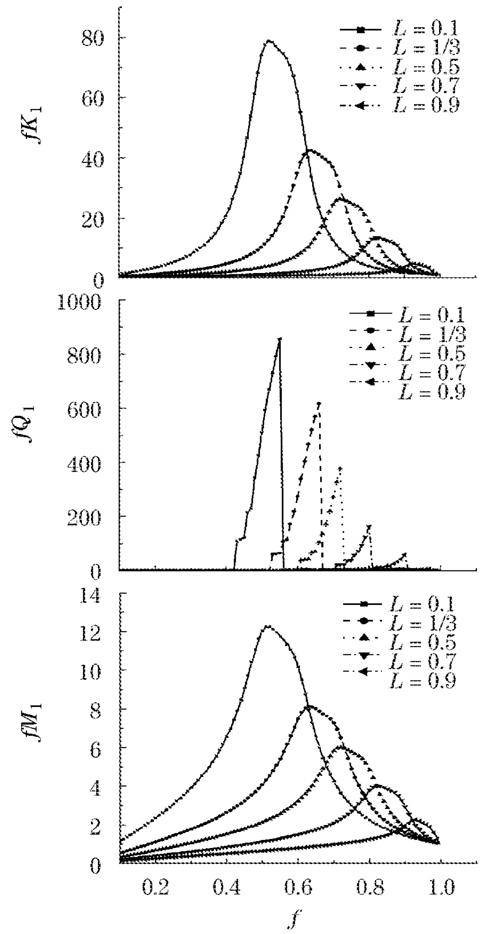

Fig. 3. NO and NMO enhancement factors fK_1, fQ_1, and fM_1 versus f with various L for the same parameters as in Fig. 1.

situation such as the shape-distributed granular composite. The calculations will further estimate experimental study on the magneto-optical nonlinearity of a magnetic granular composite.

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