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Silver-coated metallic and dielectric magnetic nanospheres: Localized surface plasmons and circular dichroism

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ABSTRACT

A thorough study of localized surface plasmons and associated strong circular dichroism, which can occur in silver-coated metallic and dielectric magnetic nanospheres, is reported by means of both quasistatic and full electrodynamic calculations taking into account the actual (magneto) optical response of the constituent materials, including dispersion and losses. It is shown that such composite magnetoplasmonic nanoparticles offer a versatile platform for engineering hybrid plasmon modes that give rise to sharp absorption resonances and subject to large magneto-optic splitting, leading to giant magnetic circular dichroism signals, by properly choosing the different materials and tuning the geometrical parameters involved.

1. Introduction

Nanostructures comprising magnetic materials exhibit unique optical properties, and strong photon confinement in the vicinity of the magnetically active components results in large enhancement of linear and nonlinear magneto-optical effects [1]. In this respect, localized modes of the electromagnetic (EM) field occurring, e.g., at a noble metal–dielectric interface, termed surface plasmons, provide an efficient means to confine light in subwavelength volumes and achieve huge local field enhancement at specific frequencies that can be tuned within a relatively broad spectrum by changing the materials and geometrical parameters involved [2]. Unfortunately, though noble metals, like gold and silver, are excellent plasmonic materials, they show very weak magneto-optical activity at moderate magnetic fields. For example, it has been reported that magnetoplasmonic modes, observed on colloidal gold nanoparticles by means of magnetic circular dichroism spectroscopy, could be useful for detecting changes in the refractive index of the surrounding medium and thus developing refractometric sensing applications but the required magnetic fields are prohibitively strong [3]. On the other hand, ferromagnetic metals that exhibit strong magneto-optical effects lack competitive plasmonic properties due to the high optical losses. In this respect, combining magnetic, either metallic or dielectric, materials with noble metals opens new routes in the development of efficient and versatile, the so-called magnetoplasmonic, architectures [4]. For example, large enhancement of the magneto-optical Faraday rotation has been observed in all-metal core–shell cobalt–silver nanoparticles due to localized surface plasmon resonance [5] while these particles exhibit, also, strong magnetic circular dichroism [6].

In general, composite magnetoplasmonic nanoparticles with a core–shell morphology have intriguing optical properties and offer impressive opportunities for tailoring in a controllable manner the light–matter interaction at subwavelength dimensions. These properties are usually analysed in the framework of the quasistatic approximation which, however, is often inadequate and thus a full electrodynamic treatment is required. In the present paper we report a thorough study of localized surface plasmon modes in silver-coated metallic or dielectric magnetic nanospheres and investigate the role of the plasmonic resonances in the occurrence of strong circular dichroism, by means of both quasistatic and full electrodynamic calculations, extending our previous work on the subject [6]. In Section 2 we present the quasistatic approximation for light absorption by spherical nanoparticles consisting of a magnetic core and a nonmagnetic shell and in Section 3 we briefly describe the multipole method for a full electrodynamic treatment of the problem. Section 4 is devoted to the analysis and discussion of our results and the last section concludes the article.

2. Quasistatic approximation

The rate of work done per unit volume on the matter by the EM field, which is dissipated as Joule heating, is generally given by

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$$\frac{dW(\mathbf{r},t)}{dt} = \mathbf{j}(\mathbf{r},t)\cdot\mathbf{E}_0(\mathbf{r},t), \tag{1}$$

where $\mathbf{E}_0$ is the external electric field and $\mathbf{j}$ the induced current density. Assuming time-harmonic, monochromatic fields of angular frequency ω, $\mathbf{E}_0(\mathbf{r},t) = \mathrm{Re}[\mathbf{E}_0(\mathbf{r})\exp(-i\omega t)]$ and $\mathbf{j}(\mathbf{r},t) = \mathrm{Re}[\mathbf{j}(\mathbf{r})\exp(-i\omega t)]$, the absorbed power, averaged over the wave period $T = 2\pi/\omega$, takes the form

$$P_{abs} = \frac{1}{2}\int d^3r\ \mathrm{Re}\left[\mathbf{j}^*(\mathbf{r})\cdot\mathbf{E}_0(\mathbf{r})\right]. \tag{2}$$

We consider a particle, much smaller than the wavelength, embedded in a homogeneous and isotropic nonabsorbing host medium, characterized by a (real) relative dielectric permittivity ϵ_h and a (real) relative magnetic permeability μ_h. Considering the particle as a point dipole, the induced electric dipole moment density has the form $\mathbf{P}(\mathbf{r},t) = \mathrm{Re}[\mathbf{p}\delta(\mathbf{r})\exp(-i\omega t)]$, where $\mathbf{p}$ is the particle dipole moment which oscillates with angular frequency ω. In this case, using Maxwell equation $\nabla\times\mathbf{H}(\mathbf{r},t) = \partial_t[\epsilon_0\mathbf{E}_0(\mathbf{r},t) + \mathbf{P}(\mathbf{r},t)] + \mathbf{j}(\mathbf{r},t)$, Eq. (2) gives

$$P_{abs} = \frac{\omega}{2}\,\mathrm{Im}\left[\mathbf{p}^*\cdot\mathbf{E}_0\right], \tag{3}$$

where $\mathbf{E}_0 = \hat{\mathbf{e}}E_0$ with E_0 being the electric field amplitude and $\hat{\mathbf{e}}$ a unit vector that denotes the polarization of the wave. On the other hand, in the quasistatic approximation, the electric dipole moment induced by the external field is given by $\mathbf{p} = \epsilon_h\overleftrightarrow{\alpha}\mathbf{E}_0$, where $\overleftrightarrow{\alpha}$ is the particle polarizability tensor. Therefore, Eq. (3) takes the form

$$P_{abs} = \frac{\epsilon_h\omega}{2}\,\mathrm{Im}\left[\mathbf{E}_0^\dagger\overleftrightarrow{\alpha}^\dagger\mathbf{E}_0\right], \tag{4}$$

where the dagger denotes adjoint operator. For a spherical particle of radius S, we define the normalized absorption cross section $\sigma_{abs} = P_{abs}/P_0$, where $P_0 = \pi S^2|E_0|^2/(2Z_h)$, with $Z_h = \sqrt{\mu_0\mu_h/(\epsilon_0\epsilon_h)}$ being the impedance of the host medium, is the incident power through a (circular) section of area πS^2. Therefore, the normalized absorption cross section takes the form

$$\sigma_{abs} = \frac{q_h}{\pi\epsilon_0 S^2}\,\mathrm{Im}\left[\hat{\mathbf{e}}^\dagger\overleftrightarrow{\alpha}^\dagger\hat{\mathbf{e}}\right], \tag{5}$$

where $q_h = \omega\sqrt{\epsilon_0\epsilon_h\mu_0\mu_h} = \omega\sqrt{\epsilon_h\mu_h}/c$, with c being the velocity of light, is the wavenumber in the host medium.

In the present work we shall be concerned with composite nanoparticles consisting of a magnetic spherical core of radius S_c coated with a nonmagnetic concentric spherical shell of outer radius S, relative electric permittivity function ϵ_s and relative magnetic permeability $\mu_s(=1)$. The optical response of magnetic materials in the visible and infrared part of the spectrum is described by a relative magnetic permeability $\mu_g = 1$ and a relative electric permittivity tensor

$$\overleftrightarrow{\epsilon}_g = \epsilon_z\begin{pmatrix}\epsilon_r & -i\epsilon_\kappa & 0\\ i\epsilon_\kappa & \epsilon_r & 0\\ 0 & 0 & 1\end{pmatrix}, \tag{6}$$

if we take the magnetization along the z-axis. The tensor components are in general complex functions of frequency due to the dissipative and dispersive behaviour of the material. The magneto-optical properties are due to the nondiagonal components, which maintain sign upon magnetization reversal and vanish above the Curie temperature.

Magnetic circular dichroism spectroscopy measures the difference between two absorption spectra acquired using light with opposite helicity in the presence of a magnetic field parallel to the incident light direction [3]. Since the wavelength of light is much longer than the size of the nanoparticles under consideration, their optical response can be described by a dipole polarizability tensor [7,8]

$$\overleftrightarrow{\alpha} = 4\pi\epsilon_0 S^3(\overleftrightarrow{\epsilon} - \epsilon_h\overleftrightarrow{I})(\overleftrightarrow{\epsilon} + 2\epsilon_h\overleftrightarrow{I})^{-1}, \tag{7}$$

where $\overleftrightarrow{I}$ is the diagonal unit tensor and $\overleftrightarrow{\epsilon}$ an effective particle permittivity tensor which, in the spirit of the quasistatic Maxwell–Garnett homogenization method, is obtained by the equation [9,10]

$$(\overleftrightarrow{\epsilon} - \epsilon_s\overleftrightarrow{I})(\overleftrightarrow{\epsilon} + 2\epsilon_s\overleftrightarrow{I})^{-1} = \left(\frac{S_c}{S}\right)^3(\overleftrightarrow{\epsilon}_g - \epsilon_s\overleftrightarrow{I})(\overleftrightarrow{\epsilon}_g + 2\epsilon_s\overleftrightarrow{I})^{-1}. \tag{8}$$

The permittivity tensor, $\overleftrightarrow{\epsilon}_g$, given by Eq. (6), can be readily diagonalized and its first two diagonal elements, $\epsilon_g^{(+)} = \epsilon_z(\epsilon_r + \epsilon_\kappa)$ and $\epsilon_g^{(-)} = \epsilon_z(\epsilon_r - \epsilon_\kappa)$, correspond to left-circular polarized (LCP) and right-circular polarized (RCP) waves, respectively, propagating along the z direction, which is taken to be along the direction of the magnetization, while the third diagonal element is ϵ_z. Therefore, in the quasistatic approximation, the corresponding diagonal elements of $\overleftrightarrow{\alpha}$, $\alpha^{(+)}$ and $\alpha^{(-)}$ provide the (normalized) absorption cross section of the core–shell particles under consideration for LCP and RCP light incident along the z direction according to Eq. (5):

$$\sigma_{abs}^{(\pm)} = \frac{q_h}{\pi\epsilon_0 S^2}\,\mathrm{Im}\,\alpha^{(\pm)}. \tag{9}$$

In the absence of gyrotropy, i.e., for unmagnetized core, $\epsilon_g^{(+)} = \epsilon_g^{(-)}$ and the two circular plasmonic modes are degenerate. Within the quasistatic approach, $\epsilon^{(+)} = \epsilon^{(-)} \equiv \epsilon$ and in the vicinity of the resonance frequency ω_0 given by the Fröhlich condition: $\mathrm{Re}\,\epsilon(\omega_0) + 2\epsilon_h = 0$, in the low-loss regime $\mathrm{Im}\,\epsilon(\omega) \ll 1$, we obtain to first order $\sigma_{abs} \cong A/[(\omega-\omega_0)^2 + \Gamma]$, where A and Γ are positive constants. In the presence of weak gyrotropy, $\epsilon^{(\pm)} \cong \epsilon \pm \delta\epsilon$ where $\delta\epsilon$ is a small quantity, and the degeneracy of the two circular plasmonic modes is removed. To first-order approximation, $\sigma_{abs}^{(\pm)}$ have the same Lorentzian-like spectral shape, as in the absence of gyrotropy, about ω_0 shifted downwards/upwards by the same small amount $\delta\omega \cong \mathrm{Re}\,\delta\epsilon(\omega_0)/(d/d\omega)\,\mathrm{Re}\,\epsilon(\omega)|_{\omega_0}$, i.e., $\omega_0 \pm \delta\omega$. Therefore, $\Delta\sigma_{abs} \equiv \sigma_{abs}^{(+)} - \sigma_{abs}^{(-)}$, which is usually adopted as a quantitative measure of circular dichroism [11], has a bipolar line shape. Whether this varies from positive to negative values with increasing frequency, or the opposite, depends on the sign of $\delta\omega$, i.e., on the specific form of the effective permittivity about the resonance-frequency, according to the above equation.

3. Multipole method

The electric field component of a harmonic, monochromatic plane EM wave, of angular frequency ω and wave vector $\mathbf{q}_h$, propagating in a homogeneous and isotropic medium characterized by a relative electric permittivity ϵ_h and a relative magnetic permeability μ_h, has the general form $\mathbf{E}_0(\mathbf{r},t) = \mathrm{Re}[\hat{\mathbf{e}}E_0\exp[i(\mathbf{q}_h\cdot\mathbf{r} - \omega t)]]$. This plane wave can be expanded into regular vector spherical waves, about a given origin of coordinates, with amplitudes $\mathbf{A}^0_{Plm}(\hat{\mathbf{q}}_h)\cdot\hat{\mathbf{e}}E_0$, $P = E, H$, $l = 1, 2, \ldots$ and $m = -l, -l+1, \ldots l$, and

$$\begin{aligned}\mathbf{A}^0_{Hlm}(\hat{\mathbf{q}}_h) &= \frac{4\pi i^l(-1)^{m+1}}{\sqrt{l(l+1)}}\Big\{\Big[\alpha_l{}^m\cos\theta e^{i\phi}Y_{l-m-1}(\hat{\mathbf{q}}_h)\\ &\quad + m\sin\theta Y_{l-m}(\hat{\mathbf{q}}_h) + \alpha_l{}^{-m}\cos\theta e^{-i\phi}Y_{l-m+1}(\hat{\mathbf{q}}_h)\Big]\\ &\quad \hat{\mathbf{e}}_1 + i\Big[\alpha_l{}^m e^{i\phi}Y_{l-m-1}(\hat{\mathbf{q}}_h) - \alpha_l{}^{-m}e^{-i\phi}Y_{l-m+1}(\hat{\mathbf{q}}_h)\Big]\hat{\mathbf{e}}_2\Big\}\end{aligned} \tag{10}$$

and