Plasmon enhanced direct and inverse Faraday effects in non-magnetic nanocomposites

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While applications of plasmonics are rapidly growing, magneto-optical effects in nanocomposites are poorly understood. We therefore devote this paper to the theoretical analysis of magneto-optical effects in nanocomposites. Based on the Drude model, we derived the constitutive equation where the dielectric and coupling functions describe the interactions of metal nanoparticles with magnetic field. In the limitation of low volume fraction of metal nanoparticles (i.e., when the material is still transparent), these functions were calculated within the Maxwell–Rayleigh theory of dilute suspensions. We showed that in the absence of external magnetic fields, a non-magnetic nanoparticle can be magnetized in the circularly polarized light beam, and the magnetization depends on the direction of rotation of the light wave. The external magnetic field alters the particle magnetization, and when the fields are weak, this change in magnetization linearly depends on the particular field. The proposed theory was applied to an analysis of the Faraday effect in nanocomposites. We predicted a resonance behavior of the Verdet function in nanocomposites and its dependence on concentration, sample thickness, and external magnetic field.

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1. INTRODUCTION

Plasmonics is an emerging field of optics, dealing with the light-induced excitations of charges in metal particles. In metals, these excitations are called "plasmons," thus emphasizing the nature of the electron oscillations described in terms of the solid state plasma [1–6]. Plasmons can be observed in very small metal nanoparticles and thin nanofilms [2]. Especially attractive are noble metals, such as gold, silver, and copper [7], where the electron–lattice and electron–electron interactions are diminished. Plasmons can be described as an assembly of free electrons moving through a positively charged matrix [2,8,9]. In this model, proposed by Drude [8,9], the matrix is considered as a continuum resisting the electron movement with a velocity-dependent force. This type of electron excitations is called volume plasmons. Within the Drude model, one can analytically derive an effective dielectric function of the macroscopic material, keeping track of electron–field interactions. One can corroborate that in the particles or films, the free electrons oscillate in phase with the applied oscillating electric field, but the oscillations significantly depend on the materials parameters. It has been shown [2–6] that the optical properties of metal colloids and nanocomposites (i.e., dielectric liquids or solids with embedded nanoparticles) are drastically different from those of the matrix and bulk metal. The vast literature on solid state plasmonics shows that metal colloids and nanocomposites demonstrate a resonant behavior in oscillating electromagnetic fields [1–6]. This resonant behavior of nanoparticles is caused by the confinement of the conduction electrons within small particle volume. Despite unknown physical origin, this effect of optical resonance has been effectively exploited for centuries, such as in formulation of different paints. The physics of interactions of nanoparticles with electromagnetic fields has become clearer only recently [3–6]. In the known applications, the observed effects are mostly probed directly as a color change in colloids. Only recently, the magneto-optical effects have attracted the interest in this field, and this interest is definitely growing as seen from recent theoretical and experimental papers [10–17]. The present work is devoted to the theoretical analysis of magneto-optical properties of nanocomposites with metal nanoparticles.

In particular, we discuss the resonances of the magneto-optical Faraday effect in nanocomposites. While the Drude model has been widely accepted for the description of volume plasmons, to the best of our knowledge, the Drude model has never been applied for the analyses of magneto-optical effects in nanocomposites. We therefore undertake what we believe to be the first step in the development of magneto-plasmonics and discuss the role of volume plasmons in Faraday effects.

In Faraday's experiments [13], the applied magnetic field is co-directed with the light wave vector. To date, most of the studies on nanocomposites have largely been experimental. The literature review and new experiments on gold-coated iron oxide nanoparticles can be found in [18]. A phenomenological theory of Faraday effect in composites has been discussed in [19,20]. The analysis of light-induced magnetization and the mechanism of the enhancement of the Faraday effect were left aside of these

theories. The authors of [20] used a perturbation method keeping only linear magnetic-field-dependent terms; this approximation is questionable near resonances when the losses are small and the Lorentz force becomes significant [21,22]. More rigorous approach to this problem can be found in [10].

In this paper, using the Drude model, we derive the basic equations describing the materials magnetization. To the best of our knowledge, this derivation has never been discussed in the literature. These basic formulas explain the microscopic nature of the inverse Faraday effect that has been phenomenologically predicted by Pitaevskii in 1961 [23]. Using this microscopic model, we apply a quasistatic approximation for the electromagnetic field of light to accurately solve the problem of nanoparticle interaction with the light. Because of the new gyromagnetic constitutive equation, this problem deserves special attention. It can be considered as a basic problem for the analysis of Rayleigh scattering on gyromagnetic nanoparticles. We extend the approach of [10] and explicitly solve the electrostatic problem. It appears that the particle polarization is a nonlinear function of the external magnetic field. In weak magnetic fields (some few Teslas), this dependence is reduced to a linear dependence, leading to the Faraday effect with the Verdet function, depending on the light frequency. With the explicit solutions for the electromagnetic fields, one can study the magnetization in nanocomposites. We show that the magnetization in nanocomposites has a resonance behavior, thus predicting the resonances for inverse Faraday effect. We then show that in the direct Faraday effect the angle of rotation of the **E**-vector of light should also have a resonance behavior. Remarkably, this resonance depends on the properties of the nanoparticle-matrix pair. We believe that our theory points to a constructive approach for design of materials for optical isolators, Faraday rotators, and other magneto-optical devices with superior magneto-optical properties.

2. LIGHT-INDUCED MAGNETIZATION OF NON-MAGNETIC METALS

Before proceeding to an analysis of interactions of the electromagnetic field with nanocomposites, consider the magnetic behavior of metals subjected to electromagnetic radiation. Using the Drude model [8,9], we can write the equation of motion for the electrons as

$$m\frac{d\mathbf{v}}{dt} + \gamma m\mathbf{v} = e\mathbf{E} + e\mathbf{v}\times\mathbf{B}, \tag{1}$$

where m is the effective mass of the electron, $\mathbf{v}=d\mathbf{r}/dt$ is the electron velocity, r is the radius-vector of the electron, γ is the damping constant, $\mathbf{E}=E_0\exp(\pm i\omega t)=E_0\cos(\omega t)\pm iE_0\sin(\omega t)=E_x\pm iE_y$ is the oscillating electric field of light, and $\mathbf{B}$ is the constant external magnetic field perpendicular to the **E**-vector. The frequency ω in this definition is always a positive number. We will call wave $\mathbf{E}^+=E_0\exp(+i\omega t)$ the "plus"-wave, and wave $\mathbf{E}^-=E_0\exp(-i\omega t)$ the "minus"-wave.

We seek a solution to Eq. (1) in the form $\mathbf{r}=r\exp(\pm i\omega t)$, where the radius-vector of the electron rotates in the same **E**-plane. In this case, the velocity-vector of the electron, $\mathbf{v}=\pm i\omega r\exp(\pm i\omega t)=\pm i\omega(x\pm iy)$, is perpendicular to the **r**-vector. Therefore, the Lorentz force is equal to $e\mathbf{v}\times\mathbf{B}=ev_yB-iev_xB=\pm eB\omega r\exp(\pm i\omega t)$. Hence, the Lorentz force is co-directed with the radius-vector of the electron. These arguments show that the movement of the electron occurs in the **E**-plane.

Within the Drude model of free electrons, the dipole moment of the ion–electron pair is defined as the quantity $\mathbf{P}_{ij}=e\mathbf{r}_{ij}$, where $\mathbf{r}_{ij}$ is the radius-vector of the "ith" electron with respect to the center of the "jth" ion [22]. Then the average dipole moment, produced by an ensemble of N electrons in volume V, is defined as the quantity $\mathbf{P}_{ij}$ averaged over the sample volume [22]: $\mathbf{P}=(1/V)\int\Sigma_{ij}e\mathbf{r}_{ij}\delta(\mathbf{R}-\mathbf{R}_j)d^3\mathbf{R}$, where, $\mathbf{R}_j$ is the radius-vector of ion "j," modeled as a point charge. The delta-function picks each ion from the ensemble. Therefore, in order to find this average, we need to solve Eq. (1) for each electron.

For weak magnetic fields (some few Teslas), the Lorentz force is small [i.e., the inequality $eB/(m\omega)\ll 1$ holds true]. Therefore, we can use a perturbation method to solve Eq. (1). In the first approximation with respect to the magnetic field, the polarization for anti-clockwise rotating field, $\mathbf{E}=E_0\exp(i\omega t)$, is obtained as

$$\mathbf{P}(\omega) = -i\frac{e^2N}{m}\frac{(\gamma-i\omega)}{\omega(\gamma^2+\omega^2)}\mathbf{E} - i\frac{e^3N}{m^2}\frac{(\gamma-i\omega)^2}{\omega(\gamma^2+\omega^2)^2}\mathbf{E}\times\mathbf{B}. \tag{2}$$

Then for the clockwise rotating field, we have $\mathbf{P}(-\omega)=\mathbf{P}^*(\omega)$, where the star denotes the complex-conjugated function. Hence, the vector of electric displacement field is introduced as

$$\mathbf{D}(\omega) = \varepsilon_0\mathbf{E}(\omega) + \mathbf{P}(\omega) = \varepsilon_0\varepsilon(\omega)\mathbf{E}(\omega) + if(\omega)\mathbf{E}(\omega)\times\mathbf{B}, \tag{3}$$

where ε_0 is the permittivity of free space, $\varepsilon(\omega)=1-i(e^2N/m\varepsilon_0\omega)(\gamma-i\omega)(\gamma^2+\omega^2)^{-1}$ is the complex permittivity, and $f(\omega)=-(e^3N/m^2\omega)(\gamma-i\omega)^2(\gamma^2+\omega^2)^{-2}$ is the coupling function describing the interaction of metal with magnetic field. For dielectric function $\varepsilon(\omega)$ and f-function $f(\omega)$, the following conjugation conditions derived from Eq. (2) hold: $\varepsilon(-\omega)=\varepsilon^*(\omega)$, $f(-\omega)=-f^*(\omega)$.

The Drude model allows one to find the light-induced magnetization [22]. Be reminded that our metal does not have any spontaneous magnetic order, i.e., it is neither ferromagnetic nor antiferromagnetic. Using the microscopic definition of magnetization vector $\mathbf{M}=(1/2V)\int\Sigma_{ij}e[\mathbf{r}_{ij}\times\mathbf{v}_{ij}]\delta(\mathbf{R}-\mathbf{R}_j)d^3\mathbf{R}$ and introducing a $\hat{\mathbf{k}}$-vector as the unit vector pointing toward the direction of light propagation, we find $\mathbf{r}\times\mathbf{v}=\pm\omega([r\cos(\omega t)]^2+[r\sin(\omega t)]^2)\hat{\mathbf{k}}=\pm\omega r^2\hat{\mathbf{k}}$. Therefore, the magnetization in a non-magnetic metallic sample placed in a circularly polarized electric field, $\mathbf{E}^\pm=E_0\exp(\pm i\omega t)$, is parallel to the light beam, $\mathbf{M}^\pm\propto e\mathbf{r}\times\mathbf{v}\propto\pm e\hat{\mathbf{k}}$. Remarkably, the sign of magnetization depends on the direction of rotation of the electric field. Substituting r^2 for the asymptotic solution of Eq. (1) as $B\to 0$, we obtain the magnetization in the first approximation with respect to the magnetic field:

$$\mathbf{M}^{\pm} = \pm\left(\frac{1}{\omega(\gamma^2+\omega^2)} + 2\frac{eB}{m}\frac{1}{(\gamma^2+\omega^2)^2}\right)\frac{Ne^3}{m^2}E_0^2\hat{\mathbf{k}}$$
$$= \pm g(\omega,B)E_0^2\hat{\mathbf{k}}. \quad (4)$$

Again, the frequency ω in this definition is always a positive number. Thus, one can clearly see within the Drude model that even without magnetic field, any metal particle will acquire a magnetic moment parallel to the beam of a circularly polarized light. Pitaevskii first predicted this effect on the basis of phenomenological electrodynamics [23]. Now it is called the "inverse Faraday effect" [23–25]. As follows from Eq. (4), in metals at high optical frequency, $\omega \gg \gamma$, the magnetization decreases as $M \propto 1/\omega^3$; and, therefore, the inverse Faraday effect is typically small. In this paper, we show that the inverse and direct Faraday effects can be significantly enhanced in metal nanoparticles embedded in a dielectric matrix.

3. LIGHT-INDUCED POLARIZATION AND MAGNETIZATION OF NANOPARTICLES

For the sake of generality, we assume that both materials in the nanocomposite (i.e., the matrix and nanoparticles) follow the same constitutive equation (3), and we will use the index l to label the matrix-liquid in the case of colloids and the index m to label the non-magnetic metal nanoparticles. The permeability of these metals is close to 1, and therefore the magnetic field $\mathbf{B}$ and magnetizing field $\mu_0\mathbf{H}$ can be considered identical. For small spherical particles of radius R, $R \ll \lambda/4\pi$, the field distribution can be found in a quasi-static limit [26]. This limit assumes that the fields satisfy electrostatic equilibrium conditions: $\nabla\cdot\mathbf{D}^i=0$, $\nabla\times\mathbf{E}^i=0$ (with $i=l,m$), where the electric displacement vectors $\mathbf{D}^{l,m}$ for the matrix and nanoparticle are defined by Eq. (3) with corresponding ε_i and f_i, where $i=l,m$. The solution to this problem will give us the field distribution around a sphere placed in a uniform electric field $\bar{\mathbf{E}}$ at infinity [26,27]. Applying the vector equality $\nabla\cdot(\mathbf{E}\times\mathbf{B})=\mathbf{B}\cdot(\nabla\times\mathbf{E})-\mathbf{E}\cdot(\nabla\cdot\mathbf{B})$, we conclude that the condition $\nabla\cdot\mathbf{D}^i=0$ is reduced to $\nabla\cdot(\varepsilon_i\mathbf{E}^i)=0$, with $i=l,m$. To satisfy this condition, it is convenient to introduce the potentials of the electric field inside (φ^l) and outside (φ^m) the particle. Then the full field is obtained as $\mathbf{E}^{l,m}=\nabla\varphi^{l,m}+\bar{\mathbf{E}}$, where $\bar{\mathbf{E}}(\omega)$ is the average electric field of the light in the nanocomposite. The average field $\bar{\mathbf{E}}(\omega)$ is considered constant for this electrostatic problem, and it defines the boundary condition for potential φ^l at infinity. Hence, the problem of electrostatic equilibrium is reduced to solution of the Laplace equation $\Delta\varphi^{l,m}=0$ for potentials φ^l and φ^m. In the spherical system of coordinates $\mathbf{r}=(r,\theta)$, with the origin at the particle center, the boundary conditions for this problem are two continuity conditions at the particle boundary $r=R$ and the condition at infinity:

$$\varphi^l = \varphi^m, \quad r=R,$$
$$(\mathbf{D}^l-\mathbf{D}^m)\cdot\mathbf{n}=0, \quad r=R, \quad (5)$$

where $\mathbf{n}$ is the normal vector. Because of the second term in the right-hand side of Eq. (3), the boundary condition $(\mathbf{D}^l-\mathbf{D}^m)\cdot\mathbf{n}=0$ of this electrostatic problem is different from the classical problem used for an analysis of optical plasma resonances [4–6,22]. In our case, the second line of Eq. (5) is rewritten as

$$\varepsilon_0\varepsilon_l(\omega)\left(\nabla\varphi^l\cdot\frac{\mathbf{R}}{R}\right)-\varepsilon_0\varepsilon_m(\omega)\left(\nabla\varphi^m\cdot\frac{\mathbf{R}}{R}\right)$$
$$+i\left\{f_l(\omega)\left([\nabla\varphi^l\times\mathbf{B}]\cdot\frac{\mathbf{R}}{R}\right)-f_m(\omega)\left([\nabla\varphi^m\times\mathbf{B}]\cdot\frac{\mathbf{R}}{R}\right)\right\}$$
$$=-\varepsilon_0(\varepsilon_l(\omega)-\varepsilon_m(\omega))\left(\bar{\mathbf{E}}\cdot\frac{\mathbf{R}}{R}\right)$$
$$-i\left\{(f_l(\omega)-f_m(\omega))\left([\bar{\mathbf{E}}\times\mathbf{B}]\cdot\frac{\mathbf{R}}{R}\right)\right\} \quad r=R, \quad (6)$$

$$\varphi^l=0 \quad r\to\infty. \quad (7)$$

When the external magnetic field is directed along the light beam, and it is perpendicular to E-field of light, we have

$$i[\bar{\mathbf{E}}\times\mathbf{B}] = i(E_yB-iE_xB) = E_xB+iE_yB = B\bar{\mathbf{E}}. \quad (8)$$

Hence, there is only one selected direction in this electrostatic problem, the direction of the E-field of light. Using the following formulas from vector analysis, $\nabla(\mathbf{b}\cdot\mathbf{r})=\mathbf{b}$, $\nabla f(r)=(\mathbf{r}/r)df/dr$, and noticing that the vector $[\mathbf{R}\times\mathbf{B}]$ is orthogonal to $\mathbf{R}$ (i.e., the equality $[\mathbf{R}\times\mathbf{B}]\cdot\mathbf{R}=0$ holds), we seek the solution to the formulated electrostatic problem in the form

$$\varphi^{l,m} = (\bar{\mathbf{E}}\cdot\mathbf{r})\Phi^{l,m}(r), \quad (9)$$

where

$$\Phi^l(r) = \frac{\beta}{r^3}(\text{liquid}), \quad \Phi^m(r) = \alpha(\text{parcticle}). \quad (10)$$

The ansatz (9) and (10) satisfies the Laplace equation inside and outside the nanoparticle [22], and the constants α and β must be found from the boundary conditions (6)–(9). Substituting Eq. (10) in Eqs. (5)–(9), we obtain

$$\alpha = \frac{\beta}{R^3} = \frac{\varepsilon_l(\omega)-\varepsilon_m(\omega)+(f_l(\omega)-f_m(\omega))B/\varepsilon_0}{2\varepsilon_l(\omega)+\varepsilon_m(\omega)+(f_m(\omega)-f_l(\omega))B/\varepsilon_0}. \quad (11)$$

Therefore, the total electric field $\mathbf{E}=\nabla\varphi^{l,m}+\bar{\mathbf{E}}=\nabla\varphi$ in the particle and in the liquid is described by the following potentials:

$$\varphi(r) = \frac{3\varepsilon_l(\omega)(\bar{\mathbf{E}}\cdot\mathbf{r})}{2\varepsilon_l(\omega)+\varepsilon_m(\omega)+(f_m(\omega)-f_l(\omega))B/\varepsilon_0}, \quad (r<0) \quad (12)$$

$$\varphi(r) = (\bar{\mathbf{E}}\cdot\mathbf{r})\cdot\left\{1+\frac{(\varepsilon_l(\omega)-\varepsilon_m(\omega))+(f_l(\omega)-f_m(\omega))B/\varepsilon_0}{2\varepsilon_l(\omega)+\varepsilon_m(\omega)+(f_m(\omega)-f_l(\omega))B/\varepsilon_0}\frac{R^3}{r^3}\right\}, \quad (r\geq 0). \quad (13)$$

Equation (13) can be interpreted as a potential of an electric field produced by a dipole with the moment $\mathbf{d}$, $\varphi=(\bar{\mathbf{E}}\cdot\mathbf{r})+(\mathbf{d}\cdot\mathbf{r})/(4\pi\varepsilon_0\varepsilon_l r^3)$, where the dipole is defined as

$$\mathbf{d}=4\pi\varepsilon_0\varepsilon_l R^3\bar{\mathbf{E}}\cdot\frac{(\varepsilon_l(\omega)-\varepsilon_m(\omega))+(f_l(\omega)-f_m(\omega))B/\varepsilon_0}{2\varepsilon_l(\omega)+\varepsilon_m(\omega)+(f_m(\omega)-f_l(\omega))B/\varepsilon_0}. \tag{14}$$

In the materials without dissipation $\mathrm{Im}(\varepsilon)=0$ and $\mathrm{Im}(f)=0$ the dipole moment is always co-directed with the E-field of light. In the absence of magnetic field, Eq. (14) is identical to the familiar equation for light-induced dipole used in plasmonics [1–6].

We now turn to the light-induced magnetization in the nanoparticle. Using the definition given by Eq. (4), we substitute for the electric field $E_0^2=|\nabla\varphi|^2$, where φ is defined by Eq. (12). Introducing the complex-valued function A we obtain the nanoparticle magnetization as

$$\mathbf{M}^{\mp}=\pm g(\omega,B)|A_{\pm}|^2\bar{E}^2\mathbf{k}, \tag{15}$$

where

$$A_{\pm}=\frac{3\varepsilon_l(\pm\omega)}{2\varepsilon_l(\pm\omega)+\varepsilon_m(\pm\omega)+(f_m(\pm\omega)-f_l(\pm\omega))B/\varepsilon_0}, \tag{16}$$

where ω is always positive. As seen from Eqs. (14)–(16), the dipole and magnetic moments of the metal particle can be significantly increased as we move to the resonance frequency corresponding to the roots of denominator in Eqs. (14) and (16).

It is instructive to estimate the magnetization acquired by different metals in electric field of light. The potential energy of a magnetized nanoparticle in magnetic filed B is written as $U^{\pm}=-\mathbf{M}^{\pm}\cdot\mathbf{B}$, where the superscript + corresponds to the plus-wave, and the superscript − corresponds to the minus-wave. The energy of the electromagnetic wave is proportional to ε_0E^2. Therefore, the ratio $U^{\pm}/\varepsilon_0E^2$ shows what is the fraction of the wave energy

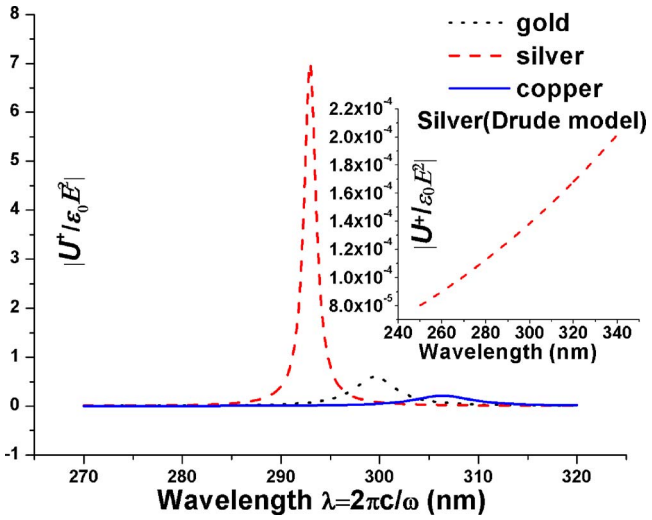

Fig. 1. (Color online) Dimensionless magnetic energy of metal nanoparticles calculated from Eqs. (15) and (16) for plus-waves. The matrix is water ($\varepsilon_l=n^2=1.77$, $f_l=6.3\times10^{-18}$ F m^{-1} T^{-1}) [the inset shows the corresponding dimensionless magnetic energy for silver calculated from Eq. (4)].

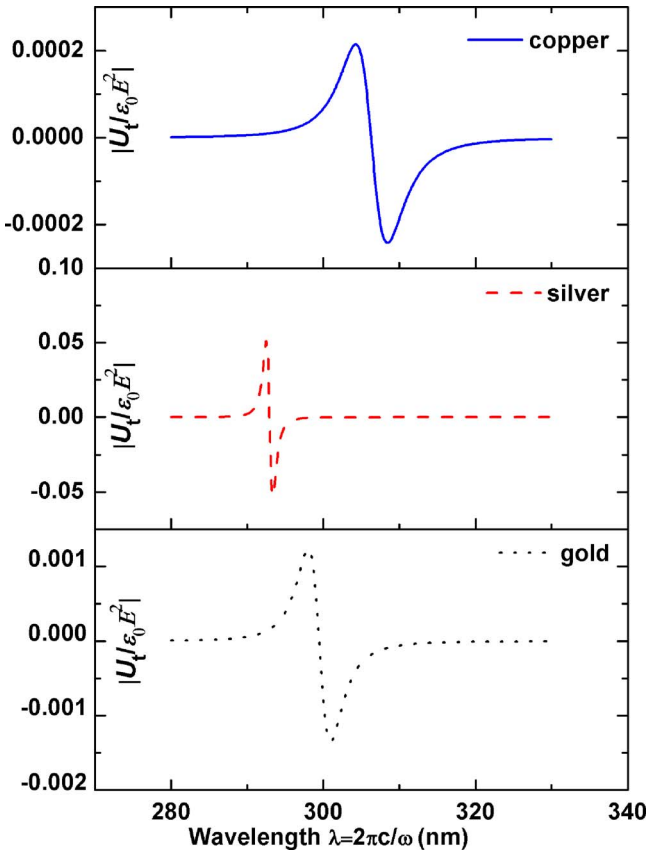

Fig. 2. (Color online) Dimensionless magnetic energy induced by plus- and minus-waves.

has been spent to magnetize the nanoparticle. As an example, we consider the magnetization in the field $B=1$ T. As seen from Fig. 1, near the resonance frequency, the magnetization induced by plus-waves is greatly enhanced. The total magnetization is the sum of plus- and minus-magnetizations, $\mathbf{M}=\mathbf{M}^+ +\mathbf{M}^-$. In Fig. 2 we show the total magnetic energy $U_t=-\mathbf{M}\cdot\mathbf{B}$. Because the waves of different circular polarizations are counter-directed, the total energy is much smaller than the energy of plus and minus magnets. Consider a laser beam with the cross-sectional area $S\sim10^{-6}$ m^2 and the beam power $P\sim10^{-3}$ W. The energy is estimated as $\varepsilon_0E^2\sim P/S/c\sim10^{-5}$ J/m^3 (c is the speed of light). From Fig. 2 we see that for silver nanoparticle the ratio U_t/ε_0E^2 is about ~0.05. Taking the volume of the nanoparitcle as $V\sim10^{-27}$ m^3, we have the energy of magnetization for each nanoparticle $U_tV\sim10^{-34}$ J. This energy is well below the energy of thermal excitations at room temperature $k_BT\sim10^{-21}$ J. Therefore, the light-induced magnetization is most likely be suppressed by the thermal fluctuations.

4. OPTICAL PROPERTIES OF NANOCOMPOSITES

This section considers a composite made of metal nonmagnetic nanoparticles. The phenomenological constitutive equation for the average electric displacement field $\bar{\mathbf{D}}$ in the nanocomposite representing a gyroscopic material takes the form of Eq. (3), $\bar{\mathbf{D}}(\omega)=\varepsilon_0\varepsilon(\omega)\bar{\mathbf{E}}(\omega)+if(\omega)[\bar{\mathbf{E}}(\omega)$

Table 1. Physical Parameters of Noble Metals [7]

	Plasma Frequency $\omega_p=(e^2N/m\varepsilon_0)^{1/2}$ (10^{16} s^{-1})	Number of Electrons per unit Volume $N(\times 10^{28})$ (1/m^3)	Effective mass m (Electron Masses)	Relaxation Time $\tau=1/\gamma$ ($\times 10^{15}$ s)
Gold	1.34	5.57	0.99	9.3
Silver	1.37	5.63	0.96	31
Copper	1.31	8.04	1.49	6.9

$\times \mathbf{B}]$, where ε is the averaged permittivity, and f is the average coupling parameter. While the average fields $\bar{\mathbf{D}}(\omega)$ and $\bar{\mathbf{E}}(\omega)$ are constants, the local fields $\mathbf{D}^l$ and $\mathbf{E}^l$ in the matrix and the fields $\mathbf{D}^m$ and $\mathbf{E}^m$ inside the nanoparticles depend on the spatial variable $\mathbf{r}$.

We assume that the concentration of nanoparticles in the nanocomposite is low. If c_0 is the number of particles per unit volume, and λ is the wavelength of light, the following inequality $(1/c_0)^{1/3} \gg \lambda$ quantitatively specifies this limit. Then the average parameters of a nanocomposite can be obtained by using Rayleigh's arguments [26] and defining the averaged fields through the following equations [22,28]:

$$\frac{1}{V}\int \{\mathbf{D}(\omega,\mathbf{r}) - \varepsilon_0\varepsilon_l(\omega,\mathbf{r})\mathbf{E}(\omega,\mathbf{r}) - if_l(\omega,\mathbf{r})[\mathbf{E}(\omega,\mathbf{r})\times\mathbf{B}]\}d^3\mathbf{r}$$

$$= \bar{\mathbf{D}}(\omega) - \varepsilon_0\varepsilon_l(\omega)\bar{\mathbf{E}}(\omega) - if_l(\omega)[\bar{\mathbf{E}}(\omega)\times\mathbf{B}]. \quad (17)$$

In this equation, $\mathbf{D}(\omega,\mathbf{r})$ and $\mathbf{E}(\omega,\mathbf{r})$ are the electric displacement field and electric field, respectively, at the point $\mathbf{r}$, and V is the sample volume. As follows from the left-hand side of Eq. (5), the integral is nonzero only inside the particles. Therefore, the integral has to be only taken over the particle volume. Substituting Eq. (12) in the right-hand side of Eq. (17), and using Eq. (8) to separate the terms containing magnetic field $\mathbf{B}$ in the nominators, we obtain the constitutive equation for nanocomposite as

$$\bar{\mathbf{D}}(\omega) = \varepsilon_0\varepsilon(\omega)\bar{\mathbf{E}}(\omega) + if(\omega)[\bar{\mathbf{E}}(\omega)\times\mathbf{B}]$$

$$= \varepsilon_0\varepsilon(\omega)\bar{\mathbf{E}}(\omega) + f(\omega)B\bar{\mathbf{E}}(\omega), \quad (18)$$

or, $D_i=\varepsilon^c_{ij}E_j$, $\varepsilon^c_{ij}=\varepsilon_0\varepsilon(\omega)\delta_{ij}+if(\omega)e_{ijn}B_n$, where the indices i, j, and n, correspond to x, y, or z components of the field vectors. δ_{ij} is the Kronecker delta, and e_{ijn} is the Levi–Civita alternating symbol [22], and the averaged dielectric and f-functions are defined through the following equation:

$$\bar{\mathbf{D}}(\omega) = \varepsilon_0\left(\varepsilon_l + \chi\frac{3\varepsilon_l(\varepsilon_m-\varepsilon_l)}{(2\varepsilon_l+\varepsilon_m)+(f_m-f_l)B/\varepsilon_0}\right)\bar{\mathbf{E}}(\omega)$$

$$+\left(f_l + \chi\frac{3\varepsilon_l(f_m-f_l)}{(2\varepsilon_l+\varepsilon_m)+(f_m-f_l)B/\varepsilon_0}\right)B\bar{\mathbf{E}}(\omega), \quad (19)$$

where χ is the volume fraction of nanoparticle. Typically, the second term in the denominator is much smaller than the first. Therefore, we can expand the fractures with respect to the magnetic field. Again accounting for Eq. (8) and resorting the B-dependent terms in Eq. (18), we can rewrite Eqs. (18) and (19) in the first approximation with respect to the field as

$$\varepsilon(\omega) = \varepsilon_l(\omega) + \chi(\varepsilon_m-\varepsilon_l)\frac{3\varepsilon_l}{2\varepsilon_l+\varepsilon_m},$$

$$f(\omega) = f_l(\omega) + \chi(f_m-f_l)\frac{9\varepsilon_l^2}{(2\varepsilon_l+\varepsilon_m)^2}, \quad (20)$$

where for the averaged dielectric and f-functions, we have the following conjugation conditions: $\varepsilon(-\omega)=\varepsilon^*(\omega)$, $f(-\omega)=f^*(\omega)$. The symbols "+" and "−" correspond to different signs in the exponent $\bar{\mathbf{E}}=E_0\exp(\pm i\omega t)$. By expanding formula 3.6(a) in [10] over the volume fraction of nanoparticles χ, as the first approximation, we obtain the same Eq. (20). This confirms that two different approaches give the same results.

Using the experimental data of [7] summarized in Table 1, we can numerically analyze Eq. (20). Figures 3 and 4 show that the dielectric and f-functions for nanocomposites have well-pronounced resonance peaks, which are redshifted as the refractive index of the matrix increases. For some materials with very large refractive indices, like gallium (III) phosphide, $n=3.5$, and silicon, $n=4.01$, the peak is expected to shift to the visible light region. As follows from the Drude model, bulk metals should not have these resonances; these resonances appear only in nanocomposites. The reason is that the specific electrostatic interactions of metal nanoparticles with the surrounding dielectric matrix lead to a significant increase in the electric field inside the particle [see Eq. (12)]. This in turn affects the polarization of the material [see Eq. (14)]. The calculations are done within the optical range of wavelength, but the tendency is expected to be the same for microwave frequencies. In order to shift the resonance toward the longer wavelength, it is better to use a matrix with a greater refractive index; for example, chalcogenide glasses can be used as the nanoparticle carriers [29–31].

5. FARADAY EFFECT IN NANOCOMPOSITES

Introducing a system of coordinates with the z-axis parallel to the direction of light propagation, we can represent the electric and magnetic fields of light ($\mathbf{E}$- and $\mathbf{B}$-vectors of light) as $\mathbf{E}=\mathbf{a}\exp[\pm i(\omega t-\mathbf{k}\cdot\mathbf{r})]$ and $\mathbf{B}=\mathbf{b}\exp[\pm i(\omega t-\mathbf{k}\cdot\mathbf{r})]$, where $\mathbf{B}$ is the magnetic flux density, and $\mathbf{k}$ is the wave vector directed along the z-axis, and ω is the wave frequency. Equations (18) and (19) can be immediately applied for the analysis of the Faraday effect. Consider first the minus-wave, $\mathbf{E},\mathbf{B}\propto\exp[-i(\omega t-\mathbf{k}\cdot\mathbf{r})]$. The Maxwell equations, $i\mathbf{k}\times\mathbf{E}=i\omega\mathbf{B}$ and $i\mathbf{k}\times\mathbf{B}=-i\omega\mu_0\mathbf{D}$, can be rewritten as $(k_ik_j-k^2\delta_{ij}+\mu_0\omega^2\varepsilon^c_{ij})E_j=0$, with $i=x,y,z$, or

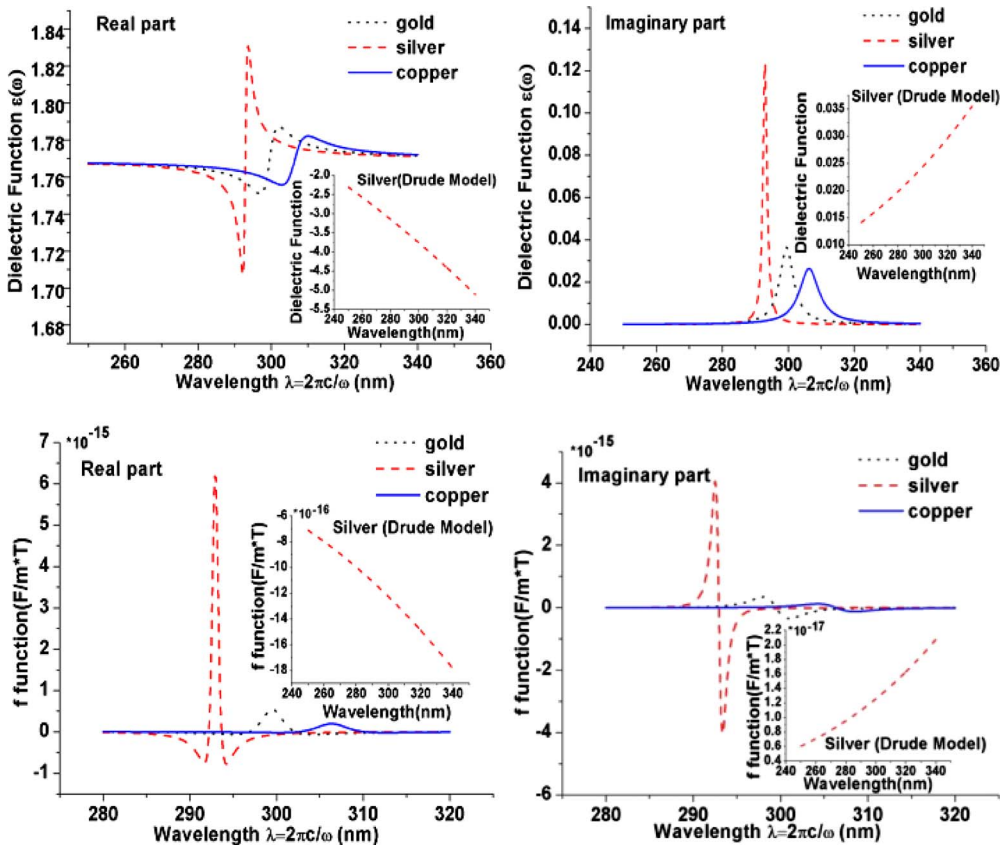

Fig. 3. (Color online) Resonance behaviors for real and imaginary parts of the dielectric function and coupling f-function for different water-based ($\varepsilon_l = n^2 = 1.77$, $f_l = 6.3 \times 10^{-18}$ F m^{-1} T^{-1}) colloids with gold, silver, and copper nanoparticles. Volume fraction of nanoparticles is $\chi = 0.0001$ (the insets show the corresponding dielectric and f-functions for bulk silver).

$$(-k^2 + \mu_0\varepsilon_0\omega^2\varepsilon(\omega))E_x + i\mu_0\omega^2 f(\omega)B_z E_y = 0, \quad (21)$$

$$(-k^2 + \mu_0\varepsilon_0\omega^2\varepsilon(\omega))E_y - i\mu_0\omega^2 f(\omega)B_z E_x + i\mu_0\omega^2 f(\omega)B_x E_z = 0, \quad (22)$$

$$\mu_0\varepsilon_0\omega^2\varepsilon(\omega)E_z - i\mu_0\omega^2 f(\omega)B_x E_y = 0. \quad (23)$$

When the magnetic field $\mathbf{B}$ is directed along the z-axis perpendicularly to the nanocomposite film, the second term in Eq. (23) disappears. Therefore, the z-component of electric field is zero; hence, the $\mathbf{E}$-vector oscillates in the x-y plane (i.e., perpendicularly to the external magnetic field). Therefore, the $\mathbf{k}$-vector of light is perpendicular to the electric field. All these conclusions are applicable for plus-waves $\mathbf{E}, \mathbf{B} \propto \exp[i(\omega t - \mathbf{k}\cdot\mathbf{r})]$ as well.

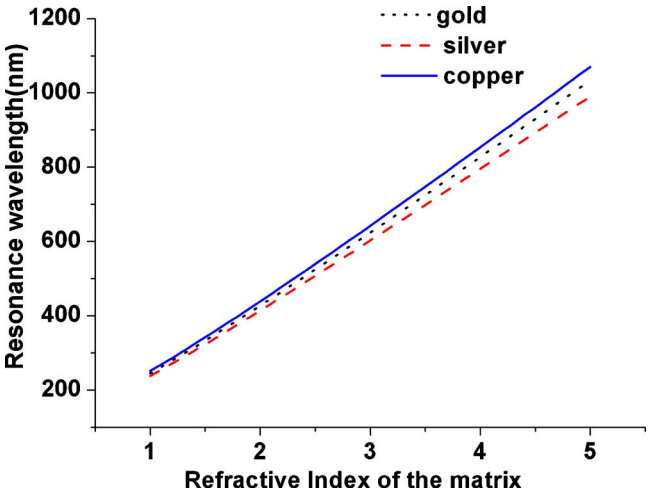

Fig. 4. (Color online) Dependence of the resonance wavelength $\lambda = 2\pi c/\omega$ for imaginary part of dielectric function as a function of refractive index of the matrix.

It appears that the external magnetic field changes the dispersion relation:

$$k_\pm^2 = \omega^2\mu_0(\varepsilon_0\varepsilon(\pm\omega) + f(\pm\omega)B). \quad (24)$$

That is, the plus- and minus-waves have different wave vectors:

$$k_\pm = \frac{\omega}{c}\sqrt{\varepsilon(\pm\omega) + f(\pm\omega)B/\varepsilon_0} = \frac{\omega}{c}\sqrt{\varepsilon' \pm f'B/\varepsilon_0 + i(f''B/\varepsilon_0 \pm \varepsilon'')}, \quad (25)$$

where c is the speed of light. The plane of light polarization is defined as "the plane passing through the $\mathbf{E}$- and $\mathbf{k}$-vectors" [22,32]. When the light hits the sample, the plane of light polarization rotates, and the angle of rotation θ is defined through the equation $\tan\theta = (E_y^+ + E_y^-)/(E_x^+ + E_x^-)$ [22]. Separating the real and imaginary parts in the wave vectors, $k_\pm = k'_\pm + ik''_\pm$, we see from Eq. (25) that the following two approximations: $k'_+ \approx k'_- + O(\omega f'B/c\varepsilon_0)$ and $k''_+ = -k'' \approx -k''_- + O(\omega f''B/c\varepsilon_0)$ hold true. Therefore, for the light wave exiting a nanocomposite film of thickness d, we can approximate the $\mathbf{E}$-vector as

$$\bar{\mathbf{E}} = \bar{\mathbf{E}}_+ + \bar{\mathbf{E}}_- = 2ae^{-k''d}e^{i(k'_+ - k'_-)d/2}\cos\left(\omega t - \frac{k'_+ + k'_-}{2}d\right). \tag{26}$$

Therefore, the angle of rotation is given by the formula

$$\tan\theta = \frac{\bar{E}_y}{\bar{E}_x} = \tan\frac{(k'_+ - k'_-)d}{2}, \quad \text{or} \quad \theta \approx \frac{\omega B d}{2\varepsilon_0 c}\mathrm{Re}\left[\frac{f(+\omega)}{\sqrt{\varepsilon(+\omega)}}\right]. \tag{27}$$

Accounting for the definition of dielectric and f-functions (18) and (19), we observe that the Faraday effect can be significantly enhanced by adding nanoparticles from noble metals. As shown in Fig. 5, even very small volume fraction of these nanoparticles significantly increases the angle of Faraday rotation. Moreover, we predict two resonances. These resonances can be clearly seen if we neglect the dissipation, $\gamma=0$. One resonance is associated with the plasmon frequency ω_r, when the denominator of f-function [see Eq. (20)] goes to zero. The plasmon frequency ω_r can be obtained as the root of the following equation: $2\varepsilon_l(+\omega_r)+\varepsilon_m(+\omega_r)=0$. Another peak in Fig. 5 is associated with the root of equation $\varepsilon(+\omega_f)=0$. This frequency can be calculated analytically as $\omega_f=\omega_r+\chi\Omega$, where

$$\Omega = -3(\varepsilon_m(\omega_r) - \varepsilon_l(\omega_r))\{d[2\varepsilon_l(\omega) + \varepsilon_m(\omega)]/d\omega|_{\omega=\omega_c}\}^{-1}. \tag{28}$$

Thus, the position of the second peak linearly depends on the nanoparticle concentration. This dependence is clearly seen in Fig. 6. We therefore conclude that the Faraday effect contains wealthy information about the nanocomposite; it can predict the plasmon resonance, as well as the concentration of the nanoparticles in the composite. Also, as seen from Fig. 5, the sign of the angle of rotation can be changed near the plasma resonance. This property is attractive for engineering applications, especially for making tunable Faraday isolators and switchers, and many other optoelectronic fiber-based devices [33–35].

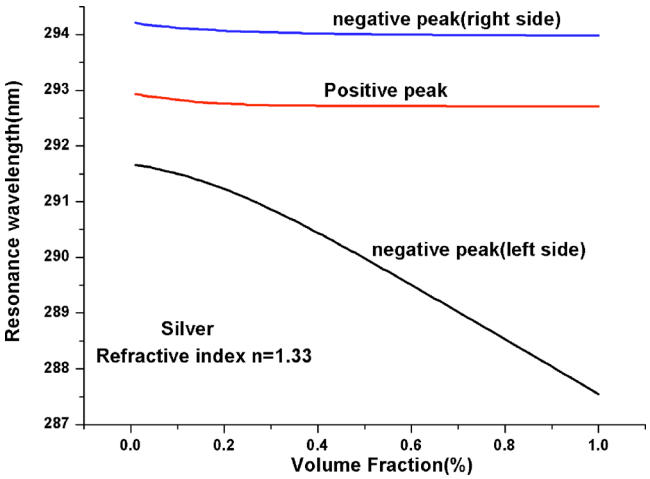

Fig. 6. (Color online) Resonance wavelengths of the Verdet function in the nanocomposites. The volume fraction of nanoparticles is ranged between $0.0001<\chi<0.01$. Observe that the left negative peak depends on the nanoparticle concentration almost linearly.

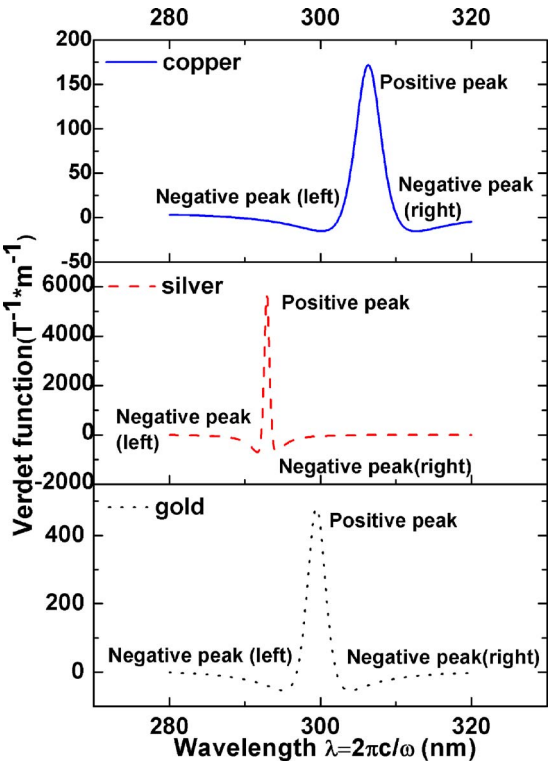

Fig. 5. (Color online) Resonance behavior of Verdet function in nanocomposites.

It should be noted, however, that the Faraday effect in nanocomposites is difficult to observe because of a strong shielding of the electromagnetic radiation in the samples. This shielding is manifested through the strong exponential decay of the E-field [see Eq. (26)]. Based on Eq. (26), the initial and final amplitudes of the E-field can be introduced as $2a$ and $2a\exp(-k''d)$, respectively. Therefore, we can introduce the transmission coefficient of light as $T=\exp(-k''d)$. As seen from Fig. 7, because of a somewhat similar resonance behavior of the imaginary part of the wave number "k," one expects to have significant light ab-

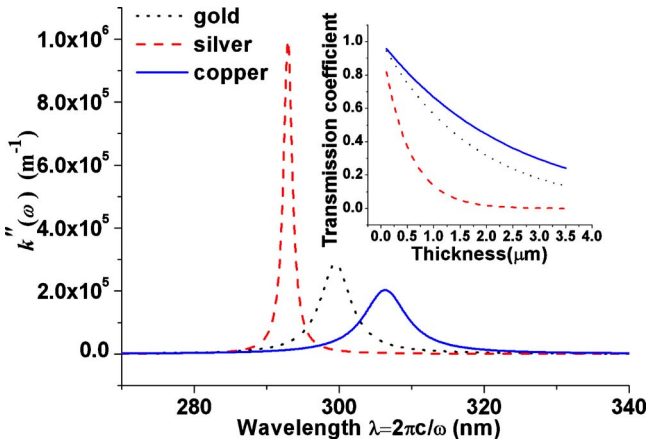

Fig. 7. (Color online) Imaginary part of the wave number (water-based colloids, $\chi=0.0001$). The inset shows the dependency of the transmission coefficient on the sample thickness. The transmission coefficient was calculated at the peak wavelength.

sorption at the resonances. Therefore, the observation of the resonance behavior of the Faraday effect is limited by the sample thickness; one needs to choose the sample thickness, which could provide as large transmission coefficient as possible. In this set of three metals, copper colloid seems to be the best candidate for the enhancement of the Faraday rotation. Although the Verdet function in copper colloid at the peak is smallest among other noble metals, this colloid has the largest transmission coefficient. As a result, the resonance in copper colloid is expected to be most prominent.

6. CONCLUSIONS

This paper is devoted to the theoretical analysis of magneto-optical effects in nanocomposites made of metal non-magnetic nanoparticles embedded into a non-metal matrix. The dispersion properties of metal nanoparticles are described by the Drude model. This model does not account for the effects of nanoparticle size, yet predicts a resonance behavior of the nanoparticle spectra. We used the microscopic definition of magnetization and obtained the magnetic properties of non-magnetic metals within the Drude model. It has been demonstrated that in the absence of external magnetic field, a bulky metal can be magnetized in the circularly polarized light beam, and the magnetization depends on the direction of rotation of the light wave. However, this effect is difficult to observe.

We analytically solved the quasi-static field equations for the case when the size of the particle is small compared to the wavelength of light. Because of the new gyromagnetic constitutive equation, the formulation of this basic problem is different from that of conventional plasmonics [1–6]. We explicitly showed that the particle polarization is a nonlinear function of the external magnetic field, and it has a resonance behavior. This resonance effect is caused by a significant enhancement of the electric field within the nanoparticle. As a result, the light-induced magnetization of the non-magnetic metal nanoparticles can be significantly increased. The external magnetic field alters the particle magnetization, and when the fields are weak this change in magnetization linearly depends on the magnetic field. The proposed theory has been applied to the analysis of Faraday effects in nanocomposites with small fractions of nanoparticles when the materials are still transparent. In order to find a constitutive equation for the electric displacement field, we used the Rayleigh theory of dilute suspensions. We predicted a resonance behavior of the Verdet constant in nanocomposites and its dependence on concentration, sample thickness, and magnetic field. Such composites might be of value for optical sensors and other devices where magneto-optical effects need to be significantly enhanced. While the Drude model is attractive because of its simplicity and clear physical picture of electron transport, more sophisticated models should be called in for a quantitative description of size effects in nanocomposites.

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