Theory of Faraday rotation in granular magnetic materials

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We present a formalism to treat Faraday rotation (FR) in composite media. The method is not limited to low concentrations of magnetically active inhomogeneities and is thus useful for understanding the magneto-optical properties of such inhomogeneous media over the whole range of concentrations. It involves the calculation of an effective magnetic-field-dependent dielectric tensor \( \epsilon_r \), using a tensor effective-medium approximation. To illustrate the technique, we present the results of several model calculations involving both paramagnetic and ferromagnetic metals. We find that such materials have strongly enhanced FR over a wide range of frequencies. The effects of particle shapes on FR are discussed.

I. INTRODUCTION

More than 150 years ago, Michael Faraday discovered that when linearly polarized light is passed through flint glass in an applied magnetic field, its plane of polarization is rotated.\(^1\) It was found that the angle of rotation \( (\theta_f) \) could be expressed in the form

\[
\theta_f = \frac{VBI}{1 + \frac{B}{N} \cos \phi}
\]

where \( B \) is the magnetic induction, \( I \) is the thickness of the glass, and \( V \) is Verdet's constant, which depends on the material and on the frequency of the light. This effect is now well known as Faraday rotation (FR). It is the basis for numerous devices based on a magnetooptical principle, and it also widely used as an experimental tool for measuring material properties.\(^2\) The new generation of fiber-optic magnetometers, for example, operates on a magnetooptical principle.\(^3\) To produce a magnetometer of maximum sensitivity, one requires a sensing element with the highest possible Verdet's constant.

In this paper, we consider Faraday rotation in granular Faraday-active materials. In the ease of low concentration of Faraday-active materials embedded or suspended in a Faraday-inactive host, Hui and Stroud\(^4\) employed the Maxwell-Garnett approximation and predicted an enhancement in Faraday rotation in the vicinity of the surface plasmon resonance frequency of the granules. Experimental evidences confirming such prediction were reported by Yusuf, Rousan, and El-Ghanem in a dilute Fe\(_3\)O\(_4\) particle magnetic fluid.\(^5\) The confirmation of predictions in Ref. 4 could mean important potential applications of granular materials in magneto-optical devices. In this paper, we generalize the theory in Ref. 4 to treat Faraday-active granular materials over the whole range of concentrations. Our present theory employs the well-known effective-medium approximation (EMA), which has successfully been used in obtaining other transport and optical properties in composite materials.\(^6\) Using this treatment, together with a mean-field decoupling reminiscent of the Weiss molecular field theory in magnetism, one can readily study composites over the entire range of concentrations of Faraday-active constituents. The results in Ref. 4 can be obtained as the dilute limit of the present theory.

We will consider both insulator-normal metal composites and insulator-ferromagnetic metal systems over a broad range of concentrations. As will be shown, such materials have strongly enhanced Verdet's constants over a wide range of frequencies. We also consider the effects of particle shapes. By comparing results from calculations based on needlelike particles, spherical particles, and plateletlike particles, we show that the frequencies at which enhancements occur depend on the shape of the particles, and that needlelike particles give the strongest enhancement. Preliminary results of the present theory have been reported previously.\(^7\)

The remainder of this paper is organized as follows. Section II describes a formal generalization of the EMA to treat the effective frequency-dependent dielectric function for a composite in a magnetic field. Section III presents two applications of this formalism: first, to a composite of insulator and free-electron metal in an applied magnetic field; next, to a composite of insulator and ferromagnetic metal. A discussion of our results follows in Sec. IV.

II. FORMALISM

The effective-medium approximation has often been applied to treat the optical properties of composites.\(^6\) In these studies, the optical response of the composite is described by an effective dielectric function \( \epsilon_r \), which can be calculated within the EMA as a function of the constituent dielectric functions, concentrations, and particle shapes. If the constituents are isotropic, and if the composite geometry is such that the composite is macroscopically isotropic, then \( \epsilon_r \) is generally a scalar, that is, a multiple of the \( 3 \times 3 \) unit tensor \( I \). In the presence of a magnetic field—whether applied or in-
ternally generated within a ferromagnetic particle—this isometry is destroyed and off-diagonal elements are introduced into the dielectric matrix. These off-diagonal elements describe the magneto-optical response of the system.

We consider a binary composite containing a volume fraction $1 - f$ of Faraday-inactive insulator and fraction $f$ of Faraday-active particle. The latter is assumed to have dielectric tensor

$$
\varepsilon = \begin{pmatrix}
\varepsilon_i (B, \omega) & i \gamma_i (B, \omega) & 0 \\
-i \gamma_i (B, \omega) & \varepsilon_i (B, \omega) & 0 \\
0 & 0 & \varepsilon_2 (B, \omega)
\end{pmatrix},
$$

(2)
in an applied magnetic field $H_0 = H_0 \hat{z}$. The elements of $\varepsilon$ depend on the local magnetic induction $B$ within the particle, which may differ from $H_0$, and on the angular frequency $\omega$. We assume that the magnetic permeability tensor $\mu = 1$, where $1$ is the unit tensor; this will normally be a good approximation at optical frequencies. The Faraday-inactive component is assumed to have an isotropic dielectric tensor $\varepsilon_i = \varepsilon_i'$. 

To simplify our calculations, we assume that the magnetically active grains are uniaxial ellipsoids with a short axis $a$ and a long axis $b$, all identically aligned, and with their principal axes parallel to the $z$ axis. We also assume that the external magnetic field is oriented parallel to the axis of symmetry. Although this geometric form may not be applicable to some cases of interest, it can easily be generalized to treat more complicated geometries, if desired.

Our goal is to calculate the effective dielectric tensor $\varepsilon_e$ of the composite. With the stated assumptions on the geometry of the composite, this dielectric tensor can be written in the same form as Eq. (1):

$$
\varepsilon_e = \begin{pmatrix}
\varepsilon_1 (B, \omega) & i \gamma_1 (B, \omega) & 0 \\
-i \gamma_1 (B, \omega) & \varepsilon_1 (B, \omega) & 0 \\
0 & 0 & \varepsilon_2 (B, \omega)
\end{pmatrix},
$$

(3)
where the unknown elements $\varepsilon_1$, $\varepsilon_2$, and $\gamma_1$ are to be determined in terms of the properties of the host and of the inclusions, using the effective-medium approximation. For the present problem, the EMA self-consistency condition takes the form,

$$
(i - \Gamma \delta e)^{-1} \delta e = 0,
$$

(4)
where

$$
\delta e = \varepsilon_i - \varepsilon_e,
$$

(5)
with $\varepsilon_i = \varepsilon_i'$ or $\varepsilon_i'$. The triangular brackets denote a volume average. The tensor $\Gamma$ is defined by a surface integral:

$$
\Gamma_{ij} = -\oint_{S'} \frac{\partial}{\partial x_j} G(x - x') n_j d^2 x',
$$

(6)
where $S'$ is the surface of a grain centered at the origin, $n_j$ is a Cartesian component of a unit normal vector $\hat{n}$ pointing outward from $S'$, $x$ is a point located within the grain, and the Green's function $G(x - x')$ is given by

$$
G(x - x') = \frac{1}{4\pi \epsilon_e (\epsilon_2)} \left[ \frac{(x - x')^2}{\epsilon_1} + \frac{(y - y')^2}{\epsilon_2} + \frac{(z - z')^2}{\epsilon_2} \right]^{-1/2}.
$$

(7)

For ellipsoidal grains, only the diagonal elements of $\Gamma$ are nonzero. For example, if the grains are spherical, $\Gamma$ has nonzero elements

$$
\Gamma_{zz} = -\frac{1}{\epsilon_2 A} \left[ 1 - \frac{1}{\lambda} \right]^{-1/2} \sin^{-1} \sqrt{\frac{2}{\lambda}}
$$

(8)

$$
\Gamma_{xx} = -\frac{1}{\epsilon_2 A} \left[ \Gamma_{zz} + (\epsilon_2 - \epsilon_1) \lambda \right]^{-1/2} \sin^{-1} \sqrt{\frac{2}{\lambda}},
$$

(9)

$$
\Gamma_{yy} = \Gamma_{zz},
$$

(10)
where $\lambda = 1 - \epsilon_1 / \epsilon_2$.

If we assume that $\Gamma$ is diagonal, and that $\Gamma_{zz} = \Gamma_{yy}$, then we obtain from Eqs. (4) a set of three self-consistent equations which can be solved for $\epsilon_1$, $\epsilon_2$, and $\gamma_e$:

$$
f \left[ 1 - (\epsilon_1 - \epsilon_2) \Gamma_{zz} \right] (\epsilon_1 - \epsilon_2) + (\gamma_e - \gamma_e) \Gamma_{xx}
$$

$$
\left[ 1 - (\epsilon_1 - \epsilon_2) \Gamma_{xx} \right] + (\gamma_e - \gamma_e) \Gamma_{yy} = 0,
$$

(11)

These equations can, in principle, be solved numerically. However, simplifications can be achieved under certain situations. For example, in the limit of low magnetic fields, the diagonal elements of $\Gamma$ will be identical through first order in $B$. The integrals for the elements of $\Gamma_{xx}$ can be readily evaluated in this limit, with the results as

$$
\Gamma_{xx} = -L_{xx}/\epsilon_{xx},
$$

(12)
where the factors $L_{xx}$ are geometry-dependent quantities given by the integrals

$$
L_{zz} = \frac{1}{2} \frac{a^2 b}{\epsilon_1} \int_0^\pi \frac{d\phi}{(\xi + b^2)^2 R_2},
$$

(13)

$$
L_{yy} = \frac{1}{2} \frac{a^2 b}{\epsilon_1} \int_0^\pi \frac{d\phi}{(\xi + a^2)^2 R_2},
$$

and

$$
R_2 = (\xi + a^2 - \sqrt{\xi + a^2 + b^2})^2. \quad \text{The three self-consistency conditions (11) hence take the form}
$$

$$
0 = f \frac{\epsilon_1 - \epsilon_{xx}}{\epsilon_1 + \frac{1}{2}(1 - L)(\epsilon_1 - \epsilon_{xx})} + (1 - f) \frac{\epsilon_0 - \epsilon_{xx}}{\epsilon_0 - \epsilon_{xx}},
$$

$$
+ \frac{\gamma_e - \gamma_e}{\epsilon_0} - \frac{\epsilon_{xx}}{2(1 - L)(\epsilon_1 - \epsilon_{xx})},
$$

(11)

$$
0 = f \frac{\gamma_e - \gamma_e}{\epsilon_0 + \frac{1}{2}(1 - L)(\epsilon_1 - \epsilon_{xx})} + (1 - f) \frac{\epsilon_0 - \epsilon_{xx}}{\epsilon_0 - \epsilon_{xx}},
$$

(11)
0 = \frac{\epsilon_1 - \epsilon_2}{\epsilon_2 + L(\epsilon_1 - \epsilon_2)} + \left(1 - f\right) \frac{\epsilon_0 - \epsilon_2}{\epsilon_2 + L(\epsilon_0 - \epsilon_2)}, \tag{14}

where we have used the relations $L_{zz} = L$, and $L_{xx} = L_{yy} = \frac{1}{2}(1 - L)$. \(^{9}\)

Once $\epsilon_1$, $\epsilon_2$, and $\gamma$, are determined, the quantity $\theta_f$, denoting the angle by which the plane of polarization is rotated per unit sample length, can be expressed as: \(^{10}\)

$$\theta_f = \frac{\omega}{c^2} \left[ \epsilon_1 (B, \omega) \pm \gamma_e (B, \omega) \right]. \tag{15}$$

where

$$k^2 = \frac{\omega^2}{c^2} \left[ \epsilon_1 (B, \omega) \pm \gamma_e (B, \omega) \right]. \tag{16}$$

Note that $\theta_f$ generally is a complex number since $\epsilon_1$ and $\gamma_e$ are complex. $\Re(\theta_f)$ represents the rotation of the plane of polarization, while $\Im(\theta_f)$ determines the extent to which the incident plane-polarized light becomes elliptically polarized on passing through the sample.

Equations (11) and (14)–(16) are our main results. They are quite general: given any specific microscopic model for dielectric function $\epsilon$ of a Faraday-active inclusion, the effective dielectric tensor and the Faraday rotation of the composite can be found by solving these equations.

### III. MODEL CALCULATIONS

We first consider a composite of free-electron metal and insulator, subjected to an external magnetic induction $B = B \hat{z}$. We assume that the metallic grains can be described by a Drude dielectric function. Then the dielectric tensor of the metal grains will be given by Eq. (2) with

$$\epsilon_1 = 1 + \frac{(1 - i\omega\tau)\omega^2}{\omega(1 - i\omega\tau)^2},$$

$$\epsilon_2 = 1 + \frac{\omega^2}{\omega(1 - i\omega\tau)},$$

$$\gamma = \frac{\omega\epsilon_1}{\omega(1 - i\omega\tau)}.$$ \tag{17}

Here $\omega_\rho = \sqrt{4\pi ne^2/m}$ is the plasma frequency, $\omega_\tau = eB/mc$ is the cyclotron frequency, and $\tau$ is a characteristic relaxation time defined in the Drude model. In the weak-field limit, $\omega_\tau < 1$, we have $\epsilon_1 \approx 1 + \omega_0^2\tau^2/\omega(1 - i\omega\tau)$, and $\gamma \approx \omega_\tau/\omega(1 - i\omega\tau)$ to first order in the field. Substituting these approximations into Eq. (14) yields an exactly solvable quadratic equation for $\epsilon_1$. To the same (first) order in the applied field, $\gamma_e$ is given by

$$\gamma_e = f' \left[ \epsilon_1 \pm \frac{\epsilon_0 - \epsilon_1}{2\epsilon_1} \right]. \tag{18}$$

Using Eq. (15), the angle of Faraday rotation in the small field limit is thus given by

$$\theta_f = \frac{\omega f}{2c\epsilon_1} \left[ \epsilon_1 \pm \frac{\epsilon_0 - \epsilon_1}{2\epsilon_1} \right]. \tag{19}$$

The frequency dependence of $\theta_f(\omega)$ is shown in Fig. 1 for several different filling fractions of metal. In these calculations, we have assumed that $\omega_\tau = 100$, typical of free electrons such as Al at room temperature, $\omega_\tau = 10^{-6}$, corresponding to an applied field of 1 Oe for Al, and $L = \frac{1}{2}$, corresponding to spherical metal grains. Both the real and imaginary parts of $\theta_f(\omega)$ have resonance peaks near $\omega = \omega_\rho/\sqrt{3}$, the frequency of the surface plasmon resonance of the small metal particles. Our calculated enhancement shows a surprisingly good agreement with that of the experiment.\(^3\) This enhancement results from a greatly increased local electric field either within or just outside the metal grains at special frequencies, relative to the applied field. The effect is most obvious in the dilute limit ($f = 0.03$).\(^4\) At higher concentrations of metal, the enhancement peaks still remain, but are considerably broadened, because of the increased disorder in the composite.

In Fig. 2 shows the dependence of $\theta_f(\omega)$ on the shapes of the metallic particles. We have plotted $\theta_f(\omega)$ at a filling fraction $f = 0.03$ for three different shape factors: $L = 0.01$ (needlelike particles), $L = \frac{1}{2}$ (spheres), and $L = 0.99$ (pla-
and D is the demagnetization tensor of the grain. For the geometry we have assumed here, D is diagonal with nonzero matrix elements $D_{xx} = L_{xx}$, and M is parallel to the applied magnetic field, $\mathbf{M} = M \mathbf{z}$. Combining these formulas together, we finally obtain

$$\mathbf{B} = \mathbf{H}_0 + 4\pi(1 - L)\mathbf{M}, \quad (22)$$

where we have used the fact that the grains are oriented with their axes of symmetry parallel to $z$ and have used the notation $L_{xy} = L_{yy} = L_{xz} = \frac{1}{2}(1 - L)$.

In order to carry out a simple model calculation, we require an approximation for the complex dielectric function of a ferromagnetic metal. For simplicity, we model this material by a Drude dielectric function in which the electrons feel an effective magnetic field which includes the local (molecular) field as well as the applied field. Then Eqs. (17) are still valid for the ferromagnetic inclusions, provided that we replace $\omega$ by

$$\omega' = \omega_c [1 + 4\pi(1 - L)\chi'], \quad (23)$$

where the “susceptibility” $\chi$ is defined as $\chi = M / H_0$, the ratio of magnetization to applied field. In a random composite, this ratio will certainly vary from one magnetic particle to another. In order to derive Eq. (23), we have made the rather crude approximation that the magnetic field within each particle can be approximated by the value that field would have, if the particle were isolated.

To estimate the field within the ferromagnetic inclusions, we approximate the susceptibility appearing in Eq. (23) by

$$\chi = \frac{C}{T - T_c}, \quad (24)$$

for $T > T_c$, and by

$$\chi = \frac{M_0}{H_0} (1 - \nu T^{3/2}), \quad (25)$$

for $T < T_c$. Here $T_c$ is the Curie temperature, $M_0$ is the zero-temperature magnetization, $C$ is the Curie constant, and $\nu$ is proportional to the spin-wave stiffness constant of the material. Equation (24) represents the Curie–Weiss approximation for the susceptibility in the paramagnetic region. Equation (25) is the spin-wave approximation for the susceptibility below the Curie temperature and becomes exact at sufficiently low temperatures. In Ni, for example, $T_c = 627$ K, $M_0 = 510$ G, $C/T_c = 0.32$, and $\nu = 7.5 \times 10^{-6}$ K$^{-3/2}$. Note that, although $\chi$ appears to diverge in the limit $H_0 \to 0$, according to Eq. (25), this apparent divergence leads only to a finite shift in $\omega'$ relative to $\omega_c$, because $\omega_c = eH_0/m\nu$ varies linearly with $H_0$.

Figure 3 displays $\theta_J(\omega)$ for a number of different metal filling fractions both above and below the percolation threshold $J$. This threshold, which occurs for spherical particles at $f = \frac{1}{3}$ within the EMA in three dimensions, corresponds to the point at which a ferromagnetic metal first forms a connected path extending across the entire sample. The calculations are carried out at a temperature $T/T_c = 0.5$ (corresponding to $T_c \approx 300$ for Ni); $\nu = 5 \times 10^{-6}$ K$^{-3/2}$, and $M_0/H_0 = 100$. At all concentrations, Fig. 3 shows that both Re($\theta_J$) and Im($\theta_J$) have huge

FIG. 2. Same as Fig. 1, but plots are carried out for a metallic filling fraction 0.03 and three different metal grain shapes, corresponding to depolarization factors $L = 0.01$ (needles), $L = 1/3$ (spheres) and 0.99 (plates).

telike particles). In all cases, the particles are assumed to be oriented parallel to the applied magnetic field. The frequency of the enhancement peaks, like that of the surface plasmon, depends on particle shape. Figure 2 suggests that, of the three shapes considered, the needlelike metal particles result in the strongest enhancement.

We turn next to a more complex example of considerable practical interest, namely, the effective dielectric function and Faraday rotation of a composite of ferromagnetic metal and insulator. Here we assume that the composite is subjected to an applied magnetic field $H_0 = H_0 \mathbf{z}$. The Faraday rotation in this case will be strongly influenced by the temperature-dependent magnetization of the ferromagnetic metal grains, denoted by $\mathbf{M} = \mathbf{M}(T)$, where $T$ is the absolute temperature. Within a metal grain, the magnetic induction takes the form

$$\mathbf{B} = \mathbf{H} + 4\pi\mathbf{M}, \quad (20)$$

where the local field $\mathbf{H}$ can be expressed as

$$H_{\text{local}} = H_0 - 4\pi\mathbf{D}\mathbf{M}, \quad (21)$$

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peaks near $\omega = \omega_p/\sqrt{3}$. These same peaks also appear in composites of free-electron metal and insulator, but in the ferromagnetic case, the rotations are at least two orders of magnitude larger. Since for typical Drude metals, $cr \sim 10^{-4}$ cm, this rotation below the Curie temperature can amount to hundreds of degrees per centimeter of sample thickness, depending on the value of the external frequency. Note also that the sign of $\theta_f$ can change at certain frequencies. Finally, we note that, as the concentration of ferromagnetic metal is increased, the position and magnitude of the enhancement peak remain roughly constant, but the peaks broaden considerably. Such a broadening is expected in a disordered system.

Figure 4 illustrates the dependence of $\theta_f$ on particle shape. Several plots of $\theta_f(\omega)$ are shown at fixed volume fraction $f$, but for different values of the depolarization factor $L$. As in the free-electron case, the frequency of the rotation peak depends on $L$; the greatest enhancement is obtained, again, for needlelike particles ($L = 0.01$).

The effects of temperature are shown in Fig. 5, where we display $\theta_f(\omega)$ for spherical ferromagnetic grains in the dilute limit at two temperatures: $t = 1.1$ and 0.5. All other parameters are the same as in Figs. 3 and 4. The enhancement peak appears for both $T > T_c$ ($t = 1.1$) and $T < T_c$ ($t = 0.5$). The rotation in the latter case is, however, two orders of magnitude larger. This increase is a result of the greatly enhanced local magnetic field within the metal grains when they are in the ferromagnetic phase. At certain frequencies, e.g., $\omega > 0.65 \omega_p$, the angle of rotation changes sign as the temperature decreases.

**IV. DISCUSSION**

We have presented in this paper a method for calculating the Faraday rotation in Faraday-active composites and have illustrated its applications in several model systems. Although our model calculations may be oversimplified, they show several interesting features characteristic of granular materials for many experimentally plausible situations. As in other composite calculations of this kind, they assume that the magnetic particles are small in comparison to the wavelength of light.

From our model calculations on ferromagnetic composites, we see there will be no Faraday rotation in zero exter-
ferromagnetic particle was chosen from a distribution, rather than being assumed to have a unique value. Another complication, namely, short-range order in the orientations of magnetic moments of neighboring particles (such as might be seen in superparamagnetic composites) would be more difficult to treat within the present formalism. The present theory can easily be extended to take into account realistic situations such as distributions in shapes and sizes of the particles.

Within the present model calculations, granular ferromagnets can produce substantial rotation of light on transmission. The theory can be slightly modified to calculate the Faraday rotation on reflection. Our theory can be used to predict the optical properties of new ferromagnetic composites, and thus to help in the design of new composites with desired optical rotation in specific frequency regimes. It would also be of great interest to have more systematic experimental studies on the various predictions here in systems such as magnetic fluids.

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