Mie scattering of magnetic spheres

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The Mie scattering intensity of a magnetic sphere has been derived by extending the classical Mie scattering approach to a media where the dielectric constant is no more a real number but a tensor with a gyrotropic form. Using a perturbation method the propagation equations of the electromagnetic field are derived. For an incident plane wave the magnetization effect could be detectable. The Mie scattering intensity is analyzed for special incident wave configurations, in particular, for the case where the magnetic field of the incident plane wave is polarized along the magnetization direction. This magnetization effect is most important for the finger pattern of the backscattering intensity. Magnetic Mie scattering is still significant for a magnetic sphere of radius larger than 10 nm.

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I. INTRODUCTION

Magnetic nanostructures are of growing interest, in particular due to their potential industrial applications to high density magnetic recording media. The conventional magnetic characterization techniques [1] are limited to the study of large clusters or to large ensembles of particles [2]. Then the interpretation of the data is not trivial due the shape and cluster size distribution. A recent experiment on the magnetic anisotropy of a single cobalt nanocluster used a new micro–superconducting quantum interference device (micro-SQUID) set up [3].

The determination of the magnetization of clusters has been the subject of many theoretical [4] and experimental studies [5] . The experimental studies are based on Stern and Gerlach deflection measurements. The aim of the present article is to show that magnetic effects can be detected by Mie scattering experiments. This would provide then an alternative way to determine the magnetization. It is well known that the interaction of an electromagnetic wave with a magnetic sphere is weak. However, as our calculations show the magnetization effects are detectable.

In Sec. II we discuss the model and in Sec. III we report on the influence of the magnetization direction on the Mie scattering intensity profile.

II. THE MODEL

Of course the classical Mie scattering [6] of an electromagnetic wave by a metallic nonmagnetic sphere is controlled by the Maxwell equations of the electromagnetic field $(\vec{E}, \vec{H})$. For a linear, isotropic, and homogeneous medium $\vec{E}$ and $\vec{H}$ are solutions of the following equations:

$$\nabla^2\vec{E}+k^2\vec{E}=\vec{0}, \tag{1}$$

$$\nabla^2\vec{H}+k^2\vec{H}=\vec{0}, \tag{2}$$

where k^2 is equal to $\omega^2\epsilon\mu$. Here, ω is the wave frequency and ϵ and μ are, respectively, the dielectric and magnetic constants of the media which are both scalar numbers.

Let us recall briefly the method developed by Mie to solve the scattering problem. We introduce the incident electromagnetic wave $(\vec{E}_i, \vec{B}_i)$, the scattered fields $(\vec{E}_{sc}, \vec{B}_{sc})$, and the inner fields $(\vec{E}_{in}, \vec{B}_{in})$. These fields have to obey Eqs. (1) and (2). At the boundary between the metallic sphere and the surrounding media we apply the continuity conditions

$$(\vec{E}_i+\vec{E}_{sc}-\vec{E}_{in})\times\vec{e}_r=\vec{0}, \tag{3}$$

$$(\vec{H}_i+\vec{H}_{sc}-\vec{H}_{in})\times\vec{e}_r=\vec{0}, \tag{4}$$

where $\vec{e}_r$ is a vector perpendicular to the sphere .

We assume now that the medium displays a magnetization along the z axis. Then, the dielectric constant ϵ is no more a scalar, but a tensor which has a gyrotropic form

$$\epsilon=\begin{pmatrix} \epsilon_0 & \epsilon_{xy} & 0 \\ -\epsilon_{xy} & \epsilon_0 & 0 \\ 0 & 0 & \epsilon_0 \end{pmatrix}. \tag{5}$$

From symmetry consideration [7] one gets that the off-diagonal terms in ϵ are linear functions of the magnetization M. They represent the magneto-optical Kerr effect contribution to ϵ and are derived from polar Kerr ellipticity and rotation measurement [8]. Note, formally we could write

$$\epsilon_{xy}=A\lambda_{s\text{-}o}M+\cdots, \tag{6}$$

where $\lambda_{s\text{-}o}$ is the spin-orbit coupling constant. In small clusters $\lambda_{s\text{-}o}$ may be larger than in bulk material [9].

Applying a coordinate axis transformation of Eq. (4) to a laboratory frame, ϵ can be rewritten for any arbitrary magnetization direction as

$$\epsilon=\epsilon_0 I+\epsilon'. \tag{7}$$

Here, ϵ' is a matrix proportional to the magnetization M. Moreover, ϵ' is different from zero only in the inner sphere region. Therefore, the wave equations for the electromagnetic fields are written as

$$\nabla^2\vec{E}+\omega^2\epsilon_0\mu\vec{E}=-\omega^2\mu\epsilon'\vec{E}, \tag{8}$$

$$\nabla^2\vec{H}+\omega^2\epsilon_0\mu\vec{H}=-\omega^2\mu\epsilon'\vec{H}. \tag{9}$$

Here, ϵ' can be regarded as a perturbation to ϵ. Thus, it seems natural to apply a perturbation approach. Hence, $\vec{E}$ and $\vec{H}$ are written as

$$\vec{E}=\vec{E}^0+\vec{E}^1+\cdots, \tag{10}$$

$$\vec{H}=\vec{H}^0+\vec{H}^1+\cdots. \tag{11}$$

$\vec{E}^0$ and $\vec{H}^0$ are the fields without the magnetization and given by Eqs. (1) and (2) as in the classical Mie approach. The terms $\vec{E}^i$ and $\vec{H}^i$ result from the magnetization of the sphere in ith order perturbation theory.

Let us investigate the first-order field corrections $\vec{E}^1$, $\vec{H}^1$ (the higher corrections are obtained similarly). It follows that

$$\nabla^2\vec{E}^1+\omega^2\epsilon_0\mu\vec{E}^1=-\omega^2\mu\epsilon'\vec{E}^0, \tag{12}$$

$$\nabla^2\vec{H}^1+\omega^2\epsilon_0\mu\vec{H}^1=-\omega^2\mu\epsilon'\vec{H}^0. \tag{13}$$

In Eqs. (12) and (13), the terms $\omega^2\mu\epsilon'\vec{E}^0$ and $\omega^2\mu\epsilon'\vec{H}^0$ can be regarded as source terms. Therefore, $\vec{E}^1$ and $\vec{H}^1$ can be obtained with the Green function method. Let us denote by $G_0^{(E,H)}(\vec{r},\vec{r}')$ the Green function for $\vec{E}$ and $\vec{H}$ if $M=0$:

$$\nabla^2 G_0^{(E,H)}(\vec{r},\vec{r}')+\omega^2\epsilon^0\mu G_0^{(E,H)}(\vec{r},\vec{r}')=-4\pi\delta(\vec{r},\vec{r}'). \tag{14}$$

Due to the spherical symmetry it is convenient to expand $G_0^{(E,H)}(\vec{r},\vec{r}')$ in terms of spherical harmonics. Thus,

$$G_0^{(E,H)}(\vec{r},\vec{r}')=G_0^{(E,H)}(r,\theta,\phi;r',\theta',\phi')$$
$$=\sum_{l,m}\vec{g}_l(r,r')Y^*_{lm}(\theta',\phi')Y_{lm}(\theta,\phi). \tag{15}$$

From Eq. (13) it follows that $\vec{g}_l(r,r')$ is a solution of

$$\left(\frac{d^2}{dr^2}+\frac{2}{r}\frac{d}{dr}+\epsilon\omega^2\mu-\frac{l(l+1)}{r^2}\right)\vec{g}_l(r,r')=-\frac{1}{r^2}\delta(r-r'). \tag{16}$$

Let a be the radius of the spherical cluster. Then $\vec{g}_l(r,r')$ has formally three different solutions:

(1) If $r<a$ and $r'<a$,

$$g_l(r,r')=j_l(kr_<)[Ah_l^1(kr_>)+Bh_l^2(kr_>)]. \tag{17}$$

(2) If $r>a$ and $r'>a$,

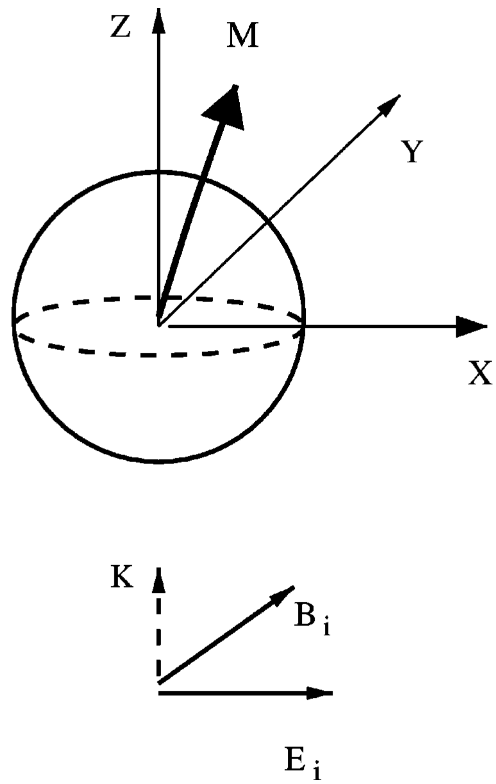

FIG. 1. Mie Scattering geometry of a magnetic spherical cluster. The incident plane wave is propagating along the *z* axis, the electric and magnetic fields are polarized along the *x* and *y* axis. The magnetization *M* has an unspecified direction.

$$g_l(r,r')=h_l^1(k'r_>)[Cj_l(k'r_<)+Dy_l(kr_<)]. \tag{18}$$

(3) For all other cases,

$$g_l(r,r')=Eh_l^1(k'r_>)j_l(kr_<). \tag{19}$$

The coefficients A, B, C, D, and E are derived from the boundary conditions. Thus, the first-order contributions to the fields due to the magnetization are

$$\vec{E}^1(\vec{r})=\int\int\int_{D_S}\epsilon'\omega^2\mu\vec{E}^0_{in}(\vec{r}')G_0^{(E)}(\vec{r},\vec{r}')d\vec{r}', \tag{20}$$

$$\vec{H}^1(\vec{r})=\int\int\int_{D_S}\epsilon'\omega^2\mu\vec{H}^0_{in}(\vec{r}')G_0^{(H)}(\vec{r},\vec{r}')d\vec{r}'. \tag{21}$$

D_S is the inner volume of the sphere. Higher order perturbation contributions to the fields are obtained straightforwardly in a similar way.

III. RESULTS

We apply our model to a cobalt sphere. Note, for radius larger than a critical radius a_{cr} the sphere is no more a monomagnetic domain (for cobalt $a_{cr}=30$ nm). Nevertheless an applied magnetic field may cause a single domain structure and will control the direction of the magnetization. The dielectric tensor is dependent on the frequency of the electro-

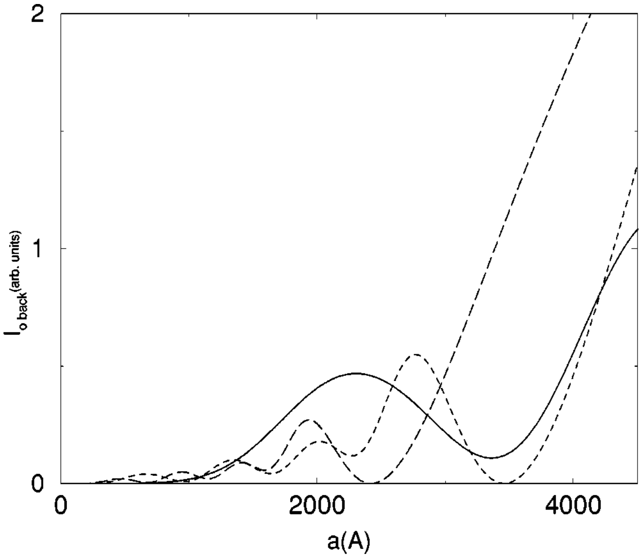

FIG. 2. Without the magnetization effect, the backscattering intensity $I_{o,back}$ in the $-Z$ direction vs the cluster radius a(Å) for different wavelength [energy $E_\omega = 1$ eV (solid line), $E_\omega = 3.5$ eV (dashed line), and $E_\omega = 5$ eV (long-dashed line)], respectively, ϵ_o worth $-29.76 + i40.00$, $-4.592 + i7.778$, and $-1.569 + i5.58$.

magnetic wave. We use the optical constant obtained by Johnson *et al.* [10] and derive the off-diagonal part of the dielectric tensor from the bulk measurement of Kerr ellipticity and rotation done by Zeper *et al.* [11]. The bulk value used are a good first guess for answering if the magnetization effect on the Mie scattering is significant or not. Note, our guess gets better for larger cluster.

The incident electromagnetic field ($\vec{E}_i, \vec{B}_i$) is a plane wave propagating along the z axis with an electric field polarized along the x axis and the magnetic field along the y axis (see Fig. 1). The direction of the magnetization $\vec{M}$ is assumed to be arbitrary. From Eqs. (20) and (21), we note that the field corrections $\vec{E}^1$ and $\vec{H}^1$ involve only integrations over a small volume of the sphere. As a consequence the corrections are weak. Therefore, generally the magnetization has a weak effect on the value of the electromagnetic field and on the total Mie scattering cross section W:

$$W = \frac{1}{2}\mathrm{Re}\int_0^{2\pi}\int_0^{\pi} d\theta d\phi r^2 \sin(\theta)^2 (E_{i\phi}H^*_{sc\theta} - E_{i\theta}H^*_{sc\phi} - E_{sc\theta}H^*_{i\phi} + E_{sc\phi}H^*_{i\theta}). \tag{22}$$

However, the magnetization effect could be detected for special directions of the differential cross section for which the contributions given by $\vec{E}^0$ and $\vec{H}^0$ are small. This is the case for the classical Mie scattering when the size of the cluster is of the same order as the wavelength. Then the backscattering spectra displays some finger pattern. In Fig. 2, the backscattered intensity $I_{o,back}$ in the $-Z$ direction has been plotted vs the cluster radius a(Å) for different wavelengths or energies E_ω of the electromagnetic waves. $I_{o,back}$ is sensitive to these two parameters. For $E_\omega = 5$ eV, $I_{o,back}$ is very small for a radius of 2400 Å.

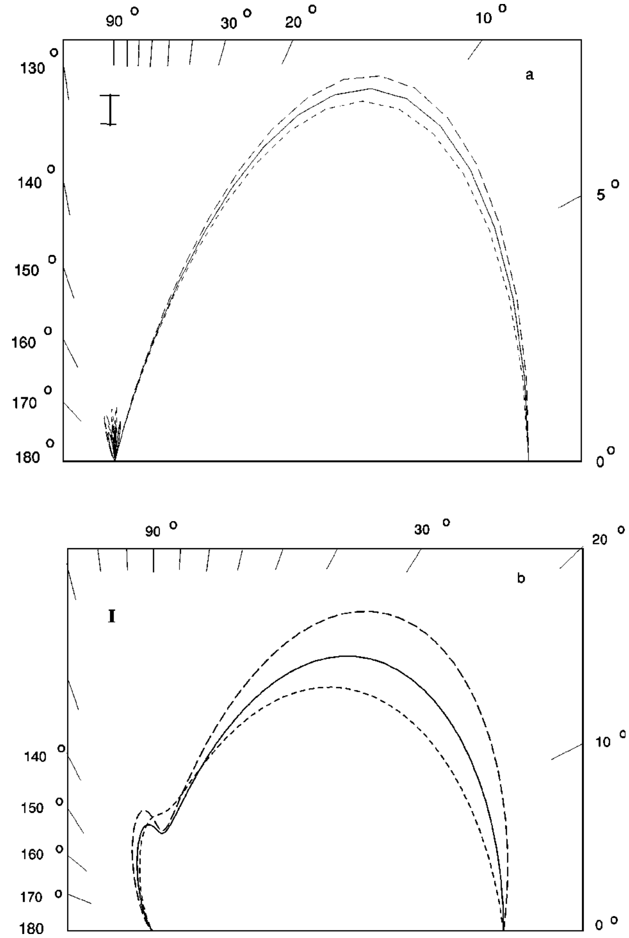

FIG. 3. Results for the angle-dependent magnetic Mie scattering intensity using a first-order calculation. The wave energy is $E_\omega = 5$ eV (1 eV). The angle refers to the polar coordinate in the z-x plane and the axis units are arbitrary. The cobalt sphere has a radius of 2600 Å. The long-dashed (dashed) curve is for a magnetization along the positive (negative) y axis. The solid curve refers to the case when no magnetization is present. We use $\epsilon_o = -29.76 + i40.00$ $(-1.569 + i5.58)$ and $\epsilon_{xy} = -1.92 + i0.41$ $(-0.059 + i0.122)$ in (b) [(a)] for $E_\omega = 1$ eV (5 eV).

Now let us discuss the effect of the magnetization on the Mie scattering. We investigate first the case where the cluster radius is of the order of the magnitude of the wavelength. We choose a value of the radius of 2600 Å such that for $E_\omega = 5$ eV the backscattering intensity is small. Our calculation shows that the effect of the magnetization is small. But it is more important and detectable for special directions of the magnetization. This is the case if the magnetization is essentially along the $\vec{B}_i$ (i.e., along the y axis).

In Fig. 3, we report results for the Mie scattering intensity for different wave energies in the half plane x-z $(X>0)$, i.e., in the plane containing $\vec{E}$ and $\vec{k}$. We use the polar representation and the axis units are arbitrary. The solid line is the classical Mie scattering curve (for $H=0$). We plot also the first-order changes due to the magnetization, when $\vec{M}$ is along the positive y axis (long-dashed curve), and when it is along the negative y axis (dashed curve).

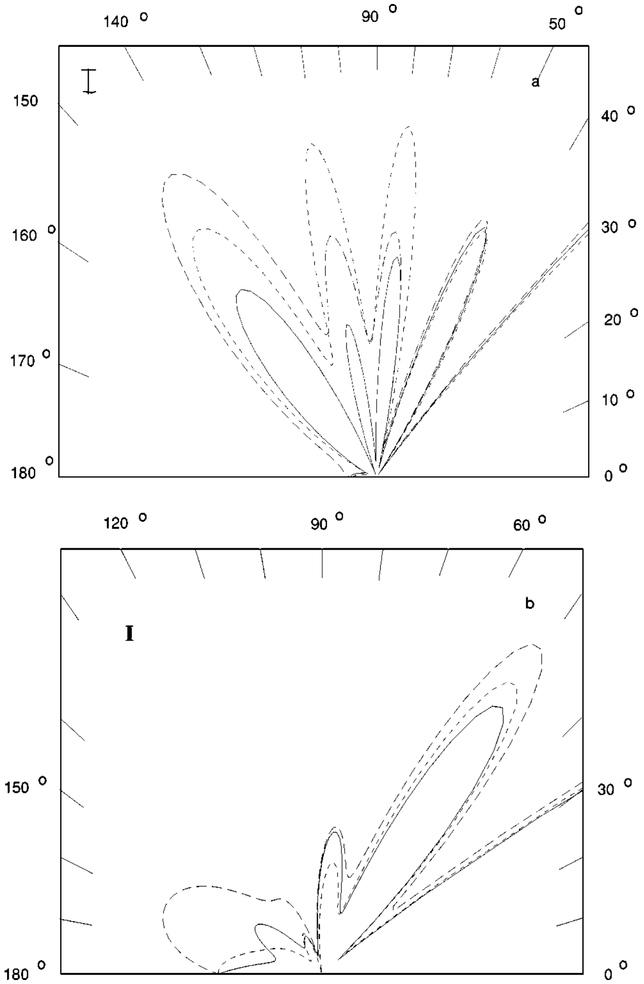

FIG. 4. Finger pattern of the first order calculation of the angle-dependent magnetic Mie backscattering Intensity for a wave energy $E_\omega = 5$ eV (3.5 eV).The angle refers to the polar coordinate in the z-x plane and the axis units are arbitrary. The cobalt sphere has a radius of 2600 Å. The long-dashed (dashed) curve refers to the case where the magnetization is along the positive (negative) y axis. The solid curve refers to the case of $M = 0$. We use $\epsilon_o = -4.592 + i7.778$ $(-1.569 + i5.58)$ and $\epsilon_{xy} = -0.1613 - i0.0733$ $(-0.059 + i0.122)$ in (b) [(a)] for $E_\omega = 3.5$ eV (5 eV).

The effect of the magnetization is maximal when $\vec{M}$ is parallel to the y axis: in fact the boundary conditions Eq. (3) and (4) give some relations on the normal components of the fields which are more affected for this M direction. The forward intensity displays a lobe shape which is increasing (decreasing) when M is along the positive (negative) y axis. The spectra is essentially modified for $\theta = \pi/4$: for $E_\omega = 5$ eV the largest effect is nearly 10% as for $E_\omega = 1$ eV it is 25%. For $\theta = 0$ (or π) the M effect does not occur, the corrections to the field are very weak. The fingering pattern of the backscattered spectra is more modified and complex (Fig. 4). The zeroth-order spectra is no more always located between the ones obtained with $\vec{M}$ parallel or antiparallel to the y axis. Note that for $\theta = \pi/2$ and $E_\omega = 5$ eV the zeroth-order intensity is very small, but the first order one due to magnetization is large.

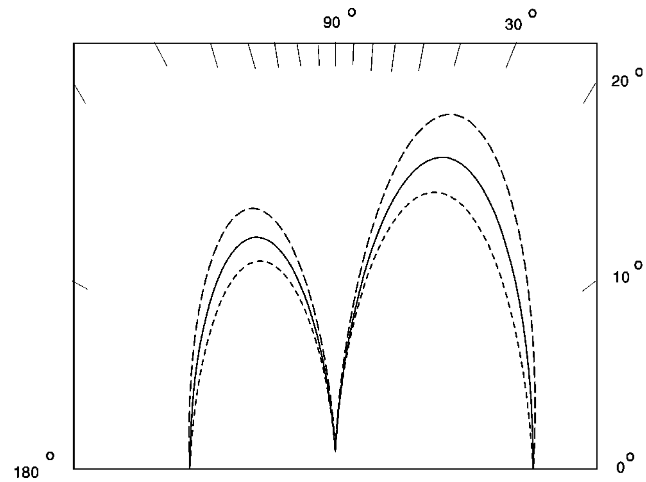

FIG. 5. First order calculation of the angle-dependent magnetic Mie scattering intensity for a wave energy $E_\omega = 5$ eV. The angle refers to the polar coordinate in the z-x plane and the axis units are arbitrary. The cobalt sphere has a radius of 260 Å. The long-dashed (dashed) curve refers to the case where the magnetization is along the positive (negative) y axis. The solid curve refers to $M = 0$. We use $\epsilon_o = -1.569 + i5.58$ and $\epsilon_{xy} = -0.059 + i0.122$.

For smaller cluster radius the finger pattern disappears. The spectra for a radius of 260 Å display a significant effect of the magnetization for $\theta = \pi/4$ or $3\pi/4$ with changes of 25% for $E_\omega = 5$ eV (Fig. 5). For a radius of 150 Å the effect amounts is only to 8% and for 100 Å only to 4%.

IV. CONCLUSION

The present calculation has shown that the Mie scattering from a magnetic sphere results most significantly in the magnetization when the incident wave is a planar electromagnetic wave with $\vec{M}$ along the incident magnetic field axis and the detection plane is determined by the incident electric field and $\vec{k}$. To control experimentally the direction of the magnetization an applied magnetic field can be applied.

For a cluster radius of the same order of magnitude as the light wavelength, the finger pattern of the intensity profile is more affected than the forward lobe shape when $\vec{M}$ is parallel or antiparallel to the incident magnetic field . Even for smaller cluster, this effect is still detectable for a radius larger than 100 Å. For smaller cluster size, it would be interesting to investigate the nonlinear optical properties.

We have seen that for small magnetic spheres magnetism affects the Mie scattering. An important problem is to determine the magnetization from Mie scattering intensity profile. Note, for small clusters the magnetization is size dependent. Hence, for known cluster sizes it is possible to determine the magnetization by fitting the experimental Mie scattering spectra to the calculated ones by using ϵ_{xy}. The analysis should be repeated for different wavelength energies.

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