Radiative corrections to the polarizability tensor of an electrically small anisotropic dielectric particle

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Abstract: Radiative corrections to the polarizability tensor of isotropic particles are fundamental to understand the energy balance between absorption and scattering processes. Equivalent radiative corrections for anisotropic particles are not well known. Assuming that the polarization within the particle is uniform, we derived a closed-form expression for the polarizability tensor which includes radiative corrections. In the absence of absorption, this expression of the polarizability tensor is consistent with the optical theorem. An analogous result for infinitely long cylinders was also derived. Magneto optical Kerr effects in non-absorbing nanoparticles with magneto-optical activity arise as a consequence of radiative corrections to the electrostatic polarizability tensor.

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1. Introduction

Electromagnetic scattering from nanometer-scale objects has long been a topic of large interest and relevance to fields from astrophysics or meteorology to biophysics and material science [1, 2, 3, 4, 5, 6]. During the last decade nano-optics has developed itself as a very active field within the nanotechnology community. Much of it has to do with plasmon (propagating) based subwavelength optics and applications [7]. Also, isolated metallic particles supporting localized plasmons have attracted a great deal of interest due to their ability to concentrate the electromagnetic field in subwavelength (some tens of nanometers) volumes. As a result, the studies in the field often involve the contributions of small elements or particles where the dipole approximation may be sufficient to describe the optical response. Examples of applications are on telecommunications [8, 9, 10], spontaneous emission rates and fluorescence [11, 12, 13], sensors [14], energy harvesting[15, 16], optical forces and trapping [17, 18, 19, 20] or medical therapy [21]. The capabilities and applicability of all these promising examples can be largely enhanced if some degree of tunability is added. These capacities can be endorsed by exploiting different x-optic effect (thermo-, electro-, magneto-, piezo-) where an external agent modifies some elements of the dielectric tensor, $\boldsymbol{\varepsilon}$, in some extent [22, 23, 24] which, in general, will be non-diagonal.

Most of the previous works on small anisotropic spherical particles, consider the dipolar approximation (DA) in the electrostatic limit [25, 26, 27, 28, 29, 30, 31]. By taking into account the fact that the polarization within the sphere is uniform, the polarizability is usually written as [25]

$$\boldsymbol{\alpha}_0 \equiv 3\mathrm{v}(\boldsymbol{\varepsilon} - \varepsilon_h \mathbf{I})(\boldsymbol{\varepsilon} + 2\varepsilon_h \mathbf{I})^{-1} \tag{1}$$

being $\mathrm{v} = 4\pi a^3/3$ the particle volume ($\mathbf{I} = \mathbf{u}_x\mathbf{u}_x + \mathbf{u}_y\mathbf{u}_y + \mathbf{u}_z\mathbf{u}_z$ is the unit dyadic) and ε_h the relative permittivity of the host medium at the point where the particle is placed. The host medium is assumed to be isotropic. Different extensions, including anisotropic and bianisotropic non-spherical particles, have been considered in the literature (for a short review see Ref. [32]). However, in most of the cases, the energy balance between absorption and scattering has not been considered. In particular, in absence of absorption, the polarizability tensor given by Eq. (1) does not fulfil the Optical Theorem. For isotropic particles (where $\boldsymbol{\alpha} = \alpha\mathbf{I}$ is a scalar quantity), radiative corrections to the electrostatic polarizability [5, 6] solve the problem of energy

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conservation in absence of absorption. Even in the case of absorbing particles, extended radiative corrections have been shown to be relevant to determine the effective permittivity of metallic nanoparticle doped composites [33]. However, these corrections have not been considered in the context of scattering from small anisotropic particles.

In this work we analyze the polarizability of small dielectrically anisotropic particles including radiative corrections. In Sec. 2 we describe the general properties of any polarizability tensor consistent with the Optical Theorem. In Sec. 3 we derive a generalized polarizability tensor equivalent to the extended polarizability tensor arising in the so-called "strong couple dipole method " (S-CDM) [34]. We show that, in absence of absorption, it is consistent with the optical theorem. Equivalent results for cylinders are also derived in Sec. 3.2. These results are of general applicability.

As an important application, we are going to restrict ourselves to the magneto-optical case (Sec. 4), where the presence of a magnetic field alters some of the non-diagonal components of the dielectric tensor (for an interesting discussion on the similarities and differences between the polarizabilities of gyrotropic and chiral particles see ref. [28, 29]). Depending on the relative orientation of the sample, incidence plane and magnetic-field the affected elements will vary, conferring different effects. In the so-called "polar" configuration, where the magnetic field is applied perpendicular to the sample plane and parallel to the light incidence plane and the main effect is a rotation of the polarization state, it has been shown that the plasmon excitation largely modifies the rotation [35, 36, 37] due to the strong enhancement and localization of the EM field. As we will show, the polarizability given by Eq. (1) wrongly predicts the absence of Kerr rotation for non-absorbing magneto-optical particles.

2. Optical Theorem for anisotropic Rayleigh particles

Let us consider a dielectric anisotropic particle with a permittivity tensor $\boldsymbol{\varepsilon}(\omega)$ and radius a in an otherwise uniform medium with relative permittivity ε_h and refractive index $n_h = \sqrt{\varepsilon_h}$. For a linear non-magnetic material and harmonic fields, the electric displacement $\boldsymbol{D}$ inside the particle is related to the electric field $\mathbf{E}$ through $\boldsymbol{D} = \varepsilon_0 \boldsymbol{\varepsilon}(\omega)\mathbf{E}$ and $\mathbf{H} = \varepsilon_0 c^2 \boldsymbol{B}$ (being c the vacuum's speed of light). For small particles ($a \ll \lambda$), the electromagnetic response is well described by the dipolar approximation (DA): an external electric field $\mathbf{E}_0$ induce an electric dipole $\mathbf{p} = \varepsilon_0 \varepsilon_h \boldsymbol{\alpha}(\omega)\mathbf{E}_0$ where $\boldsymbol{\alpha}(\omega)$ is the polarizability tensor.

The optical theorem for anisotropic particles can be easily deduced from the Poynting's theorem for harmonic fields [38]: The time-averaged rate of work done by the external field $\mathbf{E}_0$ in a volume V must be equal to the sum of dissipated and radiated powers:

$$\frac{1}{2}\Re\left\{\int_V \boldsymbol{J}^* \cdot \mathbf{E}_0 d^3\mathbf{r}\right\} = P_{dis} + P_{rad} = P_{dis} + \frac{1}{2}\Re\left\{\oint_s \mathbf{E}_s \times \mathbf{H}_s^* \cdot \boldsymbol{n} ds\right\} \tag{2}$$

where $\mathbf{E}_s$ and $\mathbf{H}_s$ are the scattered fields. For dielectric particles, the current density, $\boldsymbol{J} = -i\omega\boldsymbol{P}$, is proportional to the polarization vector $\boldsymbol{P}$. If the particle is uniformly polarized, $\boldsymbol{P} = \mathbf{p}/\mathrm{v} = \varepsilon_0 \varepsilon_h \boldsymbol{\alpha}\mathbf{E}_0/\mathrm{v}$, and we have

$$\frac{1}{2}\omega\Im\left\{\mathbf{E}_0^\dagger \cdot \mathbf{p}\right\} = P_{dis} + \frac{c}{n_h}\frac{k^4}{12\pi\varepsilon_0\varepsilon_h}|\mathbf{p}|^2 = \tag{3}$$

$$\frac{1}{2}\omega\varepsilon_0\varepsilon_h\Im\left\{\mathbf{E}_0^\dagger\boldsymbol{\alpha}\mathbf{E}_0\right\} = P_{dis} + \frac{c}{n_h}\frac{k^4}{12\pi}\varepsilon_0\varepsilon_h\mathbf{E}_0^\dagger\boldsymbol{\alpha}^\dagger\boldsymbol{\alpha}\mathbf{E}_0 \tag{4}$$

where $k^2 = \varepsilon_h\omega^2/c^2 = 2\pi/\lambda$ is the wave number and we have made use of the well known result of the total power radiated by an oscillating dipole [38]. In absence of absorption,

$$\frac{k^3}{6\pi}\mathbf{E}_0^\dagger\boldsymbol{\alpha}^\dagger\boldsymbol{\alpha}\mathbf{E}_0 = \Im\left\{\mathbf{E}_0^\dagger\boldsymbol{\alpha}\mathbf{E}_0\right\} = \Im\left\{\mathbf{E}_0^\dagger\left(\boldsymbol{\alpha}^\dagger\boldsymbol{\alpha}^{\dagger^{-1}}\right)\boldsymbol{\alpha}\mathbf{E}_0\right\} \tag{5}$$

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which can be written as

$$\Im\left\{\mathbf{p}^{\dagger}\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}\mathbf{p}\right\}=0 \quad ; \quad \boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}\equiv\left\{\boldsymbol{\alpha}^{\dagger^{-1}}-i\frac{k^3}{6\pi}\mathbf{I}\right\} \tag{6}$$

The matrix $\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}$ must be Hermitian (i.e. $\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}=\left(\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}\right)^{\dagger}$) since the expression above must hold for arbitrary $\mathbf{p}$. In absence of absorption, the inverse of the polarizability tensor must then have the following general expression:

$$\boldsymbol{\alpha}^{-1}=\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}-i\frac{k^3}{6\pi}\mathbf{I} \tag{7}$$

being $\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}$ an arbitrary hermitian matrix. Equation (7) represents the optical theorem for the polarizability tensor of non-absorbing anisotropic particles.

2.1. *Optical theorem in the presence of several scattering objects*

Let us now consider $\mathbf{E}_0$ as the field generated by "fixed" external sources and the scattered field for any other object *in absence of the small particle*. As a consequence of multiple scattering effects, in the presence of other scattering objects or inhomogeneities in the host medium, the actual polarizing field incoming towards the particle, $\mathbf{E}_{inc}$, is obviously different from $\mathbf{E}_0$. The induced dipole can be written as

$$\mathbf{p}=\varepsilon_0\varepsilon_h\boldsymbol{\alpha}\mathbf{E}_{inc}=\varepsilon_0\varepsilon_h\widehat{\boldsymbol{\alpha}}\ \mathbf{E}_0 \tag{8}$$

where $\boldsymbol{\alpha}$ is the particle's polarizability and $\widehat{\boldsymbol{\alpha}}$ is a renormalized polarizability including all the multiple scattering effects between the dipolar particle and the rest of the system.

The field radiated from the dipole can be written in terms of the dyadic Green function (Green tensor) of the system $\mathbb{G}$. $\mathbb{G}(\mathbf{r},\mathbf{r}_0)$ connects an electric-dipole source $\mathbf{p}$ at a position $\mathbf{r}_0$ to the electric field at a position $\mathbf{r}$ through the relation $\mathbf{E}(\mathbf{r})=[k^2/(\varepsilon_0\varepsilon_h)]\mathbb{G}(\mathbf{r},\mathbf{r}_0)\mathbf{p}$. The Green tensor for a homogeneous system, $\mathbf{G}(\mathbf{r},\mathbf{r}')$, is defined as the solution of

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{G}-k_0^2\varepsilon_h\mathbf{G}=\mathbf{I}\delta(\mathbf{r}-\mathbf{r}_0), \tag{9}$$

with the outgoing-wave condition at infinity. In a homogeneous medium, it reads:

$$\mathbf{G}(\mathbf{r},\mathbf{r}_0)|_{\mathbf{r}\neq\mathbf{r}_0}=\left(\mathbf{I}+\frac{\boldsymbol{\nabla}\boldsymbol{\nabla}}{k^2}\right)\frac{e^{ik|\mathbf{r}-\mathbf{r}_0|}}{4\pi|\mathbf{r}-\mathbf{r}_0|}. \tag{10}$$

The total Green function can be written as $\mathbb{G}(\mathbf{r},\mathbf{r}_0)=\mathbf{G}(\mathbf{r},\mathbf{r}_0)+\mathbf{G}_b(\mathbf{r},\mathbf{r}_0)$ where $\mathbf{G}$ is the source term for the homogeneous system and the dyadic $\mathbf{G}_b$ describes the field scattered by any other object in the system.

The total power radiated by the dipole is modified by the presence of other particles or inhomogeneities and can be written as [39]

$$P_{rad}=\frac{\omega^3\mu_0}{2}\mathbf{p}^{\dagger}\cdot\Im\left\{\mathbb{G}\left(\mathbf{r}_0,\mathbf{r}_0\right)\right\}\cdot\mathbf{p} \tag{11}$$

For a non-absorbing particle, Eq. (4) then becomes

$$\Im\left\{\mathbf{E}_0^{\dagger}\widehat{\boldsymbol{\alpha}}\mathbf{E}_0\right\} \quad = \quad k^2\mathbf{E}_0^{\dagger}\widehat{\boldsymbol{\alpha}}^{\dagger}\cdot\Im\left\{\mathbb{G}\left(\mathbf{r}_0,\mathbf{r}_0\right)\right\}\cdot\widehat{\boldsymbol{\alpha}}\mathbf{E}_0 \tag{12}$$

As a consequence, the inverse of the renormalized polarizability tensor must then have the following general expression:

$$\widehat{\boldsymbol{\alpha}}^{-1}=\widehat{\boldsymbol{\alpha}_{\boldsymbol{h}}}^{-1}-ik^2\Im\left\{\mathbb{G}\left(\mathbf{r}_0,\mathbf{r}_0\right)\right\} \tag{13}$$

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being $\widehat{\boldsymbol{\alpha}_{\boldsymbol{h}}}^{-1}$ an hermitian matrix. This generalize the optical theorem in the presence of several scattering objects. Notice that in absence of scattering objects, $\Im\{\mathbb{G}(\mathbf{r}_0,\mathbf{r}_0)\} = \Im\{\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0)\} = k/(6\pi)\mathbf{I}$ (a well known result for the Green tensor in homogeneous media) and we recover the results of Eq. (7),

$$\boldsymbol{\alpha}^{-1} = \boldsymbol{\alpha}_{\boldsymbol{h}}^{-1} - ik^2\Im\{\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0)\}. \tag{14}$$

$\boldsymbol{\alpha}$ and $\widehat{\boldsymbol{\alpha}}$ are related through a simple self-consistent equation. The actual incoming field $\mathbf{E}_{inc}$ must be given by the sum of $\mathbf{E}_0$ and the field backscattered from the other objects in the system:

$$\mathbf{E}_{inc}(\mathbf{r}_0) = \mathbf{E}_0(\mathbf{r}_0) + \frac{k^2}{\varepsilon_0\varepsilon_h}\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0)\mathbf{p} = \mathbf{E}_0(\mathbf{r}_0) + k^2\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0)\boldsymbol{\alpha}\mathbf{E}_{inc} \tag{15}$$

$$\mathbf{p} = \varepsilon_0\varepsilon_h\boldsymbol{\alpha}\left(\mathbf{I} - k^2\mathbf{G}_b(0)\boldsymbol{\alpha}\right)^{-1}\mathbf{E}_0 = \varepsilon_0\varepsilon_h\widehat{\boldsymbol{\alpha}}\mathbf{E}_0 \tag{16}$$

From Eqs. (13) and (14) we then have

$$\begin{aligned}\widehat{\boldsymbol{\alpha}}^{-1} &= \boldsymbol{\alpha}^{-1} - k^2\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0) = \boldsymbol{\alpha}_{\boldsymbol{h}}^{-1} - ik^2\Im\{\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0)\} - k^2\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0) \\ &= \left(\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1} - k^2\Re\{\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0)\}\right) - ik^2\Im\{\mathbb{G}(\mathbf{r}_0,\mathbf{r}_0)\}\end{aligned} \tag{17}$$

$$\widehat{\boldsymbol{\alpha}_{\boldsymbol{h}}}^{-1} = \boldsymbol{\alpha}_{\boldsymbol{h}}^{-1} - k^2\Re\{\mathbf{G}_b(\mathbf{r}_0,\mathbf{r}_0)\} \tag{18}$$

3. Polarizability of anisotropic Rayleigh particles

We now present a simple extension of the electrostatic approach that leads in a natural way to a polarizability consistent with Eq. (7). From Maxwell equations, the electric field follows the wave equation

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\mathbf{E} - k_0^2\varepsilon_h\mathbf{E} = k_0^2(\boldsymbol{\varepsilon}(\mathbf{r}) - \varepsilon_h\mathbf{I}).\mathbf{E} \tag{19}$$

where $k_0 = \omega/c$ and $\boldsymbol{\varepsilon}(\mathbf{r}) = \boldsymbol{\varepsilon}\Theta(a - |\mathbf{r}-\mathbf{r}_0|)$ for a particle of radius a located at $\mathbf{r} = \mathbf{r}_0$ (being Θ the Heaviside step function). In scattering problems, the total field is usually written as the sum of incoming $\mathbf{E}_0$ and scattered $\mathbf{E}_{scatt}$ fields. By using the Green tensor, Eq. (19) can be transformed into an integral equation

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + k_0^2\int\mathbf{G}(\mathbf{r},\mathbf{r}')(\boldsymbol{\varepsilon}(\mathbf{r}') - \varepsilon_h\mathbf{I})\mathbf{E}(\mathbf{r}')d^3\mathbf{r}' \tag{20}$$

In the small particle limit, we can consider that the electric field inside $\mathbf{E}_{\text{inside}}$ the particle is approximately constant. The total field **outside the particle** can then be written as

$$\begin{aligned}\mathbf{E}(\mathbf{r}) &\approx \mathbf{E}_0(\mathbf{r}) + k_0^2\mathbf{G}(\mathbf{r},\mathbf{r}_0)\mathrm{v}(\boldsymbol{\varepsilon} - \varepsilon_h\mathbf{I})\mathbf{E}_{\text{inside}}(\mathbf{r}_0) \\ &= \mathbf{E}_0(\mathbf{r}) + \frac{k^2}{\varepsilon_0\varepsilon_h}\mathbf{G}(\mathbf{r},\mathbf{r}_0).\mathbf{p}\end{aligned} \tag{21}$$

where v is the particle volume and the induced electric dipole, $\mathbf{p}$ is given by $\mathbf{p} = \varepsilon_0\varepsilon_h\mathrm{v}(\boldsymbol{\varepsilon} - \varepsilon_h\mathbf{I})\mathbf{E}_{\text{inside}} \equiv \varepsilon_0\varepsilon_h\boldsymbol{\alpha}\mathbf{E}_0(\mathbf{r}_0)$. We can use Eq. (20) to obtain a self-consistent solution for $\mathbf{E}_{\text{inside}}$,

$$\begin{aligned}\mathbf{E}_{\text{inside}} &= \mathbf{E}_0(\mathbf{r}_0) + k_0^2\left(\int\mathbf{G}(\mathbf{r}_0,\mathbf{r}')d^3\mathbf{r}'\right)(\boldsymbol{\varepsilon} - \varepsilon_h\mathbf{I}).\mathbf{E}_{\text{inside}} \\ &= \mathbf{E}_0(\mathbf{r}_0) + k_0^2\mathrm{v}\langle\mathbf{G}\rangle(\boldsymbol{\varepsilon} - \varepsilon_h\mathbf{I}).\mathbf{E}_{\text{inside}}\end{aligned} \tag{22}$$

where $\langle\mathbf{G}\rangle$ is the average of the Green tensor over the particle volume v. The self-consistent internal field is then given by

$$\mathbf{E}_{\text{inside}} = \left\{\mathbf{I} - k_0^2\mathrm{v}\langle\mathbf{G}\rangle(\boldsymbol{\varepsilon} - \varepsilon_h\mathbf{I})\right\}^{-1}\mathbf{E}_0(\mathbf{r}_0). \tag{23}$$

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Following the notation of Ref. [34], we can write

$$\mathrm{v}\langle\mathbf{G}\rangle \equiv \left(\int \mathbf{G}(\mathbf{r}_0,\mathbf{r}')d^3\mathbf{r}'\right)_{\mathrm{v}\to 0} = \left(-\frac{1}{k^2}\boldsymbol{L}+\boldsymbol{M}\right) \tag{24}$$

where $\boldsymbol{L}$ is the (real) electrostatic depolarization dyadic [40, 41, 42] and the dyadic $\boldsymbol{M}$ corresponds to the volume average of the non-singular part of the Green tensor. We can then rewrite Eq. (23) as

$$\mathbf{E}_{\mathrm{inside}} = \left\{\mathbf{I}-k_0^2\left(-\frac{1}{k^2}\boldsymbol{L}+\boldsymbol{M}\right)(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\right\}^{-1}\mathbf{E}_0(\mathbf{r}_0), \tag{25}$$

which is exactly the S-CDM internal field obtained in Ref. [34] for non magnetic particles. This leads to the S-CDM polarizability

$$\boldsymbol{\alpha} = \mathrm{v}(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\left\{\mathbf{I}-k_0^2\left(-\frac{1}{k^2}\boldsymbol{L}+\boldsymbol{M}\right)(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\right\}^{-1} \tag{26}$$

It is of interest to rewrite $\boldsymbol{M}$ as the sum of real and imaginary dyadics, $\boldsymbol{M}=\boldsymbol{M}_R+i\boldsymbol{M}_I$. Noticing that, at lowest order, $\boldsymbol{M}_I$ is given by $\approx i\mathrm{v}\Im\{\mathbf{G}(\mathbf{r}_0,\mathbf{r}_0)\} = i\mathrm{v}k^3/(6\pi)$ we have

$$\boldsymbol{\alpha} = \boldsymbol{\alpha_h}\left\{\mathbf{I}-i\frac{k^3}{6\pi}\boldsymbol{\alpha_h}\right\}^{-1} \tag{27}$$

$$\boldsymbol{\alpha_h} = \mathrm{v}(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\Big\{\varepsilon_h\mathbf{I}+(\boldsymbol{L}-k^2\boldsymbol{M}_R)(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\Big\}^{-1}. \tag{28}$$

Equations (27) and (28) apply to absorbing as well as nonabsorbing particles of *arbitrary shape* as long as they are electrically small, i.e. as long as the electric field inside the particle is approximately constant. Moreover, in absence of absorption, they exactly match the optical theorem condition (Eq. (7)), i.e. the condition that absorption be absent requires that the polarizability tensor $\boldsymbol{\alpha_h}$ is Hermitian. The dielectric tensor evidently must have the same symmetry properties $\boldsymbol{\varepsilon}=\boldsymbol{\varepsilon}^\dagger$.

3.1. Polarizability of anisotropic spheres

For a spherical particle [34, 40, 41] $\boldsymbol{L}=(1/3)\mathbf{I}$ and $\boldsymbol{M}_R=(a^2/3)\mathbf{I}+\ldots$. We then have

$$\boldsymbol{L}-k^2\boldsymbol{M}_R = \frac{1}{3}\left\{1-(ka)^2+\ldots\right\}\mathbf{I}. \tag{29}$$

Keeping the zero order term in this expansion leads to the polarizability given by Eq. (27) with $\boldsymbol{\alpha_h}=\boldsymbol{\alpha}_0$ given by Eq. (1). In the isotropic case $\boldsymbol{\alpha}_0=\alpha_0\mathbf{I}$ we recover the well known Draine's result for the polarizability of a small (Rayleigh) particle with radiative corrections [5, 6].

Including the $(ka)^2$ term in Eq. (29) we obtain

$$\boldsymbol{\alpha_h} \equiv 3\mathrm{v}(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})(\boldsymbol{\varepsilon}+2\varepsilon_h\mathbf{I}-(ka)^2(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I}))^{-1} \tag{30}$$

In the isotropic case, this result (together with Eq. (27)) leads to Lakhtakia's S-CDM polarizability (see Eq. (60) in Ref. [34]). Interestingly, including higher order terms in the expansion of $\boldsymbol{M}_I$ can unbalance the small particle approach leading to a polarizability tensor which does not fulfil the Optical Theorem.

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3.2. *Polarizability of anisotropic cylinders*

A similar argument can be applied to the polarizability of dielectric anisotropic cylinders with very small radius. We consider a long rod with its axis along the z-axis. We will restrict ourselves to the case in which the external electromagnetic fields do not depend on z, i.e. $\mathbf{E}_0(\boldsymbol{\rho})$ ($\boldsymbol{\rho} = (x,y)$) and the induced dipole (per unit length) is constant along the cylinder axis. The Green dyadic is in this case:

$$\mathbf{G}(\boldsymbol{\rho},\boldsymbol{\rho}_0)|_{\boldsymbol{\rho}\neq\boldsymbol{\rho}_0} = \left(\mathbf{I}+\frac{\boldsymbol{\nabla\nabla}}{k^2}\right)\frac{i}{4}H_0(k|\boldsymbol{\rho}-\boldsymbol{\rho}_0)|) \tag{31}$$

where H_0 is the Hankel function of the first kind. Following the same steps as with spherical particles we find the appropriate Optical Theorem for small cylinders

$$\boldsymbol{\alpha}^{-1} = -i\frac{k^2}{8}(\mathbf{I}+\mathbf{u}_z\mathbf{u}_z)+\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1} \tag{32}$$

where again $\boldsymbol{\alpha}_{\boldsymbol{h}}^{-1}$ is an arbitrary hermitian matrix. The average of the Green dyadic over the cylinder cross section, $A = \pi a^2$, gives [41]

$$\langle\mathbf{G}\rangle_{A\to 0} \simeq \left\{-\frac{1}{2k^2A}+i\frac{1}{8}\right\}\mathbf{I}_t + i\frac{1}{4}\mathbf{u}_z\mathbf{u}_z \tag{33}$$

where $\mathbf{I}_t = (\mathbf{u}_x\mathbf{u}_x+\mathbf{u}_y\mathbf{u}_y)$ is the transversal unit dyadic. Following the same steps as before we finally have

$$\boldsymbol{\alpha} = \boldsymbol{\alpha_0}\left\{\mathbf{I}-i\frac{k^2}{8}(\mathbf{I}+\mathbf{u}_z\mathbf{u}_z)\boldsymbol{\alpha_0}\right\}^{-1} \tag{34}$$

with

$$\boldsymbol{\alpha_0} = 2A(\boldsymbol{\varepsilon}-\varepsilon_h\mathbf{I})\left(\mathbf{I}_t(\boldsymbol{\varepsilon}+\varepsilon_h\mathbf{I})+2\mathbf{u}_z\mathbf{u}_z\right)^{-1} \tag{35}$$

It is easy to check that the expression exactly match the optical theorem condition (Eq. (32)) provided $\boldsymbol{\alpha}_0 = \boldsymbol{\alpha}_0^{\dagger}$. In the isotropic case $\boldsymbol{\alpha}_0$ is diagonal with

$$\alpha_{0zz} = A(\varepsilon-\varepsilon_h) \tag{36}$$

$$\alpha_{0xx} = \alpha_{0yy} = 2A\frac{\varepsilon-\varepsilon_h}{\varepsilon+\varepsilon_h} \tag{37}$$

and we recover the well known result for the polarizability of a small (Rayleigh) cylinder with radiative corrections (see for example [43, 44, 45]).

4. Magneto-Optical Kerr effect

As an important application, we will discuss the relevance of our results in the analysis of light scattering from magneto-optical (MO) nanoparticles. Even for isotropic dielectrics, in the presence of a static magnetic field $\mathbf{H}$, the tensor $\boldsymbol{\varepsilon}$ is not longer symmetrical. From the principle of symmetry of the kinetic coefficients [46] $\boldsymbol{\varepsilon}(\mathbf{H}) = \boldsymbol{\varepsilon}^{\top}(-\mathbf{H})$.

4.1. *Magneto-Optical spheres. PMOKE*

For MO spheres, we consider the so called polar magneto optical Kerr effect (PMOKE) configuration in which a constant magnetic field $\mathbf{H} = H_z\mathbf{u_z}$ is oriented in the direction of incidence of

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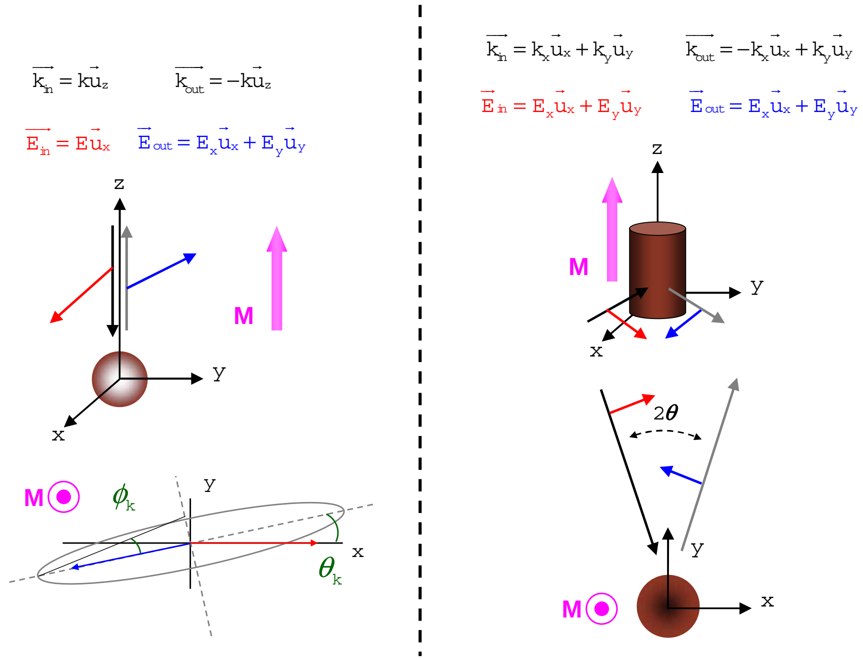

Fig. 1. **(a)** Sketch of the geometry for polar magneto optical Kerr effect (PMOKE) for a spherical nanoparticle. **(b)** Sketch of the geometry for transverse magneto optical Kerr effect (TMOKE) for a cylindrical nanoparticle.

an incoming electromagnetic plane wave $\mathbf{E}_0 = \mathbf{u}_x E_0 e^{ik_0 z}$ (for simplicity we will consider here $\varepsilon_h = 1$). The dielectric tensor can then be written as

$$\boldsymbol{\varepsilon} = \begin{pmatrix} \varepsilon & \varepsilon_{xy} & 0 \\ \varepsilon_{yx} & \varepsilon & 0 \\ 0 & 0 & \varepsilon \end{pmatrix}. \tag{38}$$

where $\varepsilon_{xy}(H_z) = \varepsilon_{yx}(-H_z)$. In absence of absorption we further have $\varepsilon_{xy} = \varepsilon^*_{yx}$. At lowest order in the magnetic field we then have $\varepsilon_{xy} = -\varepsilon_{yx} = -iQ\varepsilon$ where the magneto-optical Voigt parameter Q is proportional either to m_z the magnetization along the z-axis (being $m_z = \pm 1$ at saturation) in the case of a ferro- or antiferro-magnet or to H_z otherwise. The bare polarizability is then given by

$$\alpha_{0xx} = \alpha_{0yy} = 3v\varepsilon_0 \frac{(\varepsilon-1)(\varepsilon+2) - Q^2\varepsilon^2}{(\varepsilon+2)^2 - Q^2\varepsilon^2} \tag{39}$$

$$\alpha_{0yx} = \alpha^*_{0xy} = i\,3v\varepsilon_0 \frac{3Q\varepsilon}{(\varepsilon+2)^2 - Q^2\varepsilon^2} \tag{40}$$

$$\alpha_{0zz} = 3v\varepsilon_0 \frac{\varepsilon-1}{\varepsilon+2} \tag{41}$$

We shall focus on the properties of the backscattered reflected field. In that case the presence of the magnetic field in polar configuration induces a modification of the polarization state of

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the outgoing field with respect to the incident one, known as Kerr rotation. The complex Kerr rotation $\phi = \theta + i\varphi$ (θ is the rotation and φ is the ellipticity) can be defined in terms of the ratio between y and x components of the specular reflected field (see Figure 1a). In the limit $|R_{xy}| \ll |R_{xx}|$, we have

$$\frac{R_{yx}}{R_{xx}} = \frac{\alpha_{yx}}{\alpha_{xx}} = \left|\frac{R_{yx}}{R_{xx}}\right| e^{i\delta} \approx \theta + i\varphi \tag{42}$$

In absence of absorption, α_{0yx} is imaginary and the electrostatic approximation (without radiative corrections) predicts zero rotation ($\theta = 0$) for polar configuration. However, the actual expression of Eq. (42) is given by

$$\frac{R_{yx}}{R_{xx}} = \left\{ \frac{3Q\varepsilon}{(\varepsilon-1)(\varepsilon+2) - Q^2\varepsilon^2} \sin\delta \right\} e^{i\delta} \tag{43}$$

with

$$\frac{1}{\tan\delta} = -C\left(1 - \left|\frac{\alpha_{0yx}}{\alpha_{0xx}}\right|^2\right) \quad , \quad C \equiv \frac{k_0^3}{6\pi\varepsilon_0}\alpha_{0xx} \tag{44}$$

In the limit $Q^2\varepsilon^2 \ll (\varepsilon-1)(\varepsilon+2)$ and $C \ll 1$, we then have

$$\varphi \approx \frac{3}{\varepsilon+2}\frac{\varepsilon}{\varepsilon-1}Q \tag{45}$$

$$\theta \approx -\frac{2}{(\varepsilon+2)^2}(k_0 a)^3 \varepsilon Q \tag{46}$$

These results show that radiative corrections induce a small rotation in polar configurations that is proportional to $(a/\lambda)^3$.

4.1.1. PMOKE of a thin film

Here we are going to develop the expressions of the PMOKE for a very thin layer (thickness d) of the same MO material as the spheres are made of. We could proceed as before assuming that the electric field inside the thin layer is approximately constant and use Eq. (20) to obtain a self-consistent solution for $\mathbf{E}_{\text{inside}}$. Instead, it is simpler if we express a linearly polarized beam as the combination of two: one left and one right circularly polarized beam (σ_- and σ_+). In that case the complex Kerr rotation can be obtained from the reflection coefficients of those left and right circularly polarized beams ($\mathbf{r}_-$ and $\mathbf{r}_+$) as:

$$\frac{R_{yx}}{R_{xx}} = i\left(\frac{r_+ - r_-}{r_+ + r_-}\right) \approx \theta + i\varphi \tag{47}$$

The reflection coefficients $r_\pm$ depend on the different refractive indices ($n_\pm^2 = \varepsilon \pm Q\varepsilon$) for left and right polarizations [46] and, for $k_0 d n_\pm \ll 1$ and normal incidence, can be written as

$$r_\pm \approx i\left(n_\pm^2 - 1\right)\frac{k_0 d}{2} - \left(n_\pm^4 - 1\right)\left(\frac{k_0 d}{2}\right)^2 + \ldots \tag{48}$$

where we keep the lowest real and imaginary terms in the expansion. In the limit $Q^2\varepsilon^2 \ll (\varepsilon^2 - 1)$ we then have

$$\varphi \approx \frac{\varepsilon}{\varepsilon-1}Q \tag{49}$$

$$\theta \approx -\frac{k_0 d}{2}\varepsilon Q \tag{50}$$

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These results basically follows those obtained for a small particle: the ellipticity does not depend on the system size and the rotation scales with the size of the system, i.e. volume of the sphere and thickness of the film respectively. Neglecting radiative corrections is equivalent to keep only the lowest order in d/λ, leading to a pure imaginary $r_\pm$.

4.2. Magneto-Optical cylinders. TMOKE

We shall consider a transverse magneto-optical Kerr effect (TMOKE) where a constant magnetic field $\mathbf{H} = H_z \mathbf{u}_z$ is oriented in the direction parallel to the cylinder and perpendicular to the plane of incidence. The electric field is assumed to be in the incidence plane (p-polarization) $\mathbf{E}_0 = (\mathbf{u}_x \cos\vartheta - \mathbf{u}_y \sin\vartheta) E_0 e^{ik_0 x \sin\vartheta} e^{ik_0 y \cos\vartheta}$ (see Figure 1b). The bare polarizability is given by

$$\alpha_{0xx} = \alpha_{0yy} = 2A\varepsilon_0 \frac{(\varepsilon-1)(\varepsilon+1) - Q^2\varepsilon^2}{(\varepsilon+1)^2 - Q^2\varepsilon^2} \tag{51}$$

$$\alpha_{0yx} = \alpha^*_{0xy} = i\, 2A\varepsilon_0 \frac{2Q\varepsilon}{(\varepsilon+1)^2 - Q^2\varepsilon^2} \tag{52}$$

$$\alpha_{0zz} = A\varepsilon_0(\varepsilon - 1) \tag{53}$$

The reflected TMOKE signal for a single particle can be obtained by calculating the relative variations of the intensity for the specular intensity when the magnetization of the sample is reversed from saturation in one direction ($m_z = +1$) to saturation in the oposite direction ($m_z = -1$) in the case of ferromagnet or when the magnetic field is reversed from $+H_z$ to $-H_z$. In absence of absorption we have $\alpha_{xy} = -\alpha_{yx}$ and the (far field) TMOKE signal is simply given by

$$\frac{\Delta I_0}{I_0} \equiv \frac{I_0(Q) - I_0(-Q)}{I_0(Q) + I_0(-Q)} = \frac{I_0(Q) - I_0(-Q)}{2I_0(Q=0)} = 2\mathrm{Re}\left\{\frac{\alpha_{yx}}{\alpha_{xx}}\right\}\tan(2\vartheta) \tag{54}$$

Notice that this result is closely related to the Kerr rotation in polar configuration. In analogy with the PMOKE signal for spheres, the electrostatic approach without radiative corrections gives zero TMOKE variations. Including radiative corrections we obtain

$$\frac{\Delta I_0}{I_0} = \frac{2Q\varepsilon}{(\varepsilon-1)(\varepsilon+1) - Q^2\varepsilon^2}\sin(2\delta)\tan(2\vartheta) \tag{55}$$

with

$$\frac{1}{\tan\delta} = -\frac{k_0^2}{8\varepsilon_0}\alpha_{0xx}\left(1 - \left|\frac{\alpha_{0yx}}{\alpha_{0xx}}\right|^2\right) \tag{56}$$

In the limit $Q^2\varepsilon^2 \ll (\varepsilon-1)(\varepsilon+2)$, $(k_0 a)^2 \ll 1$, we have

$$\frac{\Delta I_0}{I_0} \approx -\frac{2Q\varepsilon}{(\varepsilon+1)^2}\frac{\pi}{4}(k_0 a)^2\tan(2\vartheta) \tag{57}$$

5. Conclusion

We have derived general expressions for the polarizability of dielectrically anisotropic nanometer size spheres and cylinders or nanorods. Our results generalize the well known radiative corrections to the electrostatic polarizability of small particles [5, 6]. Equations (7), (27) and (28) for arbitrary shaped (electrically small) particles and Eqs. (32), (34) and (35) for cylinders are

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the main general results of this work concerning the optical theorem and polarizability. Magneto optical Kerr effects in the light scattered from non-absorbing nanoparticles (Eqs. (46), (50) and (57)), absent in the electrostatic approximation, were shown to be proportional to the radiative corrections to the polarizability tensor.

We do believe that our results will have significant impact in the optical characterization of new nanostructured materials. Although strong Magneto-Optical effects occur in materials having important absorption losses, the advances in material science together with nanostructured designs will soon lead to MO systems and nanoparticles with negligible absorption. Actually, recently Osada et al. [47] reported a new nanostructured material, based on (Fe/Cu)-cosubtituted titania nanosheets, with negligible absorption and huge MO signal in the near-infrared. Moreover, and in analogy with isotropic composite materials, a correct description of the polarizability is needed to describe the dielectrical properties of MO nanoparticle based composites. Experiments along this direction are in progress.

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