Magnetochiral scattering of light: Optical manifestation of chirality

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We have investigated multiple scattering of light in systems subject to magnetochiral (MC) effects. Our medium consists of magneto-optically active dipoles placed in a chiral geometry under the influence of an external magnetic field. We have calculated the total and the differential scattering MC cross sections of this system, explicitly showing that they are proportional to *pseudoscalar* quantities. This provides an optical measure for the degree of chirality, a pseudoscalar g, of an arbitrary geometrical configuration of scatterers based on its scattering properties. We have calculated g for some simple chiral systems and we have even used it to probe the degree of optical chirality of random systems. Finally, we have compared g with other recently defined chiral measures in literature.

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I. INTRODUCTION

The breaking of fundamental symmetries of nature manifests itself in various optical phenomena and is essential for our understanding of the interaction between light and complex matter. One well-known example is the natural optical activity discovered by Arago in 1811, a nonlocal optical response in media that have broken mirror symmetry, named *chiral* media. Another example is the magnetically induced optical activity, first observed by Faraday in 1846 [1] and associated with the breaking of time-reversal symmetry by a magnetic field. Although physically distinct, the two effects manifest themselves quite similarly in homogeneous media as a rotation of the linear polarization of light. This resemblance has motivated numerous works, pioneered by Pasteur [2], to search for a link between optical activity and the Faraday effect, but in vain. The existence of an optical effect providing such a link is only allowed to occur under conditions where both mirror and time-reversal symmetries are simultaneously broken. The cross effect between natural and magneto-optical activity is called the *magnetochiral* (MC) *effect* and was first predicted in 1962 [3], followed by studies in crystalline materials [4]. The MC effect was later again predicted independently several times [5–9], both for absorption and for refraction.

In order to deduce the MC effect in homogeneous and isotropic media, let us consider the propagation of light with wave vector $\mathbf{k}$ through a magneto optically active chiral medium with dielectric tensor $\boldsymbol{\varepsilon}$ under the influence of a static magnetic field $\mathbf{B}$. Expanding $\boldsymbol{\varepsilon}$ to first order in $\mathbf{k}$ and $\mathbf{B}$, we have [4]

$$\varepsilon_{ij}(\omega,\mathbf{k},\mathbf{B}) = \varepsilon(\omega)\delta_{ij} + \alpha(\omega) i \epsilon_{ijl} k_l + \beta(\omega) i \epsilon_{ijl} B_l + \gamma(\omega)(\mathbf{B}\cdot\mathbf{k})\delta_{ij}, \quad (1)$$

where ϵ_{ijk} is the Levi-Cività tensor, $\omega/2\pi$ is the optical frequency, and α and β are associated with the natural and magneto-optical activity, respectively. The last term of Eq. (1) describes the MC effect in homogeneous and isotropic media. It is important to point out some of the major features related to this effect. First, it depends on the relative orientation of $\mathbf{k}$ and $\mathbf{B}$. Secondly, the effect has opposite sign for the two different chiral enantiometers. Finally, it is independent of the state of polarization of light. Aside from its fundamental value for the understanding of the interaction between light and matter and the underlying symmetry principles that govern it, the MC effect in absorption has been suggested to be at the origin of the homochirality of life [10], since it allows enantioselective photochemistry in a magnetic field with unpolarized light [11]. Unfortunately, the magnitude of the molecular MC effect is very weak (typically of the order of 10^{-6}) since it can be regarded, as discussed above, as a cross effect between two already small effects, the natural and magneto optical activity. That is why the MC effect has only been reported recently: in absorption by Rikken and Raupach [12], and in refraction by Kleindienst and Wagnière [13] and by Vallet *et al.* [14]. The electrical analog of the MC effect was also recently reported [15].

In the present paper, we study the MC effect in *scattering* of light. We have investigated multiple light scattering by magneto-optically active scatterers distributed in a chiral configuration under the influence of an external magnetic field. This approach is completely different from the traditional molecular one that has been employed to model the MC effect [5–9,16]. Multiple light scattering has been studied for scatterers exhibiting rotatory power [17]. In our model magnetochirality is a *collective* effect built up by multiple scattering. One single scatterer does not exhibit MC effects since it is not made of a material that is natural optically active. However, an assembly of magneto-optically active particles distributed in a chiral configuration should generate MC effects since in such a system both mirror and time-reversal symmetry are broken.

In order to investigate how scattering is affected by the MC effect, we have calculated the differential and the total cross sections for a system composed of an arbitrary number of magneto-optically active scatterers in a magnetic field. We have investigated if these quantities are sensitive to the de-

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gree of chirality associated with the geometrical configuration of the scatterers in space, and have concluded that scattering in MC media constitutes an optical manisfestation of chirality, in addition to the well-known rotatory power. An optical parameter is introduced to quantify the chirality associated with the spatial geometry, which we have calculated for some simple chiral systems. We have also employed this parameter to probe the chirality of random systems, which are in general chiral. Finally, we have compared it to recently defined chiral measures in literature [18–20].

This paper is organized as follows. In Sec. II we briefly explain how to quantify chirality and we explore this concept to define an optical chiral parameter based on the scattering properties of MC media. In Sec. III we calculate this parameter for the simplest chiral geometry, the so called "twisted H," whereas in Sec. IV we employ it to quantify the degree of chirality of random scattering systems. Finally, in Sec. V we summarize the main results of this paper.

II. MULTIPLE SCATTERING IN MAGNETOCHIRAL MEDIA

There are several scattering systems eligible for MC effects. The most obvious of them could be a system composed of an assembly of MC scatterers (i.e., made of a material exhibiting both the Faraday effect and natural optical activity). In order to study such a system, one should know precisely how an individual MC object scatters light. Mathematically, this means that the scattering T matrix of this object should be known. However, to our knowledge, no calculation exists for such a T matrix. Alternatively, it is possible to generate the MC effect in scattering systems by considering magneto-optically active scatterers distributed in chiral geometries. This is the kind of system that will be treated in the present work. Before we describe the light scattering properties of such a system, it is instructive to examine some general considerations about chirality itself and how one can quantify the degree of chirality associated with an arbitrary object or a distribution of scatterers in space. Afterward, we will discuss how a chiral configuration of scatterers affects the properties of the light scattered in MC media.

A. What is chirality and how can we quantify it?

By definition, a chiral object is an object whose mirror image cannot be rotated to coincide with itself. It can be readily seen that this implies having at least four particles for the system to be chiral. A three-particle system is necessarily achiral since it is always contained in a plane, which is automatically its mirror plane.

Recent work has addressed the problem of whether chirality can be quantified or measured, in much the same way that one can quantify the degree of order of a ferromagnet. Despite the ubiquity of chirality in nature (almost all arbitrary microscopic and macroscopic objects are chiral, from DNA to a piece of rock) and the important role played by chirality in several areas of research, from pharmaceutics to liquid crystals [21] and to the origin of life [10], only recently has real progress been made. One of the first attempts to establish a quantitative measure of chirality is due to Gilat [22]. Osipov *et al.* [23] developed a molecular measure of chirality based on the behavior of the response functions that characterize molecular optical activity. Harris *et al.* [18–20] proposed an elegant way, based on group theory, to measure the degree of chirality exhibited by a geometric object. They constructed a variety of rotationally invariant pseudoscalars and applied them to quantify the degree of chirality of molecules of arbitrary shape, showing how these parameters govern a particular observable, such as the pitch of a cholesteric liquid crystal [18–20] and the optical rotatory power [24].

The choice for a chiral "order" parameter is not unique [19], but must always be a pseudoscalar *invariant under rotations*. This last fact guarantees that no rotation of the object exists that maps the mirror image of a chiral object onto itself.

In what follows, we will describe the scattering properties of an assembly of magneto-optically active dipoles in a magnetic field.

B. Scattering by magneto-optically active particles

In our model, we deal with dielectric *pointlike* magneto-optical scatterers (i.e., radiative dipoles much smaller than the wavelength of light) located in vacuum. This model enables us to find exactly all scattering properties (i.e., the off shell T matrix) of any number of scatterers in analytic form and without any further approximation. This model exhibits a scattering resonance, where the scattering cross section of a single scattering reaches its maximum value. The 3×3 T matrix for a single scattering, up to first order in the magnetic field $\mathbf{B}$, takes the form [25]

$$\mathbf{t}(\mathbf{B},\omega)=t_0\mathbf{U}+t_1\mathbf{\Phi}, \tag{2}$$

where $\mathbf{U}$ is the unit matrix and $\mathbf{\Phi}$ is the antisymmetric Hermitian tensor $\Phi_{ij}=i\epsilon_{ijk}\hat{B}_k$. The parameter t_0 is the ordinary Rayleigh T matrix, $t_0=-4\pi\Gamma\omega^2/(\omega_0^2-\omega^2-2i\Gamma\omega^3/3c_0)$, ω_0 being the resonance frequency and Γ the resonance linewidth. The parameter t_1, defined as $t_1=-\mu k t_0^2/6\pi$ (where $\mu\equiv[9\sqrt{\varepsilon}/(\varepsilon-1)^2(ka)^3]VB/k, V$ being the Verdet constant of the scatterer, a its radius, and $k=\omega/c_0$), is related to the Faraday rotation inside the particle [25]. The knowledge of the single scattering T matrix in Eq. (2) allows us to obtain the total $3N\times3N$ T matrix of an assembly of N magneto-optical scatterers situated at the positions $\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N$:

$$\mathbf{T}_{\mathbf{k},\mathbf{k}'}=\begin{pmatrix} e^{i\mathbf{k}\cdot\mathbf{r}_1}\\ \vdots \\ e^{i\mathbf{k}\cdot\mathbf{r}_N}\end{pmatrix}^{*}\mathbf{t}\cdot(\mathbf{U}-\mathbf{G}\cdot\mathbf{t})^{-1}\cdot\begin{pmatrix} e^{i\mathbf{k}'\cdot\mathbf{r}_1}\\ \vdots \\ e^{i\mathbf{k}'\cdot\mathbf{r}_N}\end{pmatrix}, \tag{3}$$

where $\mathbf{k}$ and $\mathbf{k}'$ are, respectively, the incident and the scattered wave vectors, and $|\mathbf{k}'|=|\mathbf{k}|=k$. The elements of the $3N\times3N$ $\mathbf{G}$ matrix are equal to the Green functions calculated from the relative positions of the scatterers [26]:

$$\mathbf{G}_{NM}=\begin{cases} -\dfrac{\exp(ikr_{NM})}{4\pi r_{NM}}\left\{[\mathbf{U}-\hat{\mathbf{r}}_{NM}\hat{\mathbf{r}}_{NM}]-\left(\dfrac{1}{ikr_{NM}}+\dfrac{1}{(kr_{NM})^2}\right)[\mathbf{U}-3\hat{\mathbf{r}}_{NM}\hat{\mathbf{r}}_{NM}]\right\} & \text{for } N\neq M,\\ 0 & \text{for } N=M. \end{cases} \tag{4}$$

The set of Eqs. (3) forms a system of linear equations that determines the electric field scattered by the magneto-optical particles, and its solution requires the diagonalization of the $3N\times 3N$ scattering matrix [27]:

$$\mathbf{M}(k)\equiv\mathbf{t}(k)\cdot[\mathbf{U}-\mathbf{G}(k)\cdot\mathbf{t}(k)]^{-1}. \tag{5}$$

We have modified the numerical code developed by Rusek and Orlowski [27] in order to diagonalize the scattering matrix $\mathbf{M}(k)$ in Eq. (5) for Faraday active scatterers. This provides the scattered field amplitude for particles distributed in an arbitrary spatial configuration:

$$f(\sigma\mathbf{k}\rightarrow\sigma'\mathbf{k}')=\mathbf{g}_{\sigma}^{*}\cdot\mathbf{f}(\mathbf{k}\rightarrow\mathbf{k}')\cdot\mathbf{g}_{\sigma'}$$

with

$$\mathbf{f}(\mathbf{k}\rightarrow\mathbf{k}')=\sum_{NM}\mathbf{M}^{NM}\exp(i\mathbf{k}\cdot\mathbf{r}_N)\exp(-i\mathbf{k}'\cdot\mathbf{r}_M), \tag{6}$$

where $\mathbf{g}_{\sigma}$ and $\mathbf{g}_{\sigma'}$ are, respectively, the incident and scattered polarization vectors (expressed in the circular polarization basis) and the sum is performed over all scatterers. Since the Faraday effect is a small perturbation to the scattering, we will expand $\mathbf{f}$ *linearly* in the magnetic field, $\mathbf{f}=\mathbf{f}^0+\mathbf{f}^1$, with $\mathbf{f}^1$ given by

$$\mathbf{f}^1(\mathbf{k}\rightarrow\mathbf{k}',\mathbf{B})=-\frac{k}{4\pi}\alpha(B)\sum_{NMN'}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}\cdot\mathbf{M}_0^{MN'})\times\exp(i\mathbf{k}\cdot\mathbf{r}_N)\exp(-i\mathbf{k}'\cdot\mathbf{r}_{N'}), \tag{7}$$

where the dimensionless quantity $\alpha(B)\equiv(VB/\Gamma)(c_0/\omega_0)^2$ measures the strength of the magnetic field and $\mathbf{M}_0$ is the 3×3 scattering matrix for $\mathbf{B}=0$. For an atom subject to the Zeeman effect, $\alpha(B)$ is typically the ratio between the Zeeman splitting and the resonance linewidth [28]. The corresponding *magneto extinction cross section* $\sigma_{ext}^1(\mathbf{B},\mathbf{k})$, linear in the magnetic field and integrated over all scattered angles, can be obtained from the optical theorem [29]:

$$\sigma_{ext}^1(\mathbf{B},\mathbf{k})=-\frac{1}{k}\mathrm{Im}\sum_{\sigma}f^1(\sigma\mathbf{k}\rightarrow\sigma\mathbf{k}) =\frac{\alpha(B)}{4\pi}\mathrm{Im}\sum_{NMN'}\mathrm{Tr}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}\cdot\mathbf{M}_0^{MN'}\cdot\mathbf{\Delta}_{\mathbf{k}})\times\exp[i\mathbf{k}\cdot(\mathbf{r}_N-\mathbf{r}_{N'})], \tag{8}$$

where $(\mathbf{\Delta}_{\mathbf{k}})_{ij}\equiv\delta_{ij}-k_ik_j/k^2$ is the projector upon the space of transverse polarization, normal to $\mathbf{k}$. Notice that we have traced over polarization, which means that the Faraday effect itself cancels and that only MC effects remain. Equation (8) can be regarded as the magneto dichroism of the multiple scattering system, associated with the energy removed from the incident beam due the application of an external magnetic field.

Until now, nothing was said about how the light scattering properties of magneto-optically active particles are affected by the chiral configuration and, more important, how these properties can be related to a measure of the degree of chirality. We recall that any chiral measure must be rotationally invariant. This implies that we must perform an average over all solid rotations in the expression (8) for $\sigma_{ext}^1(\mathbf{B},\mathbf{k})$ in order to estimate magneto-optical chirality. The angularly averaged magneto extinction cross section in MC media must necessarily have the form

$$\langle\sigma_{ext}^1(\mathbf{B},\mathbf{k})\rangle_{4\pi}=g\,\alpha(B)(\hat{\mathbf{B}}\cdot\hat{\mathbf{k}}), \tag{9}$$

where g is some pseudoscalar and the angular brackets $\langle\cdots\rangle_{4\pi}$ denote the average over all solid rotations. This is mathematically equivalent to an average over the vectors $\mathbf{k}$ and $\mathbf{B}$ with fixed mutual orientation. Equation (9) is manifestly rotationally invariant and obeys the two fundamental symmetry relations (parity and reciprocity) in MC media, as can be easily verified. Using Eqs. (8) and (9), we can write g as

$$g\equiv\frac{1}{4\pi}\mathrm{Im}\sum_{NMN'}\langle\mathrm{Tr}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}_{\mathbf{k}}\cdot\mathbf{M}_0^{MN'}\cdot\mathbf{\Delta}_{\mathbf{k}})\times\exp(i\mathbf{k}\cdot\mathbf{r}_{NN'})\rangle_{\mathbf{k}\in 4\pi}, \tag{10}$$

where $\mathbf{r}_{NN'}\equiv\mathbf{r}_{N'}-\mathbf{r}_N$ and $(\Phi_{\mathbf{k}})_{ilm}=i\epsilon_{ilm}k_m$. Notice that, since $\mathbf{M}_0^{NM}$ depend only on the positions of the scatterers $\{\mathbf{r}_N\}$ and not on $\hat{\mathbf{k}}$, they will not be affected by the average. Consequently, the term in the angular brackets in Eq. (10) involves an average of the type $\langle\hat{\mathbf{k}}(1-\hat{\mathbf{k}}\hat{\mathbf{k}})\exp(i\mathbf{k}\cdot\mathbf{r})\rangle_{\mathbf{k}\in 4\pi}$, which can be obtained analytically as

$$\langle\hat{k}_i(\delta_{jk}-\hat{k}_j\hat{k}_k)\exp(i\mathbf{k}\cdot\mathbf{r})\rangle_{\mathbf{k}\in 4\pi} =i\hat{r}_i\delta_{jk}j_1(kr)-i[\hat{r}_i\delta_{jk}+\hat{r}_j\delta_{ik}+\hat{r}_k\delta_{ij}]\frac{j_2(kr)}{kr} +ij_3(kr)\hat{r}_i\hat{r}_j\hat{r}_k, \tag{11}$$

where $j_n(x)$ is the spherical Bessel function of the first kind and of order n. In addition, since $(M_0^{NM})_{ij}=(M_0^{MN})_{ji}$, we have $\mathrm{Tr}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}_{\mathbf{k}}\cdot\mathbf{M}_0^{MN'})=-\mathrm{Tr}(\mathbf{M}_0^{N'M}\cdot\mathbf{\Phi}_{\mathbf{k}}\cdot\mathbf{M}_0^{MN})$. As a result, the terms with $N=N'$ in the sum of Eq. (10) vanish. Consequently, we can simplify Eq. (10) to

$$g=\frac{1}{2\pi}\mathrm{Im}\sum_{N'<N,M} i\langle \mathrm{Tr}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}_{\mathbf{k}}\cdot\mathbf{M}_0^{MN'}\cdot\mathbf{\Delta}_{\mathbf{k}}) \times\sin(\mathbf{k}\cdot\mathbf{r}_{NN'})\rangle_{\mathbf{k}\in 4\pi}. \tag{12}$$

This clearly represents a pseudoscalar, since under a mirror operation $\{\mathbf{r}_N\}$ and $\mathbf{M}_0(\{\mathbf{r}_N\})$ transform as $\mathbf{r}_N\to-\mathbf{r}_N$ and $\mathbf{M}_0(\{\mathbf{r}_N\})\to\mathbf{M}_0(\{-\mathbf{r}_N\})=\mathbf{M}_0(\{\mathbf{r}_N\})$. Inserting Eq. (11) into Eq. (12), it follows that

$$g=\frac{1}{2\pi}\mathrm{Re}\sum_{N'<N,M}\mathrm{Tr}(\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}_{\hat{\mathbf{r}}_{NN'}}\cdot\mathbf{M}_0^{MN'}) \times\left[j_1(kr_{NN'})-\frac{j_2(kr_{NN'})}{kr_{NN'}}\right] +(\hat{\mathbf{r}}_{NN'}\cdot\mathbf{M}_0^{NM}\cdot\mathbf{\Phi}_{\hat{\mathbf{r}}_{NN'}}\cdot\mathbf{M}_0^{MN'}\cdot\hat{\mathbf{r}}_{NN'}) j_3(kr_{NN'}). \tag{13}$$

The second observable of interest is the angular distribution of the light scattered by all particles. With this purpose, we have calculated the differential magneto cross section linear in $\mathbf{B}$, $(d\sigma^1/d\Omega)(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})$, using the relation

$$\frac{d\sigma}{d\Omega}(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B}) =\frac{|f^0(\sigma\mathbf{k}\to\sigma'\mathbf{k}')+f^1(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})|^2}{(4\pi)^2}, \tag{14}$$

where $f^0(\sigma\mathbf{k}\to\sigma'\mathbf{k}')$ is the scattering field amplitude independent of $\mathbf{B}$. Using Eq. (7) to calculate $f^1(\sigma\mathbf{k}\to\sigma'\mathbf{k}')$ and inserting the resulting expression into Eq. (14) one obtains, after summing over outgoing polarization and after averaging over incident polarization,

$$\frac{d\sigma^1}{d\Omega}=-2\frac{k\alpha(B)}{(4\pi)^3}\mathrm{Re}\sum_{NN'LML'}\mathrm{Tr}(\mathbf{\Delta}_{\mathbf{k}}\cdot\mathbf{M}_0^{NN'}\cdot\mathbf{\Delta}_{\mathbf{k}'}\cdot\mathbf{M}_0^{*ML'}\cdot\mathbf{\Phi}\cdot\mathbf{M}_0^{*LM})\exp[-i\mathbf{k}\cdot\mathbf{r}_{NL}+i\mathbf{k}'\cdot\mathbf{r}_{N'L'}]. \tag{15}$$

Since we are interested in scattering quantities sensitive to chirality, we will again perform an average of the scatterers' positions over all rotations in Eq. (15), as we did for the total MC cross section. The calculation of the angularly averaged differential cross section linear in $\mathbf{B}$ $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$ implies the construction of a rotationally invariant scalar from the three vectors $\mathbf{B}$, $\mathbf{k}$, and $\mathbf{k}'$. It can be verified that the only possible form for $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$ obeying parity and reciprocity relations is

$$\left\langle\frac{d\sigma^1}{d\Omega}(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\right\rangle_{4\pi} =\mathcal{H}(\mathbf{k}\cdot\mathbf{k}')\mathbf{B}\cdot(\mathbf{k}\times\mathbf{k}')+\mathcal{C}(\mathbf{k}\cdot\mathbf{k}')\mathbf{B}\cdot(\mathbf{k}+\mathbf{k}'), \tag{16}$$

where $\mathcal{H}(\mathbf{k}\cdot\mathbf{k}')$ is a scalar representing the photonic Hall effect [30] whereas $\mathcal{C}(\mathbf{k}\cdot\mathbf{k}')$ is a pseudoscalar associated with the MC effect. We can thus explicitly obtain an expression for the function $\mathcal{C}(\mathbf{k}\cdot\mathbf{k}')$ using Eq. (15) and by putting $\hat{\mathbf{B}}=\hat{\mathbf{k}}$ in Eq. (16).

In the following, we will numerically calculate g and $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$.

III. SCATTERING CHIRAL MEASURES FOR THE TWISTED H

The "twisted H," depicted in Fig. 1, has the convenient property that its chirality depends in a simple way on the angle γ between its arms. In Fig. 2 we have numerically calculated the parameter g in Eq. (13) for four magneto-optically active pointlike scatterers placed at the vertices of the twisted H as a function of the angle γ for two different values of the wavelength λ of the incident light. Each scatterer was set on resonance with the incident radiation, i.e., we set $\lambda=\lambda_0$. In addition, g was normalized by the normal extinction cross section of the system, which, for the set of parameters used here, can be adequately approximated by $\sigma_0=N\sigma_r$ (where $N=4$ and $\sigma_r=3\lambda_0^2/2\pi$ is the on resonance extinction cross section for one scatterer) since we are in the independent scattering regime. In Fig. 2 we see that g exhibits an oscillatory behavior as a function of the angle γ and *vanishes* exactly at the configurations for which the H is *achiral*. These configurations correspond to the angles $\gamma=n\pi/2$, with n integer. If the value of the incident wavelength is modified, the dependence of g on γ changes, as one can see in Fig. 2, but g still vanishes at the same angles. For different values of the wavelength λ, g may also vanish at

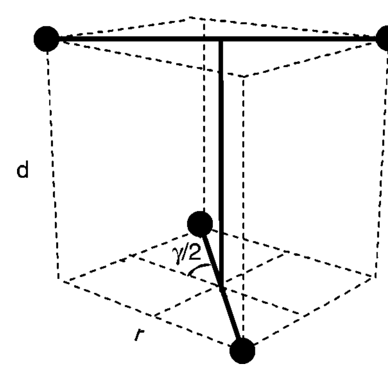

FIG. 1. Four scatterers located at the vertices of the simplest chiral geometry: the so called "twisted H." The coordinates of the scatterers are $[\pm(r/2)\cos(\gamma/2),\pm(r/2)\sin(\gamma/2),\pm d/2]$.

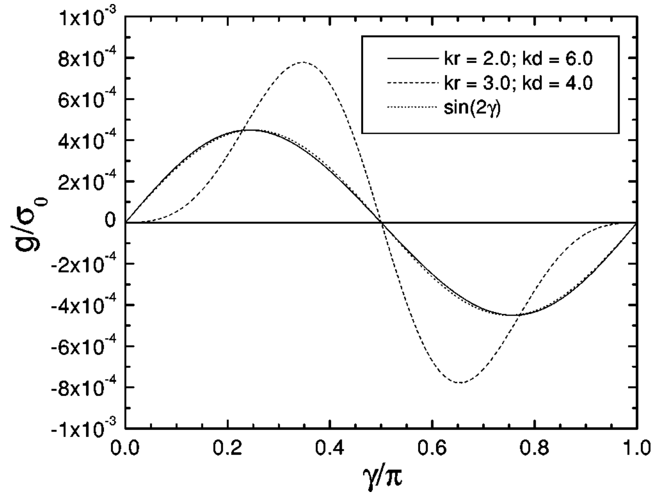

FIG. 2. The on resonance optical chiral parameter g plotted as a function of the angle γ between the "twisted H" arms. The solid curve corresponds to the values $kr=2.0$ and $kd=6.0$ (with k the light wave number). The dashed curve corresponds to the values $kr=3.0$ and $kd=4.0$. The dotted curve presents the chiral parameter $\psi\sim\sin(2\gamma)$ introduced in Refs. [18–20]. g was normalized by $\sigma_0=N\sigma_r$, where $N=4$ and $\sigma_r=3\lambda_0^2/2\pi$.

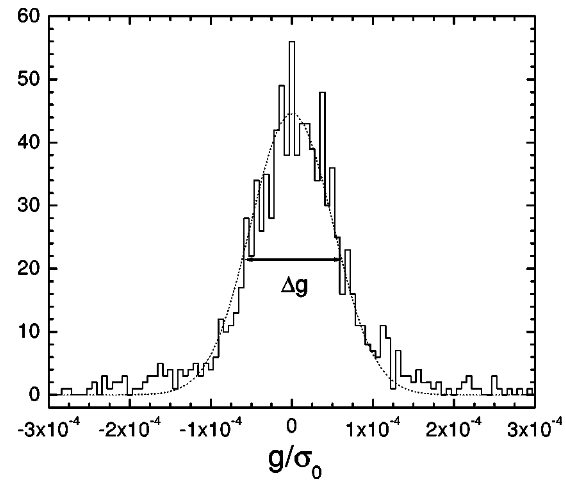

FIG. 4. A typical histogram for the on resonance values of g associated with 1000 different realizations of the positions of $N=10$ scatterers randomly distributed in a sphere of density ρ constant. The values of g were normalized by σ_0 and the parameter ζ, defined in the text, equals $\zeta\approx 0.01$. The dotted curve corresponds to a Gaussian fit to the data, whose full width at half maximum was used to determine the variance Δg.

other values of γ, but the zeros required by symmetry (at the achiral configurations) remain unchanged. For comparison, we show by a dotted line the chiral parameter $\psi\sim\sin(2\gamma)$ for the twisted H proposed by Harris *et al.* [18–20] which, for the set of values $kr=2.0$ and $kd=6.0$, nicely follows our optical parameter g.

In Fig. 3, we have numerically calculated the differential MC cross section averaged over all solid rotations, $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$, for the "twisted H" as a function of the angle γ and for the particular case where $\mathbf{k}\perp\mathbf{B}$ and $\mathbf{k}\perp\mathbf{k}'$. We observe again its oscillatory dependence on γ and, like g, its cancellation for achiral configurations. This

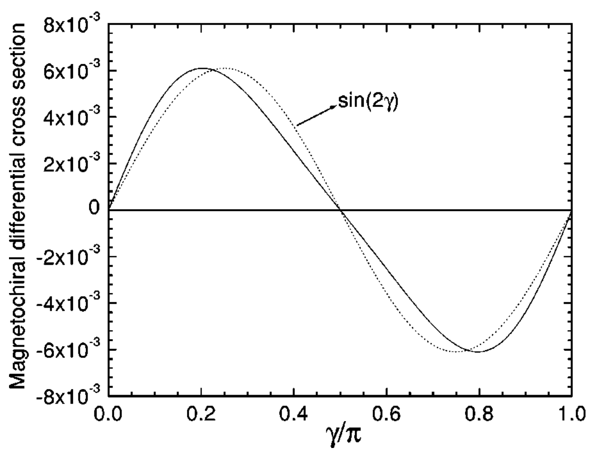

FIG. 3. The on resonance magnetochiral differential scattering cross section averaged over all solid rotations, $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$, plotted as a function of γ for the "twisted H" with $kr=3.0$ and $kd=9.0$ and for $\mathbf{k}\perp\mathbf{B}$ and $\mathbf{k}\perp\mathbf{k}'$ (solid curve). The dotted curve presents the chiral parameter $\psi\sim\sin(2\gamma)$ introduced in Refs. [18–20]. $\langle d\sigma^1/d\Omega(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$ was normalized by σ_0.

means that the intensity of the light scattered by an assembly of magneto-optically active particles under the influence of a magnetic field can also be regarded as an optical manifestation of the degree of chirality associated with the geometrical configuration of these scatterers in space.

In spite of its simplicity, the "twisted H" is an illustrative example of a chiral system. The results presented here could be experimentally verified by means of light scattering measurements performed in samples composed of identical Faraday active "twisted H" molecules dispersed in a liquid. In the single scattering regime this directly provides the optical parameters g and $\langle d\sigma^1/d\Omega\,(\sigma\mathbf{k}\to\sigma'\mathbf{k}',\mathbf{B})\rangle_{4\pi}$.

In the following, we will focus our attention on multiple scattering of light by randomly distributed particles.

IV. PROBING THE CHIRALITY OF RANDOM SCATTERING SYSTEMS

A system composed of a large number of randomly distributed particles will in general be chiral. Our purpose will be to quantify the degree of chirality of this kind of system using the scattering parameter g defined in Eq. (13). We have numerically calculated g for 1000 different random configurations of N scatterers distributed in a sphere of radius R and volume V, and have studied the behavior of g as a function of N for two distinct situations: increasing N while keeping V constant and increasing N while keeping the density $\rho=N/V$ of the scatterers constant.

In Fig. 4, we show a typical histogram for the values of g for 1000 realizations of $N=10$ scatterers randomly distributed in a sphere. The histogram shows a distribution centered at the origin, as expected on the basis of the law of large numbers. Since we have a large number of realizations of the disorder, the mirror image of any configuration is equally

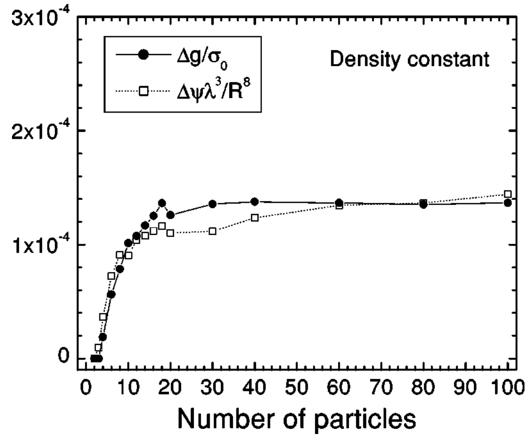

FIG. 5. The variances Δg and $\Delta\psi$ of the chiral parameters g (full circles and solid line) and ψ (empty squares and dotted line), obtained from 1000 different realizations of the disorder, as a function of the number N of scatterers randomly distributed in a sphere of density constant and with $\zeta\approx 0.01$. The values g of were calculated on resonance and normalized by $\sigma_0 = N\sigma_r$. In order to show dimensionless quantities, the values of ψ were multiplied by λ^3/R^8 (where λ is the light wavelength and R is the radius of the sphere). This reveals the proportionality relation $\Delta g/\sigma_0 \propto (\Delta\psi/R^8)\lambda^3$ between Δg and $\Delta\psi$. To allow a better comparison between Δg and $\Delta\psi$, we have multiplied the values of $\Delta\psi$ by an appropriate constant numerical factor. The lines are just a guide for the eyes.

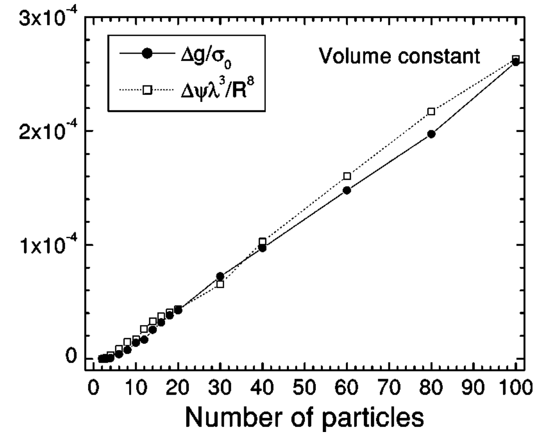

FIG. 6. As in Fig. 5, but now with N scatterers distributed in a sphere at constant volume and with $\zeta\approx 0.000\,25N$.

probable, so that $\langle g\rangle = 0$. We will consider the variance $\Delta g \equiv \sqrt{\langle g^2\rangle - \langle g\rangle^2}$ of g as a candidate to probe the typical degree of chirality of an arbitrary random configuration.

In Fig. 5, we plot the values of Δg as a function of the number of particles distributed in a sphere with ρ constant. The variances Δg were determined by taking the full width at half maximum of the Gaussian fit to the histograms of g for 1000 different realizations of the disorder (see Fig. 4). One relevant dimensionless parameter in the problem is the quantity $\zeta \equiv 4\pi\rho/k^3$, which is essentially the number of particles per cubic wavelength. For the value of ρ chosen in Fig. 5 we have $\zeta\approx 0.01$, which means that we are in the so called independent scattering regime and that only the first orders of scattering are relevant. This allows us to normalize Δg in Fig. 5 by $\sigma_0 = N\sigma_r$. As expected, Δg vanishes for $N = 2,3$, since it is impossible to generate a chiral configuration with only two or three particles. In addition, we observe that the normalized Δg increases until it reaches, for roughly $N = 30$ particles in the sphere, an asymptotic value of approximately $\Delta g/\sigma_0 \approx 1.35\times 10^{-4}$. For comparison, we also show in Fig. 5 the behavior of the variance $\Delta\psi$ of the chiral parameter ψ proposed by Harris *et al.* [18–20] as a function of N. The comparison between the two curves in Fig. 5 clearly reveals that the two chiral measures g and ψ are, statistically speaking, *proportional*. The observed relation of proportionality between Δg and $\Delta\psi$ is given by

$$\frac{\Delta g}{\sigma_0} \propto \frac{\Delta\psi}{R^8}\lambda^3, \tag{17}$$

showing that we can establish a correspondence between the optical chiral measure g and the purely geometrical one ψ/R^8 by multiplying the latter by the optical quantity λ^3. This demonstrates that the correspondence between g and ψ is not only valid for the "twisted H" case discussed in Sec. III, but applies more generally to random systems.

In Fig. 6, Δg and $\Delta\psi$ are shown as a function of N if V is kept constant, i.e., where ρ was varied. In this case, $\zeta \approx 0.000\,25N$. We observe that both Δg and $\Delta\psi$ increase linearly with the number of scatterers. The comparison between Δg and $\Delta\psi$ in Fig. 6 confirms, for V constant, the relation of proportionality between these two chiral parameters. In order to understand the linear dependence of Δg on N we recall that we need at least four particles to constitute a chiral system. This suggests considering the chiral parameter g as the sum of the contributions of groups of four particles, with random sign but with fixed absolute value g_4. Since $\langle g\rangle = 0$ and since $\binom{N}{4}$ distinct forms exist to group four particles together, we can estimate the normalized Δg as

$$\frac{\Delta g}{\sigma_0} = \frac{\sqrt{\langle g^2\rangle}}{N\sigma_r} \sim \frac{1}{N}\sqrt{\binom{N}{4}(g_4)^2}. \tag{18}$$

Taking the limit $N\to\infty$ and noticing by the analysis of Eq. (13) that $g_4 \sim 1/(kR)^3$, we can rewrite Eq. (18) as

$$\frac{\Delta g}{\sigma_0} \sim \frac{\sqrt{N(N-1)(N-2)(N-3)}}{N(kR)^3} \sim \frac{N}{(kR)^3} + \cdots \sim \rho\lambda^3 + \cdots. \tag{19}$$

This heuristic argument confirms the linear dependence of Δg on N observed in Fig. 6 for large N. However, for larger densities (typically for $\zeta\approx 1$) this relation is lost.

V. SUMMARY AND CONCLUSION

In this paper we have investigated scattering of light in magnetochiral media. In order to generate MC effects, we have studied multiple light scattering from chiral geometries of magneto-optically active pointlike scatterers. We have calculated the total and the differential scattering cross sections of such media, showing that they are proportional to pseudoscalar quantities. We have concluded that light scattering in

MC media is sensitive to the degree of chirality exhibited by the geometrical distribution of the scatterers in the space. This constitutes an optical manifestation of chirality, in addition to the well known optical rotatory power. We have introduced in Sec. II a parameter g to measure or to quantify the degree of chirality of an arbitrary configuration of particles subject to MC effects, whose construction is based on the properties of light scattered by such a configuration. As a genuine MC parameter, g is linear in the external magnetic field and independent of the polarization, in contrast to the optical rotatory power. We have shown that g is a pseudoscalar and vanishes for configurations composed of two or three particles, as required for any chiral measure.

In Sec. III we numerically calculated g for scatterers placed on the vertices of one of the simplest chiral objects, the ''twisted H.'' We showed that g vanishes when the H is achiral. Furthermore, g has the same sinusoidal behavior $g \sim \sin(2\gamma)$ as the chiral order parameter ψ defined by Harris *et al.* [18–20] for the ''twisted H.'' In Sec. IV we calculated g for N randomly distributed scatterers in a sphere of volume V and density ρ, a system which will, in general, be chiral. We studied the behavior of the variance Δg as a function of N for two distinct cases: increasing N while keeping ρ constant and increasing N while keeping V constant, i.e., increasing ρ. We have concluded that the variances Δg and $\Delta \psi$ are proportional.

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[1] M. Faraday, Philos. Mag. **28**, 294 (1846).
[2] L. Pasteur, Rev. Sci. **7**, 2 (1884).
[3] M.P. Groenewege, Mol. Phys. **5**, 541 (1962).
[4] D.L. Portigal and E. Burstein, J. Phys. Chem. Solids **32**, 603 (1971).
[5] N.B. Baranova, Yu.V. Bogdanov, and B.Ya. Zeldovich, Opt. Commun. **22**, 243 (1977).
[6] N.B. Baranova and B.Ya. Zeldovich, Mol. Phys. **38**, 1085 (1979).
[7] G. Wagnière and A. Meier, Chem. Phys. Lett. **93**, 78 (1982).
[8] G. Wagnière, Chem. Phys. Lett. **110**, 546 (1984).
[9] L.D. Barron and J. Vrbancich, Mol. Phys. **51**, 715 (1984).
[10] G. Wagnière and A. Meier, Experientia **39**, 1090 (1983); L. D. Barron, in *Physical Origin of Homochirality in Life*, edited by D.B. Cline (AIP, Woodbury, NY, 1996).
[11] G.L.J.A. Rikken and E. Raupach, Nature (London) **405**, 932 (2000).
[12] G.L.J.A. Rikken and E. Raupach, Nature (London) **390**, 493 (1997).
[13] P. Kleindienst and G.H. Wagnière, Chem. Phys. Lett. **288**, 89 (1998).
[14] M. Vallet, *et al.*, Phys. Rev. Lett. **87**, 183003-1 (2001).
[15] G.L.J.A. Rikken, J. Folling, and P. Wyder, Phys. Rev. Lett. **87**, 236602-1 (2001).
[16] G. Wagnière, Chem. Phys. **245**, 165 (1999).
[17] B.P. Ablitt *et al.*, Waves Random Media **9**, 561 (1999).
[18] A.B. Harris, R.D. Kamien, and T.C. Lubensky, Phys. Rev. Lett. **78**, 1476 (1997); **78**, 2867(E) (1997).
[19] A.B. Harris, R.D. Kamien, and T.C. Lubensky, Rev. Mod. Phys. **71**, 1745 (1999).
[20] T.C. Lubensky, A.B. Harris, R.D. Kamien, and G. Yan, Ferroelectrics **212**, 1 (1998).
[21] P.G. de Gennes and J. Prost, *The Physics of Liquid Crystals*, 2nd ed. (Oxford University Press, Oxford, 1993).
[22] G. Gilat, J. Phys. A **22**, L545 (1989).
[23] M.A. Osipov, B.T. Pickup, and D.A. Dunmur, Mol. Phys. **84**, 1193 (1995).
[24] M.S. Spector *et al.*, Phys. Rev. E **61**, 3977 (2000).
[25] B.A. van Tiggelen, R. Maynard, and T.M. Nieuwenhuizen, Phys. Rev. E **53**, 2881 (1996).
[26] A. Lagendijk and B.A. van Tiggelen, Phys. Rep. **270**, 143 (1996).
[27] M. Rusek and A. Orlowski, Phys. Rev. E **51**, R2763 (1995); **56**, 6090 (1997).
[28] G. Labeyrie, C. Miniatura, and R. Kaiser, Phys. Rev. A **64**, 033402-1 (2001).
[29] R.G. Newton, *Scattering Theory of Waves and Particles* (Springer-Verlag, New York, 1982).
[30] D. Lacoste and B.A. van Tiggelen, Europhys. Lett. **45**, 721 (1999).