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Jérôme Crassous, Axelle Amon, Jeanne Crassous. Circular differential scattering of polarized light by a chiral random medium. *Physical Review A : Atomic, molecular, and optical physics [1990-2015]*, 2012, 85 (2), pp.023806. 10.1103/PhysRevA.85.023806 . hal-00848202

HAL Id: hal-00848202

<https://hal.science/hal-00848202v1>

Submitted on 20 Aug 2013

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Circular differential scattering of polarized light by a chiral random medium

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(Received 22 May 2011; published 7 February 2012)

Differential scattering of left- and right-handed circularly polarized waves is a general property of scattering media lacking space-inversion symmetry. We show in this paper that this scattering difference has remarkable properties in the case of an almost transparent chiral random medium. By tuning the refractive indices of the materials composing the medium, this scattering difference can be varied and even inverted. Within the limit of a dense scattering material, the difference of scattering cross sections between polarizations of opposite handedness can be largely adjusted. These properties are illustrated by measurement of the difference between the transmission of right- and left-handed polarized waves through a chiral Christiansen filter.

DOI: [10.1103/PhysRevA.85.023806](https://doi.org/10.1103/PhysRevA.85.023806)

PACS number(s): 33.55.+b, 78.20.Ek, 42.25.Dd, 42.25.Fx

I. INTRODUCTION

Chirality in a medium affects propagation of vectorial waves. Pasteur first identified that molecular asymmetry in a material causes rotation of the plane of polarization of a linearly polarized wave [1]. This effect may be interpreted as a consequence of the difference between the refractive indices n_R and n_L of right- and left-handed circularly polarized waves. A consequence of this index difference is illustrated by the separation of a linearly polarized wave into two rays of opposite handedness [2–4], or the difference of scattering properties of an achiral grating embedded in a chiral material [5]. Many fields of research have special interest in the scattering properties of chiral media. For example, development of metamaterials with adjusted dielectric properties has motivated research on circular differential scattering (CDS) by a two-dimensional (2D) chiral grating [6,7]. In biochemistry, where chirality is a major issue, most characterizations use diffusion methods, motivating CDS studies [8]. Moreover, as biological systems are generally turbid, light transport in heterogeneous chiral media has very practical applications. For example, suspension of polystyrene beads in an optically active liquid has been used to mimic the scattering properties of biological tissues [9,10].

From a theoretical point of view, the effect of chirality on the scattering properties of light in a disordered material was first studied by Perrin [11]. For a material made of chiral particles, the scattering matrix is no longer symmetric, and the scattering properties are dependent on the handedness of the waves. As the difference in scattered fields for right- or left-handed incident waves depends on the difference $n_R - n_L$, which is generally very small, systems exhibiting CDS are considered to be uncommon. Nevertheless, this effect has been put forward to explain distortion in circular dichroism spectra [8,12]. CDS has mainly been reported in materials where the chirality of the geometrical structure has a length scale near the optical wavelength. Examples are the scattering of light by helical objects with pitch of micrometric length [13],

cholesteric liquid crystals [14], or chiral objects of millimetric size illuminated by millimetric electromagnetic waves [15,16].

The object of this paper is to show that this difference between the scattering of left and right circularly polarized waves can become arbitrarily large even for a constant, small difference $n_R - n_L$. The ellipticity of the transmitted wave can then be tuned independently of the optical rotation. That result is counterintuitive, as it seems to be in contradiction with the Kramers-Kronig theorem linking circular dichroism to optical rotation [17]. To demonstrate that effect, we use a chiral Christiansen filter [18], where the difference in circular scattering can be finely controlled by varying the refractive indices of the constituent materials. The circular differential scattering is then shown to strongly depend on the difference between the refractive index n_1 of the isotropic phase and the average index n of the chiral phase. A theoretical description is given, leading to the prediction of a nontrivial symmetry: exchanging the chiral media with its enantiomer $n_R \leftrightarrow n_L$ [Fig. 1 (I) \rightarrow (II)] is equivalent to the permutation of the indices of the two phases $n_1 \leftrightarrow n$ [Fig. 1 (I) \rightarrow (III)]. This effect is evidenced experimentally.

II. EXPERIMENT

We use as the scattering medium a dispersion of dielectric spherical particles in an optically active fluid. The particles are polymethyl methacrylate (PMMA) beads (Atochem, diameter $d = 100\text{--}250\ \mu\text{m}$). The optically active fluid is an 85%:15% mixture of limonene (R , S , or a mixture of R and S ; Sigma-Aldrich) with high-refractive-index microscopy oil (Santolight $n_{\text{oil}} = 1.62$) in order to reach $n \simeq n_1$. The optical activity of the fluid is measured by the optical rotation angle through an $L = 20\ \text{mm}$ cell. The PMMA particles are poured into the liquid, and the cell is gently tapped. The resultant solid fraction is ~ 0.60 . $n_1 - n$ is varied continuously by controlling the temperature of the composite material. The variation of the refractive index of PMMA with temperature is obtained from [19] $dn_1/dT = -1.20 \times 10^{-4}\ \text{K}^{-1}$. The variation of refractive index of the limonene-oil mixture is obtained from the variations of the constituents [20]: $dn/dT = -4.28 \times 10^{-4}\ \text{K}^{-1}$.

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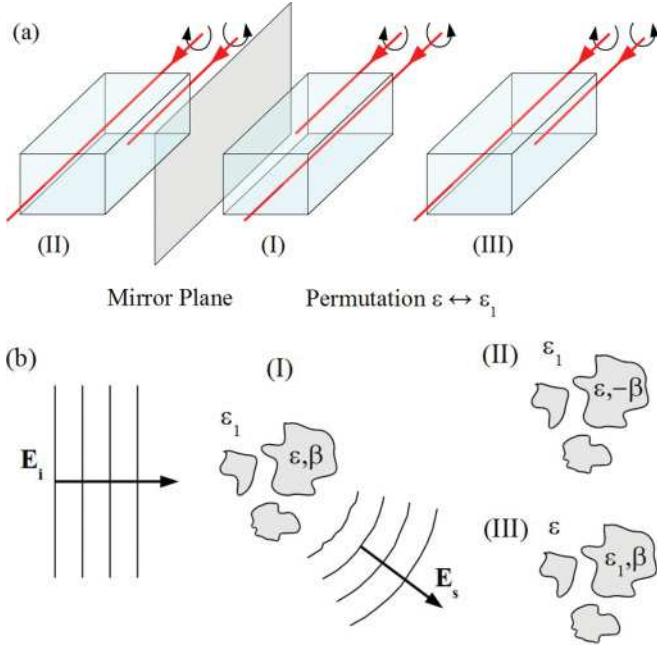


FIG. 1. (Color online) (a) Middle: a chiral material (I) differentially transmits right- and left-handed circular waves. Left (II): Symmetry imposes that the difference in transmission of right- and left-handed circular waves is reversed for the mirror material. Right (III): Near index matching, the permutation of refractive indices also reverses the transmission of right- and left-handed circular waves. (b) (I): A chiral medium with dielectric constant ϵ and chiral parameter β [see (1)] dispersed in a medium ϵ_1 scatters an incident wave \mathbf{E}_i . (II): Same as (I) but the chiral medium has been changed to its enantiomer. (III): Same as (I) but the refractive indices have been permuted.

A schematic drawing of the experimental setup is shown in Fig. 2(a). The cell temperature T is controlled to a few millikelvin precision, and the temperature is increased from 34 to 42 °C at a rate of 0.15 °C/min. The cell is illuminated with a low-power He-Ne laser operating at $\lambda = 633$ nm, and the transmitted light is collected by a 15 mm² square photodiode (PD), centered 50 cm after the exit of the cell. We first discuss the transmission I/I_0 as a function of $n_1 - n$, for a linearly polarized incident wave [Fig. 2(b), dotted line]. The value $n_1 - n$ is set to 0 at the maximum of the transmission curve. The bell-shaped curve is typical of transmission through Christiansen filters. Near index matching, the medium is almost transparent, and the transmission gradually decreases with the quality of the index matching. The finite width of the transmission curve and the imperfect transmission at index matching are due to fluctuations of the refractive indices around their mean values. Although models for transmission of Christiansen filters are still unclear [18], we can use [21] $I/I_0 = \exp\{-A[(n_1 - n)^2 + \sigma^2]\}$ with A a constant dependent on L , λ , d , and the solid fraction, and where σ is the variance of the solid refractive index. From the experimental transmission for $n_1 \simeq n$ [see the inset of Fig. 2(b)], we deduce $\sigma \simeq 2.8 \times 10^{-4}$.

The differential scattering between circular polarizations is obtained by modulation of the incident polarization with a photoElastic modulator (PEM). The imposed phase modulation is 42 kHz with a delay between the fast and slow axes of the PEM of $\lambda/4$. The modulation of the transmitted

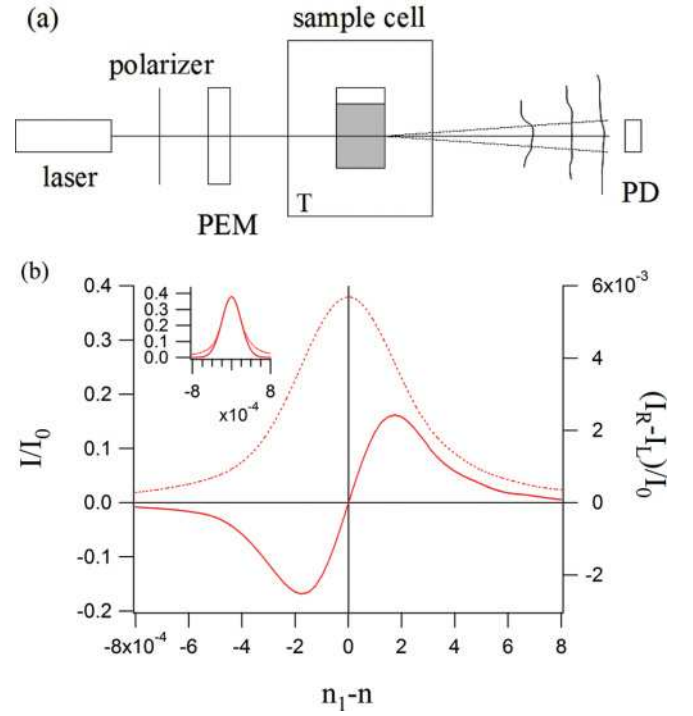


FIG. 2. (Color online) (a) Schematic of the experiment (see text for details). (b) Dotted line, transmission of the cell as a function of the refractive index difference. Solid line, difference of transmission between left and right circular polarization. Inset: Dotted line, transmission. Solid line, fit with $\exp\{-A[(n_1 - n)^2 + \sigma^2]\}$.

intensity is measured with a lock-in amplifier (Stanford Research SR830). The difference of transmission between right and left circular polarization is plotted as a function of the index matching $n_1 - n$ [Fig. 2(b), solid line]. We clearly see that the difference of transmission between opposite circular polarizations depends not only on the presence of asymmetry but also on the contrast of the refractive indices $n_1 - n$.

III. THEORY

The order of the chiral contribution to the scattered wave can be estimated. The constitutive relations for an optically active material are [22]

$$\mathbf{D} = \epsilon \mathbf{E} + \gamma \epsilon \nabla \times \mathbf{E}, \quad \mathbf{B} = \mu \mathbf{H} + \beta \mu \nabla \times \mathbf{H}, \quad (1)$$

where γ and β are parameters responsible for the optical activity. For simplicity, we restrict ourselves to materials where $\gamma = \beta$ and $\mu = \mu_0$, and without any circular dichroism [23]. The refractive indices for right n_R and left n_L monochromatic circularly polarized waves satisfy the relations $(\frac{1}{n_R} + \frac{1}{n_L})^{-1} = \frac{1}{2} \frac{\omega}{k} \sqrt{\epsilon \mu_0}$ and $\frac{1}{n_R} - \frac{1}{n_L} = 2\beta k$, where k is the wave vector in vacuum, and ω is the wave frequency. When such a material is dispersed in a medium of dielectric constant ϵ_1 , Eq. (1) may be written as

$$\mathbf{D} = \epsilon_1 \mathbf{E} + \mathbf{P}_1, \quad \mathbf{B} = \mu_0 (\mathbf{H} + \mathbf{M}) \quad (2)$$

with $\mathbf{P}_1 = (\epsilon - \epsilon_1) \mathbf{E} + \beta \epsilon \nabla \times \mathbf{E}$ and $\mathbf{M} = \beta \nabla \times \mathbf{H}$, which can be respectively considered as the polarization and magnetization of the chiral medium with reference to the isotropic phase. \mathbf{P}_1 and \mathbf{M} may therefore be viewed as the

source of the scattered field created by a plane wave propagating with a wave vector $k_1 = n_1 k$, with $n_1^2 = \epsilon_1/\epsilon_0$. The nonchiral contribution to the scattered field scales as $(\epsilon - \epsilon_1)E$ and the chiral one as $\beta \epsilon k_1 E$. The ratio of those contributions varies then as $\sim \beta k_1 \epsilon / (\epsilon - \epsilon_1) \approx (n_R - n_L)/2(n^2 - n_1^2)$. If the isotropic medium is a vacuum, $n_1 = 1$, and, as $|n_R - n_L| \ll n$, the contribution of chirality to the scattered field stays very small. In contrast, for a dispersion approaching phase matching, $\epsilon_1 \rightarrow \epsilon$, and the effect of chirality on the scattered field becomes preponderant.

For analysis of the differential transmission between the left- and right-handed circular polarization, we need to calculate the variation of the amplitude of the scattered wave as a function of $(\epsilon - \epsilon_1)$ and of β . Let us consider an incident wave $\mathbf{E}_i(\mathbf{r}, t) = \text{Re}[\tilde{\mathbf{E}}_i(\mathbf{r}, \omega) \exp(-j\omega t)]$, with a similar dependence for $\mathbf{H}_i(\mathbf{r}, t)$. The tilde indicates a complex quantity, and we will ignore the ω dependencies in the following analysis. The incident wave propagates into a homogeneous medium with dielectric constant ϵ_1 and magnetic constant μ_0 . Then a perturbation medium characterized by polarization $\tilde{\mathbf{P}}_1(\mathbf{r}) = (\epsilon - \epsilon_1)\tilde{\mathbf{E}}(\mathbf{r}) + \beta \epsilon \nabla \times \tilde{\mathbf{E}}(\mathbf{r})$ and magnetization $\tilde{\mathbf{M}}(\mathbf{r}) = \beta \nabla \times \tilde{\mathbf{H}}(\mathbf{r})$ is added to the homogeneous medium. The field $\tilde{\mathbf{E}}(\mathbf{r})$ in the presence of the perturbation medium may be written as $\tilde{\mathbf{E}}(\mathbf{r}) = \tilde{\mathbf{E}}_i(\mathbf{r}) + \tilde{\mathbf{E}}_s(\mathbf{r})$, where the scattered electric field is [24]

$$\tilde{\mathbf{E}}_s(\mathbf{r}) = \frac{1}{\epsilon_1} \left[\nabla \times \nabla \times \tilde{\mathbf{P}}_e(\mathbf{r}) + j \frac{n_1 k_1}{c} \nabla \times \tilde{\mathbf{P}}_m(\mathbf{r}) - \tilde{\mathbf{P}}_1(\mathbf{r}) \right] \quad (3)$$

with $\tilde{\mathbf{P}}_e(\mathbf{r}) = \frac{1}{4\pi} \int_V \tilde{\mathbf{P}}_1(\mathbf{r}') \frac{\exp(jk|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}'$ the electric Hertz vector, $\tilde{\mathbf{P}}_m(\mathbf{r}) = \frac{1}{4\pi} \int_V \tilde{\mathbf{M}}(\mathbf{r}') \frac{\exp(jk|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|} d\mathbf{r}'$ the magnetic Hertz vector, and V the volume of the perturbation medium. At a point $\mathbf{r} = r\mathbf{s}$ far from the scattering medium, $\tilde{\mathbf{P}}_e(\mathbf{r})$ and $\tilde{\mathbf{P}}_m(\mathbf{r})$ vary as $\frac{\exp(jk_1 r)}{r}$, and at any point outside the scattering medium $\tilde{\mathbf{P}}_1(\mathbf{r}) = \mathbf{0}$ and $\tilde{\mathbf{M}}(\mathbf{r}) = \mathbf{0}$. The scattered field may then be written as

$$\tilde{\mathbf{E}}_s(r\mathbf{s}) \sim \tilde{\mathbf{A}}(\mathbf{s}) \frac{\exp(jk_1 r)}{r}, \quad \tilde{\mathbf{H}}_s(r\mathbf{s}) \sim \frac{n_1}{\mu_0 c} \mathbf{s} \times \tilde{\mathbf{E}}_s(r\mathbf{s}) \quad (4)$$

with

$$\tilde{\mathbf{A}}(\mathbf{s}) = -k^2 \left(\mathbf{s} \times [\mathbf{s} \times \hat{\mathbf{P}}_1(k_1 \mathbf{s})] + \frac{n_1}{c} \mathbf{s} \times \hat{\mathbf{M}}(k_1 \mathbf{s}) \right), \quad (5)$$

where $\hat{\mathbf{P}}_1(\mathbf{k}) = \int_V \tilde{\mathbf{P}}_1(\mathbf{r}') \exp(-j\mathbf{k} \cdot \mathbf{r}') d\mathbf{r}'$ and $\hat{\mathbf{M}}(\mathbf{k}) = \int_V \tilde{\mathbf{M}}(\mathbf{r}') \exp(-j\mathbf{k} \cdot \mathbf{r}') d\mathbf{r}'$. For weakly scattering media, $E_s \ll E_i$, and $\tilde{\mathbf{P}}_1$ and $\tilde{\mathbf{M}}$ may be evaluated for the incident field. For a plane circular wave, $\tilde{\mathbf{E}}_i(\mathbf{r}) = \tilde{\mathbf{E}}_i \exp(jk_1 \mathbf{s}_0 \cdot \mathbf{r})$, where \mathbf{s}_0 is the incident direction. For scattering directions $\mathbf{s} \approx \mathbf{s}_0$, we then obtain

$$\tilde{\mathbf{A}}(\mathbf{s}) \sim \mathbf{s} \times \{ \mathbf{s}_0 \times [(\epsilon - \epsilon_1) \pm k_1 \beta (\epsilon + \epsilon_1)] \hat{\Theta}(\mathbf{s}_0, \mathbf{s}) \tilde{\mathbf{E}}_i \} \quad (6)$$

with $\hat{\Theta}(\mathbf{s}_0, \mathbf{s}) = \int_V \Theta(\mathbf{r}) \exp[-jk_1(\mathbf{s} - \mathbf{s}_0) \cdot \mathbf{r}] d\mathbf{r}'$, where $\Theta(\mathbf{r}) = 0$ if \mathbf{r} is in the solvent and 1 otherwise; the $-(+)$ sign stands for right-handed (left-handed) circular waves. The circular differential scattering is found in the first Born approximation and not in the second Born approximation as in [25] because we use nonisotropic polarization. The total scattered intensity is obtained by

integration of $\tilde{\mathbf{A}}(\mathbf{s}) \cdot \tilde{\mathbf{A}}^*(\mathbf{s})$ over all scattering directions and is $I_s \propto [(\epsilon - \epsilon_1) \pm \beta k_1 (\epsilon + \epsilon_1)]^2$. With $\delta n = n - n_1$ and $\Delta n = n_R - n_L$, we obtain for $|n_R - n_L| \ll n$

$$I_s \propto \left[\delta n \pm \frac{\Delta n}{2} \right]^2. \quad (7)$$

The transmitted intensity being $I_0 - I_s$, the difference between the transmission I_R of right-handed and I_L of left-handed circularly polarized waves is then in our approximation $I_R - I_L \propto \Delta n \delta n$. More generally, considering departure from the weakly scattering case, we calculate that I_s remains a function of $(\delta n \pm \frac{\Delta n}{2})$ at successive orders, so that

$$I_R - I_L = \Delta n \frac{dI}{d\delta n}, \quad (8)$$

where $I = (I_R + I_L)/2$.

IV. DISCUSSION

We plot in Fig. 3(a) the experimental difference $[I_R - I_L]/I_0$ as a function of the derivative of I/I_0 with respect to δn . Each line corresponds to a different mixture of *R* and *S* limonene. As expected, the scattering difference between right- and left-handed circular polarization varies proportionally to the derivative of the transmitted intensity. The value

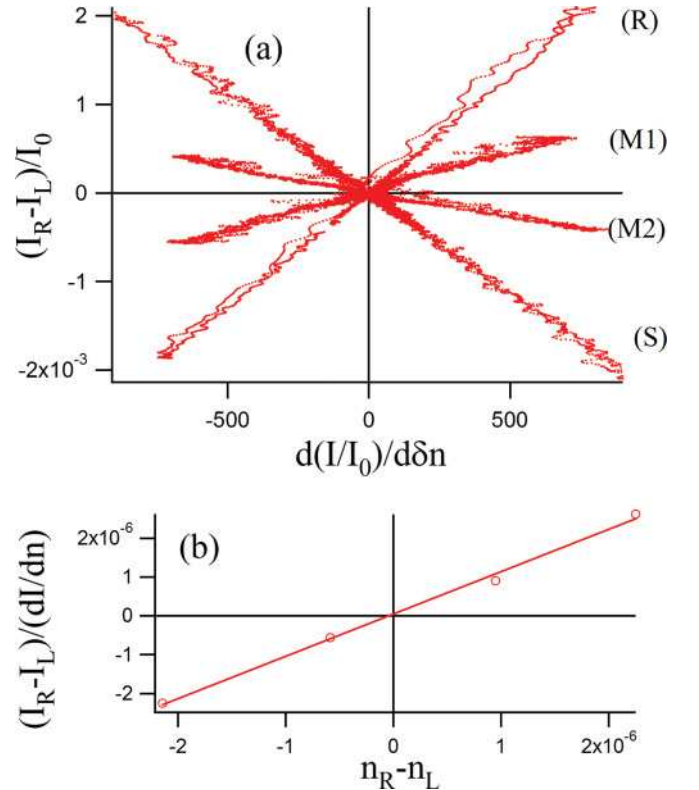


FIG. 3. (Color online) (a) Experimental difference of transmission between left and right circular waves as a function of the derivative of the total transmitted light with respect to the difference of refractive indices δn . Solutions are oil with limonene (*R*) and (*S*) and mixes (*M1*) and (*M2*) of (*R*) and (*S*). (b) Measured slopes from (a) plotted as a function of Δn ; the solid line corresponds to Eq. (8).

of Δn may be obtained from the slope $[I_R - I_L]/(dI/d\delta n)$. Figure 3(b) shows a plot of the deduced slopes as a function of Δn , measured independently from the rotation of the plane of polarization of light propagation through the liquid alone. We observe that Eq. (8) holds even far from the weakly scattering region, showing that our generalization beyond the weak-scattering limit is convincing.

The fluctuation σ of the refractive index of the beads diminishes the intensity of the transmitted light. Remarkably, the fluctuations do not modify the relation (8) even if $\Delta n \ll \sigma$. These fluctuations may be taken into account by considering $\epsilon(\mathbf{r}) = \bar{\epsilon} + \delta\epsilon(\mathbf{r}) = \bar{\epsilon} + \delta\epsilon\varphi(\mathbf{r})$, where $\bar{\epsilon}$ is the average of $\epsilon(\mathbf{r})$, $\delta\epsilon(\mathbf{r})$ is the fluctuation, $\delta\epsilon$ is the amplitude of the fluctuation, and $\varphi(\mathbf{r})$ is a function of order unity. Integration of $\hat{\mathbf{A}}(\mathbf{s}) \cdot \hat{\mathbf{A}}^*(\mathbf{s})$ over all the scattering directions shows that to the highest order $I_s \propto [(\epsilon - \epsilon_1) \pm \beta k_1(\epsilon + \epsilon_1)]^2 + \alpha[\delta\epsilon]^2$ with $\alpha = \int_s \hat{\varphi}(\mathbf{s})\hat{\varphi}^*(\mathbf{s})ds / \int_s \hat{\theta}(\mathbf{s})\hat{\theta}^*(\mathbf{s})ds$. So the fluctuations of $\epsilon(\mathbf{r})$ cancel out in the difference $I_R - I_L$ and in the derivative $dI/d\delta n$, and Eq. (8) still holds. Still, those fluctuations are responsible for the limitation of CDS by preventing divergence of $dI/d\delta n$ near phase matching.

Finally, Eq. (7) shows that the same value of $I_R - I_L$ can be obtained by simultaneously exchanging the material with its mirror image ($\Delta n \rightarrow -\Delta n$) and by changing ϵ to ϵ_1 ($\delta n \rightarrow -\delta n$). This may be simply understood. Let $\mathbf{E}_s^{(II)}$ and $\mathbf{E}_s^{(III)}$ be the fields scattered in the two experiments (II) and (III) depicted in Fig. 1(b). If we now consider the superposition of the two sources of radiation, the medium is homogeneous, and the total scattered field is null. Linearity of the Maxwell equations and of the constitutive equations (1) gives $\mathbf{E}_s^{(II)} + \mathbf{E}_s^{(III)} = 0$. Thus, the scattered intensities in the two experiments are the same. That effect is indeed observed experimentally, as

shown by the measurements for enantiomeric solutions on Fig. 3(a) and the symmetric form of the curves of Fig. 2(b). That invariance can be seen as a macroscopic analog of the CP symmetry [26]. A chiral medium lacks parity symmetry. In order to obtain invariance, charge conjugation should also be performed if time-inversion invariance is assumed [26,27]. Consider first the exchange of indices between two achiral media: i.e., changing the index of the inclusions from $\epsilon_1 + \delta\epsilon$ to $\epsilon_1 - \delta\epsilon$ while keeping the index of the surrounding medium constant at ϵ_1 . The effect of the transformation will be to change the sign of the polarization and consequently the sign of the scattered electric field. Such a transformation then gives the same result as a charge conjugation. Considering now chiral inclusions, the combination of the two transformations of charge conjugation and parity on the medium will result in an identical response to an excitation.

V. CONCLUSIONS

To conclude we have shown that circular differential scattering can be tuned in a weakly scattering material having a small constant value of $n_R - n_L$: it can be canceled, inverted, or adjusted in magnitude. Remarkable symmetry properties have been evidenced and justified.

ACKNOWLEDGMENTS

The authors thank P. Snabre for discussions and for the PMMA beads, G. Loas for help with the optical modulator measurement, and the team ‘‘Photonique et Laser’’ of Institut de Physique de Rennes for the loan of the PEM.

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