# Equivalency of the Casimir and the Landau-Lifshitz approaches to continuous-media electrodynamics and optical activity on reflection

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The controversial discussion on the existence of the specular optical activity phenomenon is reexamined. We prove here that the effect is predicted by approaches based both on the Casimir and the Landau-Lifshitz material equations and that these equations are in fact equivalent if crucial terms containing the derivatives of the nonlocal susceptibilities are taken into account in the derivation. Polarization azimuth rotation in the reflected wave of the scale $2\times10^{-6}$–$2\times10^{-4}$ rad may be expected in absorbing optically active crystals. We also show that specular optical activity is intrinsically linked with the transmission effect.

The phenomenon of optical activity, i.e., circular birefringence and circular dichroism affecting the polarization state of light propagating through a medium, is a basic and comprehensively explained optical phenomenon. Nevertheless, the controversial discussion[1–7] on the existence of optical activity on "reflection" from media exhibiting optical activity in transmission has not been completed to our knowledge. Moreover, we believe that the understanding of the electrodynamic forbiddance of this phenomenon,[3,4,6] which was thought to be backed by experimental results[4,6] and, which has been a widely accepted point of view for several years, is not correct and should be reexamined. Fresh interest in the topic of specular polarization effects in solids has been raised recently by (a) the prediction[8] and subsequent controversial experimental verification[9–12] that specular optical activity may be observed in some high-$T_c$ superconductors and (b) positive observation of specular optical activity from an $\alpha$-HgS crystal.[13] In fact the question of optical activity touches on a subject of a more general nature and greater importance, namely which is the correct material equation for the electrodynamics of a medium with nonlocal optical response?

In this paper we will approach the problem of optical activity on reflection from first principles. This will allow us to derive the correct forms of the material equation for two different treatments and to prove that specular optical activity is an allowed effect. The first treatment traditionally begins from the material equation known as the Landau-Lifshitz equation $\mathbf{D}=\epsilon\mathbf{E}+\gamma[\nabla\times\mathbf{E}]$,[14] and the second from the Casimir material equation, $\mathbf{P}=\chi^p\mathbf{E}+\theta\mathbf{B}$; $\mathbf{M}=\chi^m\mathbf{E}$.[15] It has already been established that adequate description of optical activity in transmission may be done equally successfully using either of these different starting points.[2] At the same time these two approaches have led to contradictory results in the treatment of optical activity on reflection.[2,4,6] Specifically, the Casimir formulism predicts no optical activity for normal reflection, while consideration of the Landau-Lifshitz equations leads to the existence of the phenomenon. Different authors have expressed conflicting opinions: While Natori[3] and Silverman and Badoz[5] expected no optical activity on normal reflection from an interface with an optically active medium, Bokut' and Serdyukov[1] and Val'kov, Romanov, and Shalaginov[7] did not deny the effect and Schlagheck[2] appealed for experimental verification of the controversial theoretical results.

In fact there have already been several attempts to resolve this problem by direct observation of the effect. The experiments concerning optical activity on normal reflection undertaken by Takizawa in a crystal of $TeO_2$ (Ref. 4) and by Luk'yanov and Novikov in a crystal of $LiIO_3$ (Ref. 6) gave negative results. The failure to get positive results in these experiments was, as we now understand, wrongly and prematurely considered as a decisive argument in favor of the Casimir approach. Our recent experimental results[13] show that in strongly optically active crystals of $\alpha$-HgS, in a region of strong absorption, optical activity on normal reflection exists and arises as a polarization azimuth rotation effect. We also believe that, in fact, positive observation could not be achieved in Refs. 4 and 6, since in both experiments it is likely that the experimental accuracy was below the value of the effect which might be expected.

We now understand that contradictions appearing in the use of the two treatments originate from the fact that the forms of the Casimir and the Landau-Lifshitz material equations used in Refs. 1–3, 5, and 7 were derived from different starting points. Consequently, the conditions of their validity are different. Here we will show that the Casimir and Landau-Lifshitz approaches are in fact two forms of one general material equation and consequently both are equally correct. We will also prove that the phenomenon of optical activity on reflection at normal incidence is an inevitable consequence of optical activity "in transmission." Polarization azimuth rotation on reflection however may be found in strongly absorbing materials only.

In order to examine the problem from first principles, we must first of all obtain the appropriate forms of the Maxwell equations on which the Casimir and the Landau-Lifshitz approaches are based. They may be derived from the most general form of the averaged microscopic equations for electromagnetic fields:[14]

$$[\nabla\times\mathbf{E}]=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t}\ , \tag{1}$$

$$(\nabla\cdot\mathbf{B})=0\ , \tag{2}$$

$$[\nabla\times\mathbf{B}]=\frac{1}{c}\frac{\partial\mathbf{E}}{\partial t}+\frac{4\pi}{c}\mathbf{J}\ , \tag{3}$$

$$(\nabla\cdot\mathbf{E})=4\pi\rho\ , \tag{4}$$

where $\mathbf{E}$ and $\mathbf{B}$ are, respectively, the electric-field strength and the magnetic induction in the medium and $\mathbf{J}$ and $\rho$ are the current and electric charge density, which are bound by the equation of continuity:

$$(\nabla\cdot\mathbf{J})=-\frac{\partial\rho}{\partial t}\ . \tag{5}$$

Hereafter, since we are interested in optical phenomena, we assume that there are no free charges, external electric currents or static magnetic fields involved and the media considered have no permanent magnetic moment.

The main difference between the Casimir and the Landau-Lifshitz approaches is the use of different variables in the material equations. While the Landau-Lifshitz approach operates with the strength of the electric field of the light wave and electric induction of the medium, the Casimir approach, which is designed to serve mostly magnetic problems, also involves the magnetic moment of the medium. In some sense the Casimir approach is more general, since it allows treatment of static magnetic-field phenomena. At the same time in the optics of nonmagnetic materials, specifically for the problem in question, there is no difference in their validity. We will show this below.

In order to develop the Landau-Lifshitz approach from the beginning, we substitute the following new variables into (1)–(5):

$$\mathbf{J}=\frac{\partial\mathbf{P}}{\partial t},\quad \mathbf{D}=\mathbf{E}+4\pi\mathbf{P}\ . \tag{6}$$

This leads to the Maxwell equations in the form

$$[\nabla\times\mathbf{E}]=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t}\ , \tag{7}$$

$$(\nabla\cdot\mathbf{B})=0\ , \tag{8}$$

$$[\nabla\times\mathbf{B}]=\frac{1}{c}\frac{\partial\mathbf{D}}{\partial t}\ , \tag{9}$$

$$(\nabla\cdot\mathbf{D})=0\ , \tag{10}$$

where $\mathbf{D}$ is the electric induction of the medium. For completeness the material equation $\mathbf{D}=\mathbf{D}(\mathbf{E},\mathbf{B})$ must be derived from the constitutive equation

$$D_i(\mathbf{r},t)=\int d\mathbf{r}'\int_{-\infty}^{t}dt'\psi_{ij}(\mathbf{r},\mathbf{r}',t-t')E_j(\mathbf{r}',t') +\int d\mathbf{r}'\int_{-\infty}^{t}\xi_{ij}(\mathbf{r},\mathbf{r}',t-t')B_j(\mathbf{r}',t')\ , \tag{11}$$

where the invariance against time translation, the nonlocality of the response, and the causality principle are presupposed. Here $\psi_{ij}$ and $\xi_{ij}$ are the optical response functions with the lower indices labeling the Cartesian coordinates. The dependence of $\psi_{ij}$ and $\xi_{ij}$ on $(t-t')$ reflects medium "memory," while dependence on $\mathbf{r}'$ indicates response nonlocality. We can rearrange this equation eliminating the magnetic field of the light wave by virtue of the first Maxwell equation:

$$\mathbf{B}(\mathbf{r},t)=-\int_{-\infty}^{t}dt'[\nabla\times\mathbf{E}(\mathbf{r},t')]\ . \tag{12}$$

The material equation can be rewritten in the equivalent form

$$D_i(\mathbf{r},t)=\int d\rho\int_0^{\infty}d\tau\,\epsilon_{ij}^{(1)}(\boldsymbol{\rho},r-\boldsymbol{\rho},\tau)E_j(\mathbf{r}-\boldsymbol{\rho},t-\tau)\ , \tag{13}$$

where $\epsilon_{ij}^{(1)}$ is a linear form of $\psi_{ij}$ and $\xi_{ij}$. The time-domain Fourier transform here results in

$$\mathbf{D}_i(\mathbf{r},\omega)=\int d\boldsymbol{\rho}\,\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r}-\boldsymbol{\rho},\omega)\mathbf{E}_j(\mathbf{r}-\boldsymbol{\rho},\omega)\ , \tag{14}$$

where

$$\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r}-\boldsymbol{\rho},\omega)=\frac{1}{2\pi}\int_0^{\infty}d\tau\,\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r}-\boldsymbol{\rho},\tau)\exp(i\omega\tau)\ . \tag{15}$$

The electromagnetic wave $E_i(\mathbf{r}-\boldsymbol{\rho},\omega)$ and the optical response function in (14) may be expanded as a series in powers of $\boldsymbol{\rho}$:

$$\mathbf{E}_i(\mathbf{r}-\boldsymbol{\rho},\omega)=\mathbf{E}_i(\mathbf{r},\omega)-\rho_l[\partial\mathbf{E}_i(\mathbf{r},\omega)/\partial r_l]+\cdots\ , \tag{16}$$

$$\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r}-\boldsymbol{\rho},\omega)=\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r},\omega) -\rho_l[\partial\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r},\omega)/\partial r_l]+\cdots\ . \tag{17}$$

We keep here only the first spatial derivatives, adopting the so-called first-order spatial dispersion approximation,[16] sufficient for description of the optical activity effect. If we introduce the dielectric tensor $\epsilon_{ij}$ and the nonlocality tensor $\gamma_{ijl}$,

$$\begin{aligned}\epsilon_{ij}(\mathbf{r},\omega)&=\int d\boldsymbol{\rho}\,\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r},\omega),\\ \gamma_{ijl}(\mathbf{r},\omega)&=\int d\boldsymbol{\rho}\,\rho_l\epsilon_{ij}^{(1)}(\boldsymbol{\rho},\mathbf{r},\omega)\ ,\end{aligned} \tag{18}$$

the material equation can now be presented in the Landau-Lifshitz form:

$$D_i(\mathbf{r},\omega)=\epsilon_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega) +\nabla_l[\gamma_{ijl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]+\cdots\ , \tag{19}$$

where the last term may be decomposed

$$\nabla_l[\gamma_{ijl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]=[\nabla_l\gamma_{ijl}(\mathbf{r},\omega)]E_j(\mathbf{r},\omega) +\gamma_{ijl}(\mathbf{r},\omega)[\nabla_lE_j(\mathbf{r},\omega)]\ . \tag{20}$$

The simplification of neglecting the first part of the right-hand side of (20) results in the shortened form of the Landau-Lifshitz material equation:[14]

$$D_i(\mathbf{r},\omega)=\epsilon_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)+\gamma_{ijl}(\mathbf{r},\omega)\nabla_lE_j(\mathbf{r},\omega)\ , \tag{21}$$

which is often used to describe propagation phenomena, such as natural optical activity. This simplification is justifiable only when the characteristic length of the variation of $\gamma_{ijl}(\mathbf{r},\omega)$ is sufficiently longer than the light wavelength $\lambda$, i.e., $|\nabla\gamma(\mathbf{r},\omega)| \ll |\gamma(\mathbf{r},\omega)/\lambda|$, and therefore not acceptable in the theory of reflection when boundary effects are described and material parameters such as $\gamma_{ijl}(\mathbf{r},\omega)$ change dramatically from one medium to another.

Now, using the same starting point we will derive the Casimir material equations.[15] Substitution of $\mathbf{J}=\partial\mathbf{P}/\partial t + c[\nabla\times\mathbf{M}]$, $\mathbf{D}=\mathbf{E}+4\pi\mathbf{P}$, and $\mathbf{H}=\mathbf{B}-4\pi\mathbf{M}$ into (1)–(5) leads to the use of variables adopted in this approach. This results in the following form of the Maxwell equations:

$$[\nabla\times\mathbf{E}]=-\frac{1}{c}\frac{\partial\mathbf{B}}{\partial t}\ , \tag{22}$$

$$(\nabla\cdot\mathbf{B})=0\ , \tag{23}$$

$$[\nabla\times\mathbf{H}]=\frac{1}{c}\frac{\partial\mathbf{D}}{\partial t}\ , \tag{24}$$

$$(\nabla\cdot\mathbf{D})=0\ . \tag{25}$$

Having one more variable $\mathbf{H}$ in comparison with the Landau-Lifshitz set of the Maxwell equations, two material equations must be introduced here, namely $\mathbf{M}=\mathbf{M}(\mathbf{E},\mathbf{B})$ and $\mathbf{P}=\mathbf{P}(\mathbf{E},\mathbf{B})$. As above, each of these constitutive equations should be presented in an integral form similar to (11). Again by exclusion of the magnetic induction $\mathbf{B}$ of the light wave, we can get the desired Casimir material equations:

$$P_i(\mathbf{r},\omega)=\chi^p_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)+\nabla_l[\gamma^p_{ijl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]\ , \tag{26}$$

$$M_i(\mathbf{r},\omega)=\chi^m_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)+\nabla_l[\gamma^m_{ijl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]\ . \tag{27}$$

Since we have two material equations, we have four optical susceptibilities $\chi^p_{ij}$, $\gamma^p_{ijl}$, $\chi^m_{ij}$, and $\gamma^m_{ijl}$, in comparison with only two $\epsilon_{ij}$ and $\gamma_{ijl}$ in the Landau-Lifshitz presentation. Again, the last terms in (26) and (27) may be decomposed and the contributions proportional to the spatial derivatives of $\gamma^p_{ijl}$ and $\gamma^m_{ijl}$ may be neglected when propagation phenomena are considered:

$$P_i(\mathbf{r},\omega)=\chi^p_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)+\gamma^p_{ijl}(\mathbf{r},\omega)\nabla_l E_j(\mathbf{r},\omega), \tag{28}$$

$$M_i(\mathbf{r},\omega)=\chi^m_{ij}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)+\gamma^m_{ijl}(\mathbf{r},\omega)\nabla_l E_j(\mathbf{r},\omega)\ . \tag{29}$$

Here also the simplification above should not be made when boundary effects are described and the complete form (26) and (27) should be used.

In Eqs. (26) and (27) there is no consideration of absorption and other limitations particularly arising from the fluctuation-dissipation theorem and the principle of microscopic reversibility.[17] We only presume in (16) and (17) that the material parameters and the fields vary slowly in the scale of the typical dimensions of the area forming the optical response at a particular point.

Now we come to the question of the equivalency of the Landau-Lifshitz and the Casimir material equations. As we have already mentioned above, it was considered as an established fact that these two different approaches lead to contradictory results in optics, specifically in the prediction of specular optical activity.[2,4,6] We point out here that this delusion results from comparison of the shortened form (28) and (29) of the Casimir material equation with the full form (19) of the Landau-Lifshitz Eq.,[2] while if the full forms (19) and (26) and (27) are used, this contradiction does not appear.

Evidently, being derived from the same basic principles, Eqs. (19) and (26) and (27) must produce the same results in the description of wave phenomena and moreover one of them may be transformed into the other and vice versa. In order to prove this, we mention here that the contribution to the current density associated with the susceptibility $\gamma^m$ in (27) is proportional to $\nabla\nabla\gamma^m E$ and, consequently is a second-order spatial dispersion term and must be neglected, i.e., $\gamma^m$ should be set to zero. By calculation of the current $\mathbf{J}$ in both approaches

$$J_i=-i\omega\{(1/4\pi)[\epsilon_{ij}(\mathbf{r},\omega)-\delta_{ij}]E_j(\mathbf{r},\omega) + \nabla_l[\gamma_{ijl}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]\}\ , \tag{30}$$

$$J_i=-i\omega\{\chi^p_{ij}(\mathbf{r},\omega)\mathrm{E_j}(\mathbf{r},\omega)+\nabla_\mathrm{l}[\gamma^\mathrm{p}_\mathrm{ijl}(\mathbf{r},\omega)\mathrm{E_j}(\mathbf{r},\omega)]\} + ce_{iln}\nabla_l[\chi^m_{nj}(\mathbf{r},\omega)E_j(\mathbf{r},\omega)]\ , \tag{31}$$

the remaining tensor coefficients in the Landau-Lifshitz material equation may be expressed in terms of the Casimir coefficients (as in the particular case of the Casimir presentation):

$$\begin{aligned}\epsilon_{ij}(\mathbf{r},\omega)&=\delta_{ij}+4\pi\chi^p_{ij}(\mathbf{r},\omega)\ ,\\ \gamma_{ijl}(\mathbf{r},\omega)&=4\pi[\gamma^p_{ijl}(\mathbf{r},\omega)+(ic/\omega)e_{iln}\chi^m_{nj}(\mathbf{r},\omega)]\ .\end{aligned} \tag{32}$$

The problems appearing from the fact that the three susceptibilities $\chi^p$, $\gamma^p$, and $\chi^m$ represent only two material parameters $\epsilon$ and $\gamma$ are resolved because only the linear combination $\gamma^p_{ijl}(\mathbf{r},\omega)+(ic/\omega)e_{iln}\chi^m_{nj}(\mathbf{r},\omega)$ is important in optics, since only it acts in the wave equation and only it may be measured. Consequently one can arbitrarily choose one of these linearly linked parameters. Specifically the Landau-Lifshitz Eq. (19) follows from the Casimir set (26) and (27), if $\chi^m_{nj}=0$.

As an illustration we consider below the simplest isotropic case ($\chi^p_{ij}=\chi^p\delta_{ij}$, $\gamma^p_{ijl}=\gamma^p e_{ijl}$, $\chi^m_{ij}=\chi^m\delta_{ij}$, $\gamma_{ijl}=\gamma e_{ijl}$, and $\epsilon_{ij}=\epsilon\delta_{ij}$), where the $L$-$L$ material equation takes the form

$$\mathbf{D}=\epsilon\mathbf{E}+[\mathbf{E}\times\nabla\gamma]+\gamma[\mathbf{E}\times\nabla]\ , \tag{33}$$

and the Casimir Eqs. (26) and (27) are

$$\mathbf{P}=\chi^p\mathbf{E}-\frac{i\omega}{c}\gamma^p\mathbf{B}+[\mathbf{E}\times\nabla\gamma^p]\ , \tag{34}$$

$$\mathbf{M}=\chi^m\mathbf{E}\ . \tag{35}$$

Here the magnetic field $\mathbf{B}=(c/i\omega)[\nabla\times\mathbf{E}]$ is reintroduced in order to obtain the usual Casimir form.[15] As a rule[2,4,6] the shortened form of the Casimir equation is used and the term $[\mathbf{E}\times\nabla\gamma]$ is neglected. Again the arguments produced above do not allow this simplification in boundary problems.

Now we are in a position to show how the correct material equation inevitably leads to the existence of the phenomenon of specular optical activity. We will prove this for the simplest example of an interface between a vacuum and an isotropic, gyrotropic medium. The

Landau-Lifshitz approach will be used, i.e., the Maxwell Eqs. (7)–(10) and material Eq. (31). For simplicity, step-like behavior of $\epsilon$ and $\gamma$ near the interface plane at $z=0$ is assumed, i.e., $\epsilon=1$ and $\gamma=0$ at $z<0$ and $\epsilon\neq 1$ and $\gamma\neq 0$ at $z>0$.

Evidently the form of the boundary conditions is material-equation dependent. The correct procedure for obtaining them has been developed in Ref. 7 and will be used below. From the Maxwell equation $[\nabla\times\mathbf{B}]=(1/c)\partial\mathbf{D}/\partial t$, by integration over the area of a closed loop partially immersed in the optically active medium ($z>0$) and partially remaining in the vacuum ($z<0$), in the limit as the loop size vanishes, the material equation for the magnetic induction of the Einstein-Maxwell wave may be found:

$$[(\mathbf{B}^{(1)}-\mathbf{B}^{(2)})\times\mathbf{n}]=\frac{1}{c}(-\gamma)\left[\frac{\partial\mathbf{E}}{\partial t}\times\mathbf{n}\right], \tag{36}$$

where the unit vector $\mathbf{n}$ is normal to the plane, and the indices (1) and (2) label the vacuum and gyrotropic medium, respectively. The right-hand side of this boundary condition is associated with the induced surface current. It arises only if the term $[\mathbf{E}\times\nabla\gamma]$ in the material equation discussed above is taken into consideration. Similarly from the Maxwell equation $[\nabla\times\mathbf{E}]=-(1/c)\partial\mathbf{B}/\partial t$ we get the second boundary equation:

$$[(\mathbf{E}^{(1)}-\mathbf{E}^{(2)})\times\mathbf{n}]=0\ . \tag{37}$$

Optical wave reflection coefficients may now easily be calculated for the normal incidence condition. The electric field in the vacuum $\mathbf{E}^{(1)}$ is given by

$$\mathbf{E}^{(1)}=\mathbf{E}_i\exp(-i\omega t+ikz)+\mathbf{E}_r\exp(-i\omega t-ikz)+\text{c.c.}\ , \tag{38}$$

where $\mathbf{E}_i$ and $\mathbf{E}_r$ are correspondingly the magnitudes of the incident and the reflected waves and $k=\omega/c$. In an isotropic, gyrotropic medium the eigenwaves are circularly polarized and have different wave vectors given by $q_\pm=kn_\pm$, $n_\pm=\pm(k\gamma/2)+[\epsilon+(k\gamma/2)^2]^{1/2}$,[14] i.e., the electric field of the transmitted light has the following form:

$$\mathbf{E}^{(2)}=\mathbf{e}_+E_{t+}\exp(-i\omega t+iq_+z)+\mathbf{e}_-E_{t-}\exp(-i\omega t+iq_-z)+\text{c.c.}\ , \tag{39}$$

where $\mathbf{e}_\pm=1/\sqrt{2}.\ (\mathbf{i}\pm i\mathbf{j})$ are the right and left circular polarization vectors ($\mathbf{i}$ and $\mathbf{j}$ are the unit vectors along the $x$ and $y$ axes). Ignorance of the difference between the left and the right wave vectors leads to the loss of the phenomena of optical activity on reflection.[3] We can now express the boundary conditions for the circularly polarized components of the incident ($i$), the reflected ($r$) and transmitted ($t$) waves:

$$E_{i\pm}+E_{r\pm}=E_{t\pm}\ , \tag{40}$$

$$E_{i\pm}-E_{r\pm}=n_\mp\cdot E_{t\pm} \tag{41}$$

where $E_{i,r\pm}=(\mathbf{e}_\pm\mathbf{E}_{i,r})$ are the magnitudes of the eigenwaves. Thus the reflection amplitude coefficients $\mathbf{R}_\pm$ of the circularly polarized waves are equal to

$$\mathbf{R}_\pm=\frac{E_{r\pm}}{E_{i\pm}}=\frac{1-n_\mp}{1+n_\mp}\ . \tag{42}$$

The polarization state of the waves may be presented in terms of ellipticity angle $\eta=(1/2)\sin^{-1}(s_3/s_0)$ and angle of rotation of the polarization azimuth $\alpha=(1/2)\tan^{-1}(s_2/s_1)$, where $s_m$ ($m=0\ldots 3$) are the Stokes parameters: $s_0=E_+E_+^*+E_-E_-^*$, $s_1=E_+E_-^*+E_-E_+^*$, $s_2=i(E_-E_+^*-E_+E_-^*)$, $s_3=E_+E_+^*-E_-E_-^*$. If the incident wave is linearly polarized and the polarization change of the reflected waves is small ($\delta\eta_r\ll\pi,\delta\alpha_r\ll\pi$) then

$$\delta\eta_r+i\delta\alpha_r=\frac{k\gamma}{(1-\epsilon)}\ . \tag{43}$$

We shall note here one important peculiarity predicted by the formula (43); optical activity on reflection from the interface between two media with close dielectric constants may be very strong, since the denominator of the right-hand side of (43) tends to zero when the dielectric constants on either side of the border converge (i.e., $\epsilon$ tends to 1 in the case above). The border between a perfect optically active crystal and an amorphous layer of the same material (exhibiting no optical activity) would be the most evident but not the only example. A similar enhancement of the specular polarization effect has already been mentioned for reflection from nonlinear anisotropic media.[18]

Close similarity between Eq. (43) and the formula describing the optical activity phenomenon in transmission,

$$\delta\eta_t+i\delta\alpha_t=\frac{k\gamma}{\lambda}i\pi z \tag{44}$$

can be seen. It is clear from (43) and (44) that polarization change in transmission is inevitably linked with specular effects and that one may be expressed in terms of the other:

$$(\delta\eta_r+i\delta\alpha_r)=-\frac{i\lambda}{\pi(1-\epsilon)}\frac{\partial(\delta\eta_t+i\delta\alpha_t)}{\partial z}\ . \tag{45}$$

Here the term $\partial(\delta\alpha_t)/\partial z$ is known as specific optical rotatory power in units [rad/cm]. However, there is a dramatic difference between the two phenomena. In order to make it clear we take derivatives of Eqs. (43) and (44) with respect to $z$:

$$\frac{\partial(\delta\eta_r+i\delta\alpha_r)}{\partial z}=\frac{k}{(1-\epsilon)}\frac{\partial\gamma}{\partial z}=\frac{k}{(1-\epsilon)}\gamma\delta(z)\ , \tag{46}$$

$$\frac{\partial(\delta\eta_t+i\delta\alpha_t)}{\partial z}=i\pi\frac{k\gamma}{\lambda}\ . \tag{47}$$

In the last equation we have taken into account that the propagation phenomenon is considered for $z>0$, where the medium is homogeneous and $\gamma$ is presumed to be independent of $z$. It may now be clearly seen from Eq. (46) that $\partial\gamma/\partial z$ is indeed responsible for specular optical activity and consequently this contribution must not be ignored either in the Landau-Lifshitz material Eq. (19) or the Casimir-type Eqs. (26) and (27). In our case of step-like behavior of $\epsilon$ and $\gamma$, $\partial\gamma/\partial z=\gamma\delta(z)$. Here $\delta(z)$ is the Dirac delta function and the reflected light polarization change appears when the wave reaches $z=0$. Eventually any longitudinal inhomogeneity of the medium resulting in a coordinate dependence of $\gamma$ will lead to the alteration of the reflected light polarization. If the transitional surface layer is significantly thinner than the light wavelength, the resulting effect on the reflected light polarization may be expressed in the form

$$(\delta\eta_r+i\delta\alpha_r)=\frac{k}{1-\epsilon}\int_{-\infty}^{+\infty}dz\frac{\partial\gamma}{\partial z}\ . \tag{48}$$

In some sense the specular effect is "the complex conjugated phenomenon" with respect to optical activity in transmission [see (45)]. It may lead to both polarization plane rotation and light elliptization. Specifically a linearly polarized incident wave reflected from a transparent medium ($\mathrm{Im}\{\epsilon\}=0$, $\mathrm{Im}\{\gamma\}=0$) becomes elliptically polarized but no polarization azimuth rotation may be observed. Polarization azimuth rotation in the reflected wave may only appear as a result of reflection from an absorbing crystal where either $\mathrm{Im}\{\gamma\}$ or $\mathrm{Im}\{\epsilon\}$ exists. Table I summarizes the features of both the effects.

For optically active crystals such as $\alpha$-HgS, $ZnP_2$, $LiIO_3$, $TeO_2$, and $Bi_{12}SiO_{20}$ specific polarization rotation power in the transparent range is 10–500 deg/mm, i.e., 2–80 rad/cm, while $\epsilon$ is always around 6–10. Thus $\delta\eta_r=[\lambda/\pi(1-\epsilon)]\partial(\delta\alpha_t)/\partial z$ and an ellipticity angle may be expected in the range of $4\times10^{-6}$–$2\times10^{-4}$ rad. Correspondingly the relative intensity coming from a "crossed" ideal analyzer is $\simeq(\delta\eta_r)^2$, i.e., $2\times10^{-11}$–$4\times10^{-8}$. As far as we are concerned, one of the possible explanations for the failure of detection of specular optical activity in the experiments, Refs. 4 and 6, is that the experimental accuracy in Refs. 4 and 6 was probably below the value of $\simeq(\delta\eta_r)^2=4\times10^{-8}$, predicted by (43), which is indeed a very small effect.

For absorbing crystals the reflected wave is elliptically polarized with different polarization azimuth with respect to the incident wave. By expressing the imaginary part of $\epsilon$ in terms of the absorption coefficient $\xi$, one can estimate the polarization azimuth rotation as

TABLE I. Relationships between the optical activity phenomenon and the material parameters of isotropic media.

| | $\mathrm{Re}\{\gamma\}$ | $\mathrm{Im}\{\gamma\}$ |
|---|---|---|
| | Optical activity "on reflection" | |
| $\mathrm{Re}\{1-\epsilon\}$ | Elliptization | Rotation |
| $\mathrm{Im}\{1-\epsilon\}$ | Rotation | Elliptization |
| | Optical activity "on transmission" | |
| $\mathrm{Re}\{\epsilon\}$ | Rotation | Elliptization |
| $\mathrm{Im}\{\epsilon\}$ | Rotation | Elliptization |

$$\delta\alpha_r=\frac{\lambda^2\xi n}{2\pi^2(1-n^2)^2}\frac{\partial(\delta\alpha_t)}{\partial z}\ , \tag{49}$$

i.e., specular rotation is proportional not only to the specific rotation in transmission $[\partial(\delta\alpha_t)/\partial z]$, but also to the strength of absorption. In a range of strong absorption, where $\xi$ may be as big as $10^3$–$10^4$ cm$^{-1}$, and the rotation power is stronger than in the transparent range, typically 1000–10 000 deg/mm, (i.e., 200–2000 rad/cm), the polarization azimuth rotation on reflection may be in the scale of $2\times10^{-6}$–$2\times10^{-4}$ rad, which is accessible by modern polarization-sensitive detection techniques. Recent positive experiments on observation of specular polarization rotation in $\alpha$-HgS (Ref. 13) confirm this estimate and the whole treatment derived above.

Summarizing in conclusion, it is shown here that both the Casimir and the Landau-Lifshitz approaches to the electrodynamics of optically nonlocal media are equivalent if the crucial terms containing the derivatives of the nonlocal susceptibilities are taken into account. Both of them inevitably predict the existence of optical activity on normal reflection from an interface with a medium exhibiting optical activity in transmission. Polarization azimuth rotation on reflection may be found only in optically active crystals in an area of sufficient absorption. In optically transparent media only elliptization of initially linearly polarized light may be expected with normal reflection.

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