# Features of light reflection off metals with destroyed mirror symmetry

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Features of the electromagnetic field induced by the interaction with polar conductors, i.e., conducting crystals whose symmetry with respect to reflection in a plane perpendicular to some axis (the polar axis) is broken, is considered. As distinct from conventional, centrosymmetric metals, the constitutive relations of Maxwell's equations in such materials (which relate the electric current and the magnetization to the electric and the magnetic field) are modified to include cross terms which relate the magnetization to the electric field and the electric current to the magnetic field. It is found that due to the cross terms (i) the reflection of light depends on the sign of the product $\mathbf{n} \cdot \mathbf{c}$, where $\mathbf{n}$ is the normal to the surface of the metal and $\mathbf{c}$ is the direction of the polar axis, i.e., depends upon the polar-axes direction, and (ii) in the general case the electromagnetic wave in such materials should have a longitudinal component.

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It is well known that the lack of central symmetry of crystals and molecules (in solutions or gases) may have unusual effects on their macroscopic properties. A most remarkable example is the phenomenon of optical activity when right- and left-handed circular polarized light waves propagate through active media at slightly different speeds (for a review see, e.g., Ref. 1). It is the purpose of this Brief Report to consider optical properties of another class of materials with destroyed space parity—conductors of polar symmetry. The doped $A_2B_6$ semiconductor CdS of the wurtzite structure (space group $C_{6v}^4$) apparently was the first conducting medium of polar symmetry whose electromagnetic properties were investigated.[2] At the present time, quite a number of metals, whose symmetry group includes a polar axis, is known. These are, for example, $Mo_3AlC$ (space $P4_132$, $Mo_3P$ (space group $I\bar{4}$),[3] and ternary intermetallic silicides of the general formula $RTSi_3$ (space group $I4mm$), where $R$ represents rare-earth metals and $T$ represents transition metals.[4] The main feature of polar conductors, which for the first time has been revealed by the study of CdS, consists of a peculiar form of their electron spectrum. Because from the symmetry viewpoint the existence of a polar axis is equivalent to the constant electric field, the electron Hamiltonian acquires a term of spin-orbital origin that couples the electron's momentum and spin[5]

$$H_{so} = \alpha(\mathbf{p} \times \mathbf{c}) \cdot \boldsymbol{\sigma}, \tag{1}$$

where $\alpha$ is a constant characteristic of the crystal, $\mathbf{c}$ is the unit vector directed along the polar axis, $\mathbf{p}$ is the momentum operator, and $\boldsymbol{\sigma}$ represents the spin Pauli matrices. Polar metals, as well as asymmetrical two-dimensional (2D) semiconductor heterostructures where the spin-orbit coupling of form (1) is also present, becomes nowadays the focus of intense research: the bulk metals—since some of them turn into superconductors with uncommon properties (for a review see, e.g., Ref. 6) and both the metals and 2D structures—since the band spin-orbit coupling gives hope to find a way to control the spin dynamics by means of the electric field only, i.e., without the help of the magnetic field (for a review see, e.g., Ref. 7).

Searches for uncommon electrodynamic properties of polar conducting media were initiated by a paper[8] where the magnetoelectric effect (MEE) has been predicted, which states that if, under the action of an electric field $\mathbf{E}$, the electric current $\mathbf{J} \sim \mathbf{E}$ passes through the system, it induces the spin magnetization of the carriers proportional to $\mathbf{c} \times \mathbf{E}$.[9] Afterwards, the MEE has apparently been experimentally confirmed.[10] Another anomalous property of a polar conducting medium is that the circular polarized electromagnetic wave is capable to induce in the medium the same effects as a constant magnetic field: (i) The circular polarized light wave (with the electric field $\mathbf{E}$) induces a permanent spin magnetization $\mathbf{M} \sim i\mathbf{c}(\mathbf{c} \cdot \mathbf{E} \times \mathbf{E}^*)$ which means the inverse Faraday effect (IFE),[11] and (ii) if the constant electric current $\mathbf{J}$ passes through the system, the circular wave induces the Hall-like current $\mathbf{J}_H \sim i(\mathbf{c} \times \mathbf{J})(\mathbf{c} \cdot \mathbf{E} \times \mathbf{E}^*) = \mathbf{M} \times \mathbf{J}$, which means the zero-magnetic-field Hall effect (ZMFH).[12] A common trait of the effects mentioned above is that all of them are consequences of the electron kinetics modified by the presence of the band spin-orbit coupling (BSOC) expressed by the Hamiltonian (1). In other words, they are anomalous responses of the *matter* to a *given* electromagnetic field. From the other hand, it is natural to expect that the electromagnetic field *itself* can acquire peculiar properties due to the interaction with a medium whose behavior is free from constraints imposed by the invariance under space reflections. The goal of the present Brief Report is to reveal such properties in the light reflection off a polar metal.

Consider the normal incidence of light when the polar axis is perpendicular to the surface of the metal. As is known, the standard macroscopic electrodynamics[13] supposes that properties of a conducting medium enter the Maxwell's equations

$$\nabla \times \mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{B}}{\partial t}, \tag{2}$$

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

$$\nabla \cdot \mathbf{B} = 0, \quad \nabla \cdot \mathbf{E} = 4\pi\rho, \quad \mathbf{H} = \mathbf{B} - 4\pi\mathbf{M}, \tag{4}$$

where $c$ is the velocity of light, through the so-called constitutive relations. The relations express the electric current $\mathbf{J}$ and the electron magnetization $\mathbf{M}$ through the electric and magnetic fields, **E** and **B**, in the medium. In polar metals, the constitutive relations have the form[14]

$$\mathbf{M} = \chi \mathbf{B} + \gamma \mathbf{E} \times \mathbf{c}, \tag{5}$$

$$\mathbf{J} = \sigma \mathbf{E} + \gamma \frac{\partial \mathbf{B}}{\partial t} \times \mathbf{c}. \tag{6}$$

The presence of the cross terms of this form is a specific property of a conducting media of polar symmetry;[15] two terms in the right-hand side of the equations have opposite parity under space inversion. The magnetoelectric susceptibility $\gamma$ is a material characteristic additional to the electric conductivity $\sigma$ and the magnetic susceptibility $\chi$. Since these equations satisfy the polar symmetry requirements, they can be viewed as phenomenological equations. But they can be also derived microscopically by considering a model electron system with the Hamiltonian $H = H_0 + H_{so} + H_{\text{imp}}$, where $H_0 = \frac{\mathbf{p}^2}{2m}$ is the operator of the kinetic energy of the Fermi gas, $H_{so}$ is given by Eq. (1), and the Hamiltonian $H_{\text{imp}}$ accounts for the electron impurity scattering. Then the equations are obtained as linear responses of the current and the spin magnetization densities to the electric and magnetic fields. Strictly speaking, Eqs. (5) and (6) were derived in Ref. 14 for 2D systems. An analogous analysis of a 3D system with the same Hamiltonian gives rise to the same equations with

$$\gamma_\omega = \frac{\gamma}{1 - i\omega\tau_{so}}, \quad \gamma = \alpha \left(\frac{eg\mu_B}{2\pi^2}\right) m p_F \tau, \tag{7}$$

where $e$ and $\mu_B$ are the electron charge and the Bohr magneton, $m$ and $g$ are the effective electron mass and $g$ factor, $p_F$ is the Fermi momentum, $\tau$ is the mean electron lifetime, and $\tau_{so} = 3\tau(2\alpha p_F \tau)^{-2}$ is the spin-relaxation time due to impurity scattering in the presence of the BSOC.[16] Note that $\tau_{so} \gg \tau$ at small $\alpha$ and short $\tau$. Equation (7) has been written taking into account the frequency dispersion; in the following, for the sake of simplicity, we confine ourselves to the case of low frequencies $\omega\tau \ll 1$, $\omega\tau_{so} \ll 1$. We shall also neglect the magnetic susceptibility $\chi$ assuming it very small and treat $\gamma$ as being small so that all powers of $\gamma$ in excess of the first can be ignored. By excluding the magnetic field $\mathbf{H}_{\mathbf{r},\omega}$ from Maxwell's equations and neglecting terms quadratic in $\gamma$ we obtain the electric field wave equation in the form

$$\nabla \times [\nabla \times \mathbf{E}_{\mathbf{r},\omega}] - \frac{\omega^2}{c^2}\mathbf{E}_{\mathbf{r},\omega} - \frac{4\pi i\omega}{c^2}\sigma(\mathbf{r})\mathbf{E}_{\mathbf{r},\omega} = \frac{4\pi i\omega}{c}\gamma(\mathbf{r})[(\mathbf{c}\times\nabla)\times\mathbf{E}_{\mathbf{r},\omega}] + \frac{4\pi i\omega}{c}\nabla\gamma(\mathbf{r})\times(\mathbf{E}_{\mathbf{r},\omega}\times\mathbf{c}). \tag{8}$$

For the metal occupying the homogeneous half space $z \leqslant 0$, the coordinate dependence of the conductivity and the magnetoelectric susceptibility is given by

$$\begin{pmatrix}\sigma(\mathbf{r})\\ \gamma(\mathbf{r})\end{pmatrix} = \begin{pmatrix}\sigma\\ \gamma\end{pmatrix}\Theta(-\mathbf{n}\cdot\mathbf{r}), \tag{9}$$

where **n** is the external normal to the surface of the metal and $\Theta(z)$ is the Heaviside step function. Since

$$\nabla\gamma(\mathbf{r})\times(\mathbf{E}_{\mathbf{r},\omega}\times\mathbf{c}) = -\delta(\mathbf{n}\cdot\mathbf{r})[\mathbf{E}_{\mathbf{r},\omega}(\mathbf{n}\cdot\mathbf{c}) - \mathbf{c}(\mathbf{n}\cdot\mathbf{E}_{\mathbf{r},\omega})], \tag{10}$$

the last term on the right-hand side of Eq. (8) is a surface term. It is this term which does not turn to zero at the normal incidence. The wave equation (8) can be solved following the wave-vector-space method proposed in Ref. 17. Below, only the logic and results of the application of the method to polar metals are presented; for details the reader is referred to the original paper. The Fourier transform of Eq. (8) is

$$\mathbf{q}^2\mathbf{E}_{\omega,\mathbf{q}} - \mathbf{q}(\mathbf{q}\cdot\mathbf{E}_{\omega,\mathbf{q}}) - q_0^2\left[\mathbf{E}_{\omega,\mathbf{q}} - \frac{4\pi i\sigma}{\omega}\int\frac{dq_z'}{2\pi i}\frac{\mathbf{E}_{\omega,\mathbf{q}'}}{q_z - q_z' - i0}\right] = -\frac{4\pi\omega}{c}\gamma\int\frac{dq_z'}{2\pi i}\left[\frac{(\mathbf{c}\times\mathbf{q}')\times\mathbf{E}_{\omega,\mathbf{q}'}}{q_z - q_z' - i0} - \mathbf{n}\times(\mathbf{E}_{\omega,\mathbf{q}'}\times\mathbf{c})\right], \tag{11}$$

where $\mathbf{q}' = (q_x, q_y, q_z')$. Further, it is assumed that the electric field can be written as a sum of two functions, $\mathbf{E}_{\mathbf{r},\omega} = \mathbf{E}^{(+)}_{\mathbf{r},\omega} + \mathbf{E}^{(-)}_{\mathbf{r},\omega}$, one of which, $\mathbf{E}^{(+)}_{\mathbf{r},\omega}$, is responsible for a mode propagating in the medium while another, $\mathbf{E}^{(-)}_{\mathbf{r},\omega}$, corresponds to modes propagating in the vacuum. Accordingly, the transform $\mathbf{E}_{\omega,\mathbf{q}}$ can be written as a sum of two functions, $\mathbf{E}^{(+)}_{\omega,\mathbf{q}}$, which has poles only in the upper half complex **q** plane, and $\mathbf{E}^{(-)}_{\omega,\mathbf{q}}$, which has poles only in the lower half complex **q** plane. Note that only the component $q_z$ is a true variable in $\mathbf{E}_{\omega,\mathbf{q}}$, $q_x$ and $q_y$ being fixed by the problem definition. In the case of the orthogonal incidence $\mathbf{E}^{(\pm)}_{\omega,\mathbf{q}} = (2\pi)^2\delta(q_x)\delta(q_y)\mathbf{E}^{(\pm)}_\omega(q_z)$, where

$$\mathbf{E}^{(+)}_\omega(q_z) = \frac{E_0}{i}\frac{t\mathbf{e}_3}{q_z - q_M - i0},$$
$$\mathbf{E}^{(-)}_\omega(q_z) = -\frac{E_0}{i}\left[\frac{\mathbf{e}_1}{q_z - q_V + i0} + \frac{r\mathbf{e}_2}{q_z + q_V + i0}\right]. \tag{12}$$

Here $E_0$ is a given incident electric field amplitude, $r$ is a reflection coefficient, $t$ is a transmission coefficient, $\mathbf{e}_1$, $\mathbf{e}_2$, and $\mathbf{e}_3$ are as yet undetermined unit vectors, and positions of the poles $q_M$ and $q_V$ are determined by the dispersion relations of the medium and the vacuum, respectively. It can be directly verified that the dispersion relation of the medium coincides with that of an ordinary metal up to perturbations quadratic in $\gamma$ so that $q_M = q_0\sqrt{\epsilon_\omega}, q_V = q_0$, where $q_0 = \frac{\omega}{c}$ and $\epsilon_\omega = 1 + \frac{4\pi i}{\omega}\sigma$. The substitution of Eqs. (12) into Eq. (11) gives rise to the equations

$$-\mathbf{e}_1 - r\mathbf{e}_2 + t\mathbf{e}_3 = 0,$$
$$-\mathbf{e}_1 + r\mathbf{e}_2 + t\mathbf{e}_3[\sqrt{\epsilon_\omega} - 4\pi\gamma(\mathbf{n}\cdot\mathbf{c})] = 0, \tag{13}$$

the solution to which has the form

$$\mathbf{e}_1 = \mathbf{e}_2 = \mathbf{e}_3,$$

$$r \cong \frac{\sqrt{\epsilon_\omega} - 1}{\sqrt{\epsilon_\omega} + 1}\left[1 - \frac{8\pi\gamma}{\epsilon_\omega - 1}(\mathbf{n}\cdot\mathbf{c})\right],$$
$$t \cong \frac{2}{\sqrt{\epsilon_\omega} + 1}\left[1 + \frac{4\pi\gamma}{1 + \sqrt{\epsilon_\omega}}(\mathbf{n}\cdot\mathbf{c})\right]. \tag{14}$$

Thus despite that the electromagnetic wave propagating along the polar axis of the *infinite* metal does not feel (with the adopted accuracy) the absence of mirror symmetry, it is sensitive to the orientation of the polar axis in the case of the *half space* metal. At higher frequencies when the conditions $\omega\tau \ll 1$ and $\omega\tau_{so} \ll 1$ are not fulfilled, Eqs. (14) remain to be valid if one replaces $\sigma$ and $\gamma$ with the dynamic kinetic coefficients $\sigma_\omega$ and $\gamma_\omega$, where $\sigma_\omega = \sigma(1 - i\omega\tau)^{-1}$ and $\gamma_\omega$ is given by Eq. (7).

The evaluation of the light reflection at an arbitrary angle of incidence is out of the scope of the present Brief Report. Nevertheless, to show an exceptional character of the reflection at the normal incidence, we mention here a physical consequence of the modified Maxwell's equations that may play a role at the oblique incidence. Consider a wave in the bulk. For a single frequency propagation, $\mathbf{E},\mathbf{H} \sim \exp\{-i[\omega t - \mathbf{q}\cdot\mathbf{r}]\}$, the Maxwell's equations with the adopted accuracy yield

$$\mathbf{H}_{\omega,\mathbf{q}} = \frac{1}{q_0}(\mathbf{q} + 4\pi\gamma q_0\mathbf{c}) \times \mathbf{E}_{\omega,\mathbf{q}},$$
$$\epsilon_\omega\mathbf{E}_{\omega,\mathbf{q}} = -\frac{1}{q_0}(\mathbf{q} - 4\pi\gamma q_0\mathbf{c}) \times \mathbf{H}_{\omega,\mathbf{q}}. \quad (15)$$

Thus just as in the case of conventional metal, $\mathbf{E}_{\omega,\mathbf{q}} \perp \mathbf{H}_{\omega,\mathbf{q}}$, but opposed to that both the fields $\mathbf{E}_{\omega,\mathbf{q}}$ and $\mathbf{H}_{\omega,\mathbf{q}}$ do not have to be orthogonal to the wave vector $\mathbf{q}$ except for the case $\mathbf{q}\|\mathbf{c}$, i.e., they contain longitudinal components. (Nevertheless, one can show that the equality $\mathbf{q}\cdot\mathbf{B}_{\omega,\mathbf{q}} = 0$ remains valid.) It follows from Eqs. (15) that

$$i\left(\frac{\mathbf{q}}{q_0}\cdot\mathbf{E}_{\omega,\mathbf{q}}\right) \cong \gamma\frac{\omega}{\sigma}\left(\frac{\mathbf{q}}{q_0}\right)^2(\mathbf{c}\cdot\mathbf{E}_{\omega,\mathbf{q}}). \quad (16)$$

In the coordinate space,

$$\nabla\cdot\mathbf{E}_{\mathbf{r},t} \cong -\gamma\frac{c}{\sigma}\nabla^2(\mathbf{c}\cdot\mathbf{E}_{\mathbf{r},t}). \quad (17)$$

The nonzero value of the longitudinal component of $\mathbf{E}_{\omega,\mathbf{q}}$ means that the plane wave should be accompanied by a perturbation of the electron density, i.e., should induce a local violation of the electrical neutrality. This anomalous perturbation does not appear if $(\mathbf{c}\cdot\mathbf{E}_{\mathbf{r},t}) = 0$, in particular, when the electromagnetic wave propagates along the polar axes. It should be noticed that the possibility of a $P$ polarized wave at non-normal incidence to give rise to $\nabla\cdot\mathbf{E} \neq 0$ within a metal is known (see, e.g., Refs. 18). However, in the case of a conventional metal, it is caused by the presence of the surface and takes place under the anomalous-skin-effect conditions on account of the nonlocality (the space dispersion) of the conductivity tensor. As opposed to that, the induced density oscillations in the case under consideration are a volume effect that takes place when the length of the electromagnetic wave exceeds both the mean free path and the field penetration depth, and the frequency of the electromagnetic field is less than the inverse impurity scattering time.

The values of $\alpha$ in the metallic systems mentioned are not determined yet. For doped CdS crystals with $m = 0.2m_0$ and $g = 1.8$, it has been found by means of the spin-flip inelastic light scattering[2] that $\alpha p_F \simeq 0.3$ meV at at the electron density $n = 1.8 \times 10^{18}$ cm and the mean free time $\tau \simeq 10^{-13}$ sec. Thus $\alpha \simeq 1.3 \times 10^5$ cm sec$^{-1}$. It follows from Eq. (7) that

$$\gamma \simeq \left(\frac{e^2}{\hbar c}\right)\left(\frac{m}{m_0}\right)\left(\frac{\alpha p_F\tau}{\hbar}\right)\left(\frac{g}{2\pi^2}\right), \quad (18)$$

hence $\gamma \simeq 3\times 10^{-6}$. To find a reliable way to measure $\alpha$ in dirty metals is an actual problem.

In summary, we have considered the propagation of electromagnetic waves in a metal of polar symmetry. In a particular case of the normal incidence of a wave on the metal with the surface perpendicular to the polar axes, the problem of the light reflection has been solved. It has been found that the wave feels the absence of the mirror symmetry in the metal in the sense that the coefficients of reflection and transmission of the wave depend on the scalar product $(\mathbf{n}\cdot\mathbf{c})$, where $\mathbf{n}$ is the external normal to the surface of the metal and $\mathbf{c}$ is the polar direction, i.e., they change at $\mathbf{c} \to -\mathbf{c}$. The effect could be experimentally detected by comparing the light reflection off two opposite oriented domains of the crystal. It has been also found that at an arbitrary direction of the propagation with respect to the polar axis, the wave cannot be pure transversal, hence it should be accompanied by a perturbation of the electron density. The disturbance disappears in the limit of long wavelength so that the uniform electric field does not disturb the electron density.

It would be interesting to consider in detail the case of the light reflection at the oblique incidence that could reveal additional distinctive features of optics of polar metals. An account of effects of the space dispersion, which can be anomalous in crystals with destroyed mirror symmetry subject to an external magnetic field (see, for example, Refs. 2 and 19), is also of interest. Solution of these problems could be of importance for polar superconductors as well.

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