Inverse Faraday Effect in Conducting Crystals Caused by a Broken Mirror Symmetry

Victor M. Edelstein*

Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka Moscow Region, Russia
(Received 15 September 1997)

We consider how the absence of mirror crystal symmetry influences electron kinetics of polar metals, semiconductors, and asymmetric semiconductor hererostructures, as well as some layered antipyroelectric high-T_c compounds. We find that illumination of such crystals by circularly polarized infrared light should induce a permanent spin orientation of current carriers in an amount proportional to $i\mathbf{c}(\mathbf{c} \cdot \boldsymbol{\epsilon} \times \boldsymbol{\epsilon}^*)$, where $\boldsymbol{\epsilon}$ is the polarization vector, and $\mathbf{c}$ is a unit vector directed either along the polar or antipyroelectric axis or perpendicular to the semiconductor heterostructure. [S0031-9007(98)06289-9]

Searches for and investigations of uncommon properties which materials can possess owing to the absence of mirror symmetry of their crystal structure always attracted considerable interest both from the pure scientific point of view and because of possible device application. At least three groups of such *conducting* materials are known at present. First, these are bulk polar (doped) A_2B_6 semiconductors of the wurtzite structure with space group C_{6v}^4. A_2B_6 heterostructures and some other semiconductor heterostructures with violated "up-down" symmetry, such as $In_xGa_{1-x}As/In_yAl_{1-y}As$ [1] and GaSb/InAs [2], can also be placed into this group. Recently synthesized ternary intermetallic silicides [3] of the general formula RTSi$_3$ (where R represents rare-earth metals and T represents transition metals), which crystallize in the BaNiSn$_3$ structure with space group $I4mm$ and, hence, are polar (pyroelectric) metals, fall into the second group. The third group consists of some high-T_c compounds, e.g., YBa$_2$Cu$_3$O$_{7-x}$ (YBCO). Indeed, the nearest environment of CuO$_2$ layers is asymmetric: The layers are surrounded by yttrium ions on one side and by a Ba-O plane on the other side. Such compounds may be viewed as layered antipyroelectrics because their elementary cell contains two asymmetric and oppositely oriented CuO$_2$ layers [4].

The loss of the up-down symmetry means the existence of the intracrystalline electric field acting on the carriers. The presence of the field adds the term

$$H_{\text{s.o.}} = \frac{\alpha}{\hbar}(\mathbf{p} \times \mathbf{c}) \cdot \boldsymbol{\sigma} \qquad (1)$$

to the Hamiltonian of the carriers [5], where $\mathbf{p}$, $\boldsymbol{\sigma}$, and $\mathbf{c}$ are, respectively, the electron momentum, the Pauli matrices, and a unit vector directed along the field. The purpose of this paper is to point out a novel phenomenon which must take place due to the band spin-orbit (s.o.) coupling (1), i.e., due to the broken space parity. That is, the infrared circular polarized light induces (on the penetration length) a permanent spin polarization of the carriers. In essence, this nonlinear effect is nothing but a rectification of currents excited by the light wave. The result of this

rectification is, however, the permanent spin rather than a permanent current.

Yet, before a formal derivation, let us give heuristic arguments. The term (1) can be considered as the Zeeman energy in a fictitious magnetic field $\mathbf{B}_f = \alpha(\mathbf{p} \times \mathbf{c})/\mu_B g$. The field can influence the electron kinetics in two ways. First, it stochastically changes its direction by impurity scattering providing an additional channel for the spin relaxation [6]. Second, if, under the action of an electric field $\mathbf{E}$, the electric current $\mathbf{J} \sim \mathbf{E}$ passes through the system (so that the ensemble-averaged momentum $\langle \mathbf{p} \rangle \sim \mathbf{J}$), the average field $\langle \mathbf{B}_f \rangle$ induces a spin polarization of the carriers proportional to $\mathbf{c} \times \mathbf{E}$ [7]. This magnetoelectric effect (MEE) means the appearance of a correction to the electron distribution function proportional to $\boldsymbol{\sigma} \cdot (\mathbf{c} \times \mathbf{E})$ under the action of the driving electric field $\mathbf{E}$. In the case under study, the driving field is the radiation field which can be presented as $\mathbf{E}_\Omega e^{i\Omega t} + \mathbf{E}_{-\Omega} e^{-i\Omega t}$, where because of reality of the field $\mathbf{E}_{-\Omega} = \mathbf{E}_\Omega^*$. One can assume that in the second order perturbation theory the field may induce a correction to the distribution function of the form $[\boldsymbol{\sigma} \cdot (\mathbf{c} \times \mathbf{E}_\Omega)][\boldsymbol{\sigma} \cdot (\mathbf{c} \times \mathbf{E}_{-\Omega})]$, which is nothing but $i(\boldsymbol{\sigma} \cdot \mathbf{c})(\mathbf{c} \cdot \mathbf{E}_\Omega \times \mathbf{E}_\Omega^*)$. Thus one may expect that illumination of the system by the circular polarized light can induce a permanent spin polarization. It should be stressed that the mechanism does not suppose any interband electron transitions. In this respect, the effect differs from the well-investigated optical orientation effects in semiconductors [8], when the angular momentum of absorbing photon transfers (according to selection rules) into a creating photoelectron in the conduction band and a photohole in the valence band. It resembles the inverse Faraday effect (IFE) predicted for transparent dielectrics [9,10] and molecular systems [11]. Both effects are induced by a s.o. coupling. A fundamental difference is that the effect under discussion has the inherent kinetic nature and takes place in dissipative media. Its mechanism, as it has been mentioned, is based on the linear MEE, $\langle \boldsymbol{\sigma} \rangle \sim \mathbf{c} \times \mathbf{E}$, which would be forbidden in

nonabsorbing nonmagnetic media because of time reversal symmetry. (In dissipative media, the invariance to time reflection is violated in the presence of electric currents due to the Joule heat production.) The kinetic, dissipative character of electronic processes involved is clearly seen from calculations which follow. This is the presence of R-A section in the Feynman diagrams responsible for the effect as well as the strong dependence of the result, Eq. (8), on the spin relaxation times.

In addition to the spin relaxation channel due to $H_{s.o.}$, there can be other reasons for spin-flip events as well. We consider in this paper the magnetic impurity scattering as a representative of all spin-nonconserving processes that are not related to $H_{s.o.}$. Thus, first of all keeping in mind the asymmetric semiconductor structures and the CuO_2 layers, we consider degenerate 2D Fermi gas of independent electrons of charge e and spin $\frac{1}{2}$ scattered by normal and magnetic impurities (3D systems can be treated quite analogously). Then the Hamiltonian is written as

$$H = H_o(\mathbf{p}) + H_{\text{imp}}, \tag{2}$$

$$H_0 = \frac{\mathbf{p}^2}{2m} + \alpha(\mathbf{p} \times \mathbf{c}) \cdot \boldsymbol{\sigma},$$

$$H_{\text{imp}} = \sum_i \left(u + \frac{1}{2} v \boldsymbol{\sigma} \cdot \mathbf{S}_i\right) \delta(\mathbf{r} - \mathbf{R}_i). \tag{3}$$

Here $\mathbf{r}$ is the position of an electron, and $\mathbf{R}_i$ are the positions of the arbitrary distributed short-range impurities of concentration n_{imp}. In Eqs. (2) and (3) and below, we set $\hbar = 1$. We wish to determine the effect of an alternating electric field described by the transverse ($\nabla \cdot \mathbf{A} = 0$) vector potential in the interaction Hamiltonian $H_{\text{int}} = -(e/c)\mathbf{v} \cdot \mathbf{A}$, where the velocity operator of the system $\mathbf{v} = i[H_0(\mathbf{p}), \mathbf{r}] = \mathbf{p}/m + \alpha(\mathbf{c} \times \boldsymbol{\sigma})$, in addition to the usual vector part also has a spin-vector component. As it is known [12], to study dynamic properties of a many-body system, the general procedure is to first write down certain imaginary-time-ordered thermal correlation functions, and then to perform analytic continuation from the positive imaginary frequency axis to the real frequency axis, with respect to every external frequency, in or-

der to obtain the corresponding physical, causal correlation functions. The spin polarization is given by the expression

$$M(\omega_n, \mathbf{q}) = T \sum_{\epsilon_l} \int \frac{d^2 p}{(2\pi)^2} \operatorname{Tr} \frac{1}{2}$$
$$\times \boldsymbol{\sigma} G(\epsilon_l, \epsilon_l - \omega_n; \mathbf{p}, \mathbf{p} - \mathbf{q}), \tag{4}$$

where the electron Green function G is assumed to be exact in the sense that it includes, besides the external alternating field, also the impurity self-energies. By expanding G in powers of the external field, one gets in the second order one fermion loop, threaded through three frequency bearing vertices (which include one response spin vertex and two cause field vertices). After the analytical continuation, such a diagram becomes four diagrams: one retarded diagram, one advance diagram [which does not contribute to the magnetization with the accuracy up to $(\epsilon_F \tau)^{-1}$, ϵ_F being the Fermi energy and τ the relaxation time], and two "kinetic" diagrams beginning with the retarded and ending with the advanced free Green function. Since we are interested in the permanent uniform polarization, we can now set $\omega = \mathbf{q} = 0$. The latter two diagrams are shown in Fig. 1. The free Green function (single line) has the form

$$G_{\alpha\beta}^{R(A)}(\zeta, \mathbf{p}) = \sum_{\nu=\pm} \prod_{\alpha\beta}^{(\nu)}(\mathbf{p}) \left[\zeta - E_\nu \pm \frac{i}{2\tau}\right]^{-1}, \tag{5}$$

where $E_\pm = p^2/2m \pm \alpha p$ are the energies of the two branches of the energy spectrum of $H_0(\mathbf{p})$ of positive and negative helicities (the projection of a spin on the $\mathbf{p} \times \mathbf{c}$ direction), and $\prod^\pm(\mathbf{p}) = \frac{1}{2}[1 \pm (\hat{\mathbf{p}} \times \mathbf{c}) \cdot \boldsymbol{\sigma}]$ are the projection operators on these branches. The difference between the diagram technique used and the standard one [13] consists in the spinor structure of the free Green functions and the changed form of the velocity operator. We consider the case of high light frequency $\Omega\tau \gg 1$, therefore the impurity renormalization of the field vertices can be neglected. To evaluate the diagrams, one needs to perform three operations: (i) a spin-angle integration with respect to $\hat{\mathbf{p}}$, (ii) a "radial" integration with respect to $\xi = p^2/2m - \epsilon_F$, and (iii) a calculation of the impurity renormalized $\boldsymbol{\sigma}$ vertex. The first step can be performed with the help of the straightforwardly verified equations

$$\int \frac{d\hat{\mathbf{p}}}{2\pi} \operatorname{Tr}\left\{\prod^{(\mu)}(\mathbf{n} \cdot \boldsymbol{\sigma}) \prod^{(\lambda)} \frac{p_i}{m} \prod^{(\kappa)} \alpha(\mathbf{c} \times \boldsymbol{\sigma})_j + \prod^{(\mu)}(\mathbf{n} \cdot \boldsymbol{\sigma}) \prod^{(\lambda)} \alpha(\mathbf{c} \times \boldsymbol{\sigma})_i \prod^{(\kappa)} \frac{p_j}{m}\right\}$$
$$= \alpha \frac{P}{m} \frac{i}{4} (\operatorname{sgn}\mu - \operatorname{sgn}\lambda)(\mathbf{n} \cdot \mathbf{c}) e_{ijk} c^k (\delta_{\mu\kappa} - \delta_{\lambda\kappa}), \tag{6}$$

$$\int \frac{d\hat{\mathbf{p}}}{2\pi} \operatorname{Tr}\left\{\prod^{(\mu)}(\mathbf{n} \cdot \boldsymbol{\sigma}) \prod^{(\lambda)} \alpha(\mathbf{c} \times \boldsymbol{\sigma})_i \prod^{(\kappa)} \alpha(\mathbf{c} \times \boldsymbol{\sigma})_j\right\} = \alpha^2 \frac{i}{4}(\mathbf{n} \cdot \mathbf{c}) e_{ijk} c^k (1 - \operatorname{sgn}\mu \operatorname{sgn}\lambda),$$

where $\mathbf{n}$ is an arbitrary vector, and the letters μ, λ, and κ stand for the helicity of electron states shown in Fig. 1 (each of them can be +1 or −1). Note that the vector structure of our result, $\mathbf{M} \sim i\mathbf{c}(\mathbf{c} \cdot \boldsymbol{\epsilon} \times \boldsymbol{\epsilon}^*)$, is already seen from these
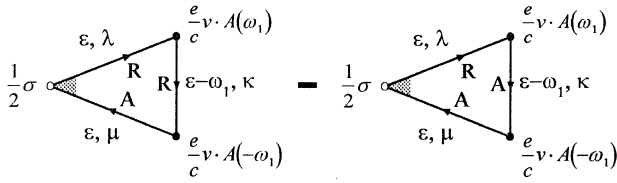
FIG. 1. Diagrams for the inverse Faraday effect. The common factor $(1/4\pi i)\{\tanh[(\epsilon - \omega_1)/2T] - \tanh(\epsilon/2T)\}$ is implicit.

equations. The radial integration is carried out in the usual manner [13], with the only exception being that two energy branches $E_\pm(\mathbf{p})$ have different Fermi momenta $p_\pm = p_F \mp \alpha m$, $p_F = (2m\epsilon_F)^{-1/2}$. The impurity renormalization of the spin vertex also can be performed in the standard way [13]. As a result, at $|v| \ll |u|$ and $\alpha p_F \tau \ll 1$,

$$(\mathbf{c} \cdot \boldsymbol{\sigma}) \to (\mathbf{c} \cdot \boldsymbol{\sigma})\left(\frac{\tau}{\tau_S} + \frac{\tau}{\tau_{\text{s.o.}}}\right)^{-1},$$

$$\frac{1}{\tau_{\text{s.o.}}} = \frac{1}{\tau}(\alpha p_F \tau)^2,$$

$$\frac{1}{\tau_S} = mn_{\text{imp}}|v|^2 \frac{1}{3} S(S+1), \qquad (7)$$

where $\tau^{-1} = mn_{\text{imp}}[|u|^2 + \frac{1}{4}|v|^2 S(S+1)]$. One should also take into account that, at light frequency Ω, the frequency ω_1 in Fig. 1 can be Ω as well as $-\Omega$. In this way, one gets

$$\mathbf{M} = i\mathbf{c}(\mathbf{c} \cdot \boldsymbol{\epsilon} \times \boldsymbol{\epsilon}^*)n_2 K_\Omega,$$

$$K_\Omega = \left(\frac{eE_\Omega \tau}{p_F}\right)^2 \frac{1}{2\pi\epsilon_F \tau}(\Omega\tau)^{-3}\frac{\tau/\tau_{so}}{\tau/\tau_{so} + \tau/\tau_S}, \qquad (8)$$

where $n_2 = p_F^2/\pi$ is the 2D electron density, $\boldsymbol{\epsilon}$ is the polarization, and E_Ω is the amplitude of the light field. The last factor in (8) tends to unity when the Dyakonov-Perel' process provides the fastest spin relaxation and becomes $\tau_S/\tau_{\text{s.o.}}$ when other processes dominate.

The existence of the band s.o. coupling (1) is firmly established by now in materials of the first group. It was found that $\alpha(\text{CdS}) = 1.6 \times 10^{-10}$ eV cm [14], $\alpha(\text{InGa}_{1-x}\text{As}/\text{InAl}_{i-y}\text{As}) = 1.4 \times 10^{-10}$ eV cm [1], and $\alpha(\text{GaSb}/\text{InAs}) = 9 \times 10^{-10}$ eV cm [2]. The role of this coupling in physics of the silicides and high-T_c materials remained unrevealed yet. To some extent, it is because experimental methods used previously to study the s.o. in semiconductors, e.g., spin-flip electronic Raman scattering [14] and especially a beating pattern in the amplitude of the conductivity magneto-oscillations [1,2], are not suitable in investigating dirty metals. It is desirable, therefore, to find an effect that could be observed in all three groups of materials. To be observable in YBCO, such an effect would have to satisfy an additional

condition—it should be equal for all CuO_2 sheets; i.e., it should be even under $\mathbf{c} \to -\mathbf{c}$ substitution. The effect presented remarkably meets all of these conditions and, therefore, may appear to be a useful tool in studying the broken reflectional symmetry and determining the value of α in the silicides and high-T_c materials as well. To obtain a crude, tentative estimate of α in YBCO, one can make use of the relation [14]

$$\frac{\alpha}{\hbar}(\mathbf{p} \times \mathbf{c}) \cdot \boldsymbol{\sigma} = \sum_{n,m} \frac{\langle c|V_\mathbf{p}|n\rangle}{E_n - E_c}\langle n|V_{\text{s.o.}}|m\rangle \frac{\langle m|V_{\text{St}}|c\rangle}{E_m - E_c}$$

$$+ (V_\mathbf{p} \leftrightarrow V_{\text{St}}), \qquad (9)$$

where $|c\rangle$ refers to the conduction band, $V_{\text{St}} = e\mathbf{Er} \cdot \mathbf{c}$ is the Stark energy due to the crystalline electric field $\mathbf{E}$, $V_{\text{s.o.}} = \zeta \mathbf{l} \cdot \boldsymbol{\sigma}/2$ is the intra-atomic s.o. energy, $V_\mathbf{p} = m^{-1}(-i\mathbf{p} \cdot \nabla)$ is the so-called $\mathbf{k} \cdot \mathbf{p}$ perturbation energy, and the sum is over the other appropriate bands. It is seen from (9) that the value of the s.o. constant α of the conduction band, which is presumably made up of the oxygen orbitals, is determined *not* by s.o. coupling in oxygen but by those nth bands, which are formed from wave functions of heavier atoms provided that the electron transitions from $|c\rangle$ to the bands are dipole allowed. For the nth band contribution to the $H_{\text{s.o.}}$, one has $(ip/2\omega_{nc})$. $[\langle c|\mathbf{r}V_{\text{s.o.}}|n\rangle\langle n|V_{\text{St}}|c\rangle - \langle c|V_{\text{St}}|n\rangle\langle n|V_{\text{s.o.}}\mathbf{r}|c\rangle]$. The corresponding contribution to the s.o. energy, $\alpha p_F/\hbar$, is roughly equal to $(k_F x_{cn})(\zeta/\omega_{nc})(eEz_{nc})$, where $\hat{x}$ lies in the a-b plane, and $\hat{z}$ is directed along the c axis. Let us suppose that the Cu:3d orbital is the major constituent of the nth band. Then, even if $|c\rangle$ is composed of the O:2p orbitals positioned only in the CuO_2 sheet, both matrix elements x_{cn} and z_{nc} can simultaneously be different from zero owing to the puckering of the sheet, because the deviation of the oxygen ions from the plane spanned by the copper ions is known [15] to result in an admixture of p_z character into the oxygen orbital contribution to $|c\rangle$. The magnitude of the local electric field in YBCO can amount to [16] 0.03–0.07 $e/\text{Å}^2$, and $\zeta(\text{Cu}:3d)$ is known to be [17] $\simeq 800$ cm^{-1}. If one adopts the typical values of the matrix elements $\langle c|x|n\rangle \simeq 4\langle c|z|n\rangle \simeq 1$ Å, the electric field $E \simeq 0.03$ $e/\text{Å}^2$, and the energy gap $E_n - E_c \simeq 3$ eV [18], then the nth band contribution to αk_F equals ~ 50 K at $k_F \simeq 3 \times 10^7$ cm^{-1} [19]. Thus we obtain $\alpha k_F \sim 10^2$ K; i.e., $\alpha(\text{YBCO}) \sim 10^{-10}$ eV cm may be of the same order of magnitude as in the semiconductors. It is difficult to give any estimate for the silicides because their band structure is not determined at present; yet heavy elements the compounds are composed of and strong mirror symmetry violation allow one to expect an appreciable value of α.

It is seen from (8) that strong disorder as well as small Fermi energy, i.e., proximity to a metal-insulator transition, favors the effect. Among the substances mentioned above, the largest magnitude of the spin polarization can be expected in the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{In}_y\text{Al}_{1-y}\text{As}$

heterostructure. For the fairly dirty heterostructure with $m^* = 0.046 m_0$, $n = 6 \times 10^{11}$ cm^{-2}, $k_F \simeq 2 \times 10^6$ cm^{-1}, $\epsilon_F \simeq 25$ meV, $\hbar/\tau \simeq 10$ meV, and for the far infrared ($\lambda = 100 \, \mu$m) pulse laser of power $P = 10^2$ W focused on the structure of dimension 1 mm^2, one has $K_\Omega \simeq 10^{-3}$.

In closing this paper, we would like to emphasize once more that IFE is common to all three groups of conductors with destroyed reflectional parity. One can expect the effect to show in NMR experiments, in dc transport (as the appearance of Hall voltage under no magnetic field), and in optical experiments (e.g., through a change of optical constants in the presence of the pump circularly polarized laser). It is not difficult to distinguish IFE reported here from that [10,11] not connected with the broken symmetry. In the case of the copper oxides, it is sufficient to compare high-T_c cuprates with asymmetric (e.g., YBCO) and symmetric (e.g., La$_{2-x}$Sr$_x$CuO$_4$) CuO$_2$ layers. The similar approach can also be used for asymmetric and symmetric semiconductor quantum wells.

---

*Present address: edelsh@issp.ac.ru

[1] B. Das, S. Datta, and R. Reifenberger, Phys. Rev. B **41**, 9278 (1990).

[2] J. Luo, H. Munekata, F. F. Feng, and P. J. Stiles, Phys. Rev. B **38**, 10 142 (1988).

[3] P. Haen *et al.,* J. Less-Common Met. **110**, 321 (1985).

[4] V. M. Edelstein, Phys. Rev. Lett. **75**, 2004 (1995).

[5] E. I. Rashba, Fiz. Tverd. Tela (Leningrad) **1**, 407 (1959) [Sov. Phys. Solid State **1**, 366 (1959)]; R. C. Gasella, Phys. Rev. Lett. **5**, 371 (1960); F. J. Ohkawa and Y. Uemura, J. Phys. Soc. Jpn. **37**, 1325 (1974).

[6] M. I. Dyakonov and V. I. Perel', Fiz. Tverd. Tela (Leningrad) **13**, 3581 (1971) [Sov. Phys. Solid State **13**, 3023 (1972)].

[7] V. M. Edelstein, Solid State Commun. **73**, 233 (1990).

[8] G. Lampel, Phys. Rev. Lett. **20**, 491 (1968); R. R. Parsons, *ibid.* **23**, 1132 (1969). For a review, see B. P. Zakharchenya *et al.,* Usp. Fiz. Nauk **136**, 459 (1982) [Sov. Phys. Usp. **25**, 143 (1982)].

[9] L. P. Pitayevskii, Zh. Eksp. Teor. Fiz. **39**, 1450 (1960) [Sov. Phys. JETP **12**, 1008 (1961)]; L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1983), Chap. XI.

[10] P. S. Pershan, Phys. Rev. **130**, 919 (1963).

[11] See, for example, M. W. Evans, Mod. Phys. Lett. B **5**, 1533 (1991), and references therein.

[12] L. P. Gor'kov and G. M. Eliashberg, Zh. Eksp. Teor. Fiz. **54**, 612 (1968) [Sov. Phys. JETP **27**, 328 (1968)].

[13] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskii, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, NJ, 1963).

[14] R. Romestain, S. Geshwind, and G. E. Devlin, Phys. Rev. Lett. **39**, 1583 (1977).

[15] J. K. Burdett and G. V. Kulharni, Phys. Rev. B **40**, 8909 (1989).

[16] H. Lee and R. P. Sharma, Phys. Rev. B **43**, 7756 (1991).

[17] A. Abraham and B. Bleaney, *Electron Paramagnetic Resonance of Transition Ions* (Clarendon, Oxford, 1970).

[18] High-T_c cuprates exhibit several optical bands with energies in the range from midinfrared to ultraviolet [see, for example, M. Shimida *et al.,* Physica (Amsterdam) **193C**, 353 (1992), and references therein]. The energy gap 3 eV has been chosen as a characteristic quantity.

[19] D. R. Harshmann and A. P. Mills, Phys. Rev. B **45**, 10 684 (1992).