

Theory of the High-Frequency Chiral Optical Response of a $p_x + ip_y$ Superconductor

Victor M. Yakovenko

Department of Physics, University of Maryland, College Park, Maryland 20742-4111, USA

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The optical Hall conductivity and the polar Kerr angle are calculated as functions of temperature for a two-dimensional chiral $p_x + ip_y$ superconductor, where the time-reversal symmetry is spontaneously broken. The theoretical estimate for the polar Kerr angle agrees by the order of magnitude with the recent experimental measurement in Sr_2RuO_4 by Xia *et al.* [Phys. Rev. Lett. **97**, 167002 (2006)]. The theory predicts that the Kerr angle is proportional to the square of the superconducting energy gap and is inversely proportional to the cube of frequency, which can be verified experimentally.

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Xia *et al.* [1] recently reported experimental observation of the polar Kerr effect in the superconducting state of Sr_2RuO_4 . In the absence of an external magnetic field, reflected light shows rotation of polarization, which is a clear signature of the spontaneous time-reversal-symmetry breaking in the superconducting state [2]. Previous muon spin relaxation measurements [3] suggested the time-reversal symmetry is broken in Sr_2RuO_4 , but the polar Kerr experiment [1] gives a much more convincing evidence for this remarkable effect.

Sr_2RuO_4 consists of weakly coupled two-dimensional (2D) metallic layers. It was proposed theoretically that the superconducting pairing in this material is spin-triplet [4] and has the chiral $p_x + ip_y$ symmetry [5]. Such an order parameter breaks the time-reversal symmetry and is analogous to the 2D superfluid $^3\text{He-A}$ [6]. There is substantial experimental evidence in favor of the spin-triplet and odd orbital symmetry of the superconducting pairing in Sr_2RuO_4 [7], which includes measurements of the spin susceptibility [8] and the Josephson effect [9] (see, however, an alternative interpretation [10]). On the other hand, the chiral character was not so well established experimentally (see Ref. [11] for interpretation of tunneling measurements).

Although the experimental demonstration [1] of the spontaneous polar Kerr effect is very convincing, a theory of this effect for chiral superconductors is not well developed. Theories [12,13] concluded that there is no chiral term in the single-particle response of a $p_x + ip_y$ superconductor, although Fig. 1 of Ref. [12] shows a nonzero Kerr effect for a different state, and Ref. [13] found some chiral response from collective excitation. On the other hand, Ref. [6] obtained the intrinsic quantum Hall effect in the single-particle response of a $p_x + ip_y$ superconductor, which was then studied in much detail in Ref. [14]. Following Refs. [1,12,15], the polar Kerr angle θ_K can be expressed in terms of the imaginary part of the ac Hall conductivity $\sigma''_{xy}(\Omega)$ at a frequency Ω

$$\theta_K = \frac{4\pi}{n(n^2 - 1)\Omega d} \sigma''_{xy}(\Omega), \quad (1)$$

where n is the refraction coefficient. We write Eq. (1) in terms of the 2D Hall conductivity σ_{xy} per one layer, which is related to the 3D one via the interlayer distance d . The natural dimensional scale for the 2D σ_{xy} is e^2/h .

In this Letter, we calculate the ac Hall conductivity $\sigma_{xy}(\Omega)$ at a finite frequency Ω as a function of temperature T for a $p_x + ip_y$ superconductor. We generalize the method of Refs. [6,14] and obtain the Chern-Simons-like term in the effective action at finite T and Ω . In the intermediate calculations, we set the Planck constant to unity $\hbar \rightarrow 1$, but restore it in the final results. The Lagrangian of electrons $L = i\partial_t - H$ (where H is the Hamiltonian) for the 2D $p_x + ip_y$ superconductor has the form [6]

$$L = \begin{pmatrix} i\partial_t + \nabla^2/2m + \mu & i(\nabla \cdot \Psi + \Psi \cdot \nabla)/2 \\ i(\nabla \cdot \Psi^* + \Psi^* \cdot \nabla)/2 & i\partial_t - \nabla^2/2m - \mu \end{pmatrix}. \quad (2)$$

Here ∂_t and $\nabla = (\partial_x, \partial_y)$ represent time and space derivatives, m and μ are the mass and the chemical potential of the electrons. We assume the parabolic dispersion law $\varepsilon(\mathbf{p}) = \mathbf{p}^2/2m - \mu$, where $\mathbf{p} = (p_x, p_y)$ is the electron momentum. The superconducting order parameter is $\Psi = \Delta_x \hat{x} + i\Delta_y \hat{y}$, where \hat{x} and \hat{y} are the unit vectors in the x and y directions. Because of the square symmetry in Sr_2RuO_4 , we have $\Delta_x = \Delta_y$, but it is convenient to label the two components of Ψ differently for the clarity of calculations. In momentum representation, Eq. (2) can be written as

$$L = i\omega - \varepsilon(\mathbf{p})\tau_3 - p_x \Delta_x \tau_1 + p_y \Delta_y \tau_2, \quad (3)$$

where the Pauli matrices τ act on the spinor $[\psi(\mathbf{p}), \psi^+(-\mathbf{p})]$ consisting of the particle and hole operators. We do not write the spin indices of electrons explicitly. The two spin components give the same contributions to the Hall conductivity, so the final results should be multiplied by 2. However, by introducing electron and hole operators, we artificially doubled the number of components, so the final result should be divided by 2 [6]. Thus, we can obtain the correct result by considering just one spin component, as implied in Eq. (3). In Eq. (3), we use

the Matsubara frequency $i\omega$, because we will be doing calculations at a finite temperature. From Eq. (3), the Green function of electrons $G = L^{-1}$ is

$$G(\vec{p}) = -\frac{i\omega + \varepsilon(\mathbf{p})\tau_3 + p_x\Delta_x\tau_1 - p_y\Delta_y\tau_2}{\omega^2 + E^2(\mathbf{p})}, \quad (4)$$

where $\vec{p} = (\omega, \mathbf{p})$ is the three-component frequency-momentum vector, and $E(\mathbf{p}) = \sqrt{\varepsilon^2(\mathbf{p}) + p_x^2\Delta_x^2 + p_y^2\Delta_y^2}$ is the electron dispersion in the superconducting state.

To calculate an electromagnetic response of the system, we introduce the electromagnetic potentials $\vec{A} = (A_0, A_x, A_y)$ by using the long derivatives $-i\nabla \mp e\mathbf{A}/c$ and $-i\partial_t \pm eA_0$ in the diagonal terms of Eq. (2), where e is the electron charge, and c is the speed of light [6]. We also assume that the superconducting order parameter has a space-time-dependent phase φ , so that it can be written as $\Psi = e^{i\varphi}\Psi_0$, where Ψ_0 is uniform with the real Δ_x and Δ_y . We will see that the effective action depends only on gradients of φ . To the first order in \vec{A} and φ , we find the following addition to the Lagrangian:

$$\Gamma = \begin{pmatrix} -eA_0 - i\frac{e\mathbf{A}\cdot\nabla}{mc} & -\frac{\nabla\cdot\Psi_0\varphi + \varphi\Psi_0\cdot\nabla}{2} \\ \frac{\nabla\cdot\Psi_0^*\varphi + \varphi\Psi_0^*\cdot\nabla}{2} & eA_0 - i\frac{e\mathbf{A}\cdot\nabla}{mc} \end{pmatrix}. \quad (5)$$

$$C_1 = \frac{1}{2}\text{Tr}[-eA_0(\vec{q})\tau_3 + ep_xA_x(\vec{q})/mc + ep_yA_y(\vec{q})/mc - \varphi(\vec{q})p_x\Delta_x\tau_2 - \varphi(\vec{q})p_y\Delta_y\tau_1][i(\omega + \Omega/2) + \varepsilon(\mathbf{p} + \mathbf{q}/2)\tau_3 + (p_x + q_x/2)\Delta_x\tau_1 - (p_y + q_y/2)\Delta_y\tau_2][-eA_0(-\vec{q})\tau_3 + ep_xA_x(-\vec{q})/mc + ep_yA_y(-\vec{q})/mc - \varphi(-\vec{q})p_x\Delta_x\tau_2 - \varphi(-\vec{q})p_y\Delta_y\tau_1][i(\omega - \Omega/2) + \varepsilon(\mathbf{p} - \mathbf{q}/2)\tau_3 + (p_x - q_x/2)\Delta_x\tau_1 - (p_y - q_y/2)\Delta_y\tau_2] \quad (10)$$

The calculation of the effective action (7) is conceptually similar to the calculation of electromagnetic response in the BCS theory of superconductivity [12,16]. However, we focus only on obtaining the Chern-Simons-like term responsible for σ_{xy} [6,14]. Picking the A_0 term from the first factor in Eq. (10) and the A_x or A_y term from the third factor, we obtain a nonzero contribution after taking trace over the τ matrices. The same procedure works for the A_x or A_y term from the first factor and the A_0 term from the third factor. Combining these terms and changing the variable of integration $\vec{q} \rightarrow -\vec{q}$ in the latter term, we obtain one contribution to C_1

$$C_1^{(a)} = A_0(\vec{q})[-q_yA_x(-\vec{q})p_x^2 + q_xA_y(-\vec{q})p_y^2]\frac{2i\Delta_x\Delta_ye^2}{mc}. \quad (11)$$

In deriving (11), we omitted the terms proportional to the product p_xp_y , which would vanish after integration over p_x and p_y . The integration over momentum \mathbf{p} in Eq. (8) is concentrated near the Fermi surface, so we can replace $p_x^2 \rightarrow p_F^2/2$ and $p_y^2 \rightarrow p_F^2/2$ in Eq. (11), because $p_x^2 + p_y^2 \approx p_F^2$. Making the Fourier transform of Eq. (11) to the coordinate space, we find

$$C_1^{(a)} = A_0(\partial_yA_x - \partial_xA_y)\frac{\Delta_0^2e^2}{mc}, \quad (12)$$

The Fourier transform of Eq. (5) can be written as

$$\Gamma = -eA_0\tau_3 + e\mathbf{A}\cdot\mathbf{p}/mc - \varphi p_x\Delta_x\tau_2 - \varphi p_y\Delta_y\tau_1. \quad (6)$$

Here the variables \vec{A} and φ are assumed to be functions of the Fourier variable $\vec{q} = (\Omega, q_x, q_y)$. Thus, the vertex $\Gamma(\vec{q}, \vec{p})$ (6) is a function of two vector arguments.

The effective action of the system to the second order in Γ is

$$S = \frac{1}{2}\sum_{\vec{q}, \vec{p}}\text{Tr}\Gamma(\vec{q}, \vec{p})G(\vec{p} + \vec{q}/2)\Gamma(-\vec{q}, \vec{p})G(\vec{p} - \vec{q}/2). \quad (7)$$

Substituting Eqs. (4) and (6) into Eq. (7), we write S in the form

$$S = \sum_{\vec{q}, \vec{p}}\frac{C_1}{C_2}, \quad (8)$$

where the denominator is

$$C_2 = [(\omega + \Omega/2)^2 + E^2(\mathbf{p} + \mathbf{q}/2)] \times [(\omega - \Omega/2)^2 + E^2(\mathbf{p} - \mathbf{q}/2)], \quad (9)$$

and the numerator is

where $\Delta_0 = \Delta_x p_F = \Delta_y p_F$ is the energy gap at the Fermi level.

In addition, picking the last two terms in the first factor in Eq. (10) and the A_x and A_y terms in the third factor or vice versa, we obtain another contribution to C_1 :

$$C_1^{(b)} = i\Omega\varphi(\vec{q})[-q_yA_x(-\vec{q})p_x^2 + q_xA_y(-\vec{q})p_y^2]\frac{\Delta_x\Delta_ye}{mc}. \quad (13)$$

Replacing the Matsubara frequency by the real frequency $i\Omega \rightarrow \Omega$ in Eq. (13) and Fourier-transforming to the coordinate space, we find

$$C_1^{(b)} = \partial_t\varphi(\partial_yA_x - \partial_xA_y)\frac{\Delta_0^2e}{2mc}. \quad (14)$$

Combining the contributions (12) and (14) to C_1 and substituting into (8), we find a Chern-Simons-like term in the effective action [6,14]

$$S_{\text{CS}} = \sigma_{xy} \int dt dx dy (A_0 + \partial_t\varphi/2e)(\partial_yA_x - \partial_xA_y)/c, \quad (15)$$

where

$$\sigma_{xy} = \frac{\Delta_0^2 e^2}{m} \sum_{\vec{p}} \frac{1}{C_2} \quad (16)$$

is the effective Hall conductivity. Indeed, taking the variational derivative of Eq. (15), we find electric current

$$\mathbf{j} = c \frac{\delta S_{CS}}{\delta \mathbf{A}} = \sigma_{xy} \left[\mathbf{E} - \frac{1}{2e} \partial_t (\nabla \varphi - \frac{2e}{c} \mathbf{A}) \right] \times \hat{z}, \quad (17)$$

where $\mathbf{E} = -\nabla A_0 - \partial_t \mathbf{A}/c$ is the electric field, and the last term in Eq. (17) is proportional to the time derivative of the London supercurrent $\mathbf{j}_s = (\rho_s e/2m)[\nabla \varphi - (2e/c)\mathbf{A}]$. Obtaining a self-consistent equation of motion for the superconducting phase φ is a complicated problem [17]. However, one may argue that the supercurrent contribution in Eq. (17) is ineffective at high frequencies, so the last term can be omitted, and we obtain the standard relation for the Hall conductivity $\mathbf{j} = \sigma_{xy} \mathbf{E} \times \hat{z}$. The Chern-Simons-like term (15) was derived in Ref. [6] at $T = 0$ and in Ref. [14] near T_c via the Ginzburg-Landau expansion. Notice that it does not have the component $A_x \partial_t A_y$, which is present in the standard Chern-Simons term $\epsilon^{\mu\nu\lambda} A_\mu F_{\nu\lambda}$. Nevertheless, Eq. (15) is gauge invariant, because the gauge transformation of A_0 in the first factor is compensated by transformation of the superconducting phase φ , and the last factor is manifestly gauge invariant [6].

Now we substitute Eq. (9) into Eq. (16) and derive an explicit expression for the Hall conductivity. To obtain optical response at a finite temperature, we do analytical continuation from the Matsubara to real frequencies, which is well known in the BCS theory [16]. We take the limit $\mathbf{q} \rightarrow 0$ while keeping finite frequency, as appropriate for optical response, and find

$$\sigma_{xy}(\Omega) = \frac{e^2 \Delta_0^2}{8\pi} \int_{-\infty}^{\infty} d\varepsilon \frac{1 - 2n(E/T)}{E^2} \times \left(-\frac{1}{\Omega + i\gamma - 2E} + \frac{1}{\Omega + i\gamma + 2E} \right), \quad (18)$$

where $E = \sqrt{\varepsilon^2 + \Delta_0^2}$, $n(E/T)$ is the Fermi distribution function, Ω is the real frequency, γ is a relaxation rate, and we replaced the integration over $dp_x dp_y / (2\pi)^2 m$ by the integration over $d\varepsilon / 2\pi$. First we take the dc limit $\Omega \rightarrow 0$ at zero temperature $T \rightarrow 0$ in Eq. (18) and find

$$\sigma_{xy}^{\text{dc}} = \frac{e^2}{4\pi} = \frac{e^2}{2h}, \quad (19)$$

where we restored the dimensional factor $\hbar = h/2\pi$ in the denominator. Equation (19) demonstrates the half-integer quantum Hall conductivity in agreement with Ref. [6]. As discussed in Ref. [14], it is difficult to measure the dc Hall conductivity experimentally because of screening by supercurrents.

At a finite temperature T , the dc Hall conductivity $\sigma_{xy}^{\text{dc}}(T) = (e^2/2h)f_d(T)$ is reduced by the factor

$$f_d(T) = \frac{\Delta_0^2}{2} \int_{-\infty}^{\infty} d\varepsilon \frac{1 - 2n(E/T)}{E^{3/2}}. \quad (20)$$

The factor $f_d(T)$ interpolates between 1 at $T = 0$ and 0 at $T = T_c$ and behaves as $f_d(T) \propto \Delta \propto \sqrt{T_c - T}$ near T_c . The same factor (20) describes temperature dependence of the quantum Hall effect in the magnetic-field-induced spin-density-wave (FISDW) state of the quasi-one-dimensional organic conductors (TMTSF)₂X [18,19]. Equation (20) represents the dynamic limit of the dc electromagnetic response [18,19]. If the limit $\Omega \rightarrow 0$ is taken in Eq. (16) first and then $\mathbf{q} \rightarrow 0$, that would generate the static limit $f_s(T)$ for the reduction function, which has the same temperature dependence as the London superfluid density $\rho_s(T)$, particularly $f_s(T) \propto \Delta^2 \propto (T_c - T)$ near T_c (see discussion in Sec. VI of Ref. [19]). Ref. [14] obtained the static limit for σ_{xy}^{dc} near T_c by doing the Ginzburg-Landau expansion.

Now we calculate the imaginary part of the Hall conductivity $\sigma_{xy}''(\Omega)$ at a high-frequency $\Omega \gg \Delta_0$. One contribution originates from the pole at $\Omega = 2E$ in Eq. (18). This term represents creation of an electron pair above the energy gap Δ_0 or a hole pair below the gap by absorption of a photon with the frequency Ω . By integrating over $\varepsilon \approx \pm E$ in the vicinity of the resonance, we find

$$\sigma_{xy}''(\Omega) = \frac{e^2 \Delta_0^2}{2\Omega^2} = \frac{e^2}{2\hbar} \left(\frac{\Delta_0}{\hbar\Omega} \right)^2. \quad (21)$$

Calculating this term, we set $E^2 = (\Omega/2)^2$ in the denominator of the first factor in Eq. (18) and put $n(E/T) = 0$, because $\Omega \gg T$. We observe that Eq. (21) does not depend on the relaxation rate γ and is reduced relative to Eq. (19) by the factor $(\Delta_0/\hbar\Omega)^2/2\pi$. The temperature dependence of $\sigma_{xy}''(\Omega)$ is given by $\Delta_0^2(T)$.

There is another contribution to the integral (18) originating from the peak in the density of states at $E \approx \Delta_0$. By changing the variable on integration from ε to E , we rewrite Eq. (18) as follows:

$$\sigma_{xy}(\Omega) = -\frac{e^2 \Delta_0^2}{\pi} \int_{\Delta_0}^{\infty} dE \frac{1 - 2n(E/T)}{\sqrt{E^2 - \Delta_0^2}} \times \frac{1}{(\Omega + i\gamma)^2 - 4E^2}. \quad (22)$$

Integral (22) over dE is logarithmic between Δ_0 and Ω . For simplicity, we consider low temperatures, where $n(E/T) \approx 0$, and find the following contribution $\tilde{\sigma}_{xy}$ to the Hall conductivity

$$\begin{aligned} \tilde{\sigma}_{xy}(\Omega) &\approx -\frac{e^2}{\pi} \frac{\Delta_0^2}{(\Omega + i\gamma)^2} \ln\left(\frac{\Omega}{\Delta_0}\right), \\ \tilde{\sigma}_{xy}''(\Omega) &\approx -\frac{4e^2}{h} \frac{\Delta_0^2 \hbar \gamma}{(\hbar\Omega)^3} \ln\left(\frac{\hbar\Omega}{\Delta_0}\right). \end{aligned} \quad (23)$$

Equation (23) is reduced relative to Eq. (21) by the factor γ/Ω and is enhanced by the factor $\ln(\hbar\Omega/\Delta_0)$. Using the

numbers from Ref. [1] and given below, we conclude that the reduction of Eq. (23) is much greater than the enhancement, so we focus only on Eq. (21).

Substituting Eq. (21) into Eq. (1), we find the Kerr angle

$$\theta_K = \frac{2\pi}{n(n^2 - 1)} \frac{e^2}{d} \frac{\Delta_0^2}{(\hbar\Omega)^3} = \frac{\alpha}{n(n^2 - 1)} \frac{\lambda}{d} \frac{\Delta_0^2}{(\hbar\Omega)^2}, \quad (24)$$

where $\alpha = e^2/\hbar c = 1/137$ is the fine structure constant, and λ is the wavelength of light. Using the interlayer distance $d = 1.3$ nm [7] and the values $n(n^2 - 1) = 3$ and $\lambda = 1550$ nm from Ref. [1], we find that the first two factors in Eq. (24) give 2.9. Using the BCS formula $\Delta_0 = 1.76k_B T_c = 0.23$ meV for $T_c = 1.5$ K [1] and $\hbar\Omega = \hbar c/\lambda = 0.8$ eV, we find that the last factor in Eq. (24) is $(\Delta_0/\hbar\Omega)^2 = 8 \times 10^{-8}$. The resultant Kerr angle (24) is $\theta_K = 230$ nanorad. This estimate is 3.6 times greater than the experimentally observed value of 65 nanorad [1]. The experimental Kerr angle may be reduced relative to the theoretical estimate for variety of reasons. For example, the effective value of Δ_0 at high energies may be lower than at the Fermi level. We conclude that the theoretical formula (24) reasonably agrees with the experiment by the order of magnitude.

A different theoretical formula with $\theta_K \propto \Delta_0$ was proposed phenomenologically in Ref. [1] motivated by the experimental temperature dependence of $\theta_K(T)$. On the other hand, our formula (24) gives $\theta_K \propto \Delta_0^2$. The error bars in the experiment [1] are quite big, so deciding between the linear or quadratic dependences of θ_K on Δ_0 may require more precise measurements. The appearance of Δ_0^2 in Eqs. (21) and (24) is quite natural, originating from the product $\Delta_x \Delta_y$, which changes sign when the chirality of the order parameter changes from $p_x + ip_y$ to $p_x - ip_y$, as observed experimentally [1]. This product can be also written as the vector $\Psi \times \Psi^*$ pointing along \hat{z} [14], which is consistent with Eq. (17). It would be very interesting to verify experimentally the Ω^{-3} frequency dependence of the Kerr angle predicted by Eq. (24).

Experiments indicate that the superconducting gap may have the so-called horizontal lines of nodes in Sr_2RuO_4 (see Ref. [11], and references therein). In this case, Δ_0 should be considered a function of the electron momentum p_z perpendicular to the layers: $\Delta_0 \rightarrow \Delta_0 \cos(p_z d)$ for triplet pairing [11] or $\Delta_0 \rightarrow \Delta_0 \sin(p_z d)$ for singlet pairing [10]. In both cases, averaging $\Delta_0^2(p_z)$ over p_z generates an additional factor 1/2 in Eq. (24) and no changes in Eq. (19). Thus, although experiment [1] directly proves the chiral character of the superconducting pairing in Sr_2RuO_4 , it does not discriminate between triplet pairing and the chiral singlet pairing $(p_x + ip_y) \sin(p_z d)$ proposed in Ref. [10].

In conclusion, we derived the Chern-Simons-like term in the effective action of the 2D chiral $p_x + ip_y$ superconductor, generalizing previous results [6,14] to finite fre-

quency and temperature. The resultant dc Hall conductivity has the half-quantum value $\sigma_{xy} = e^2/2h$ at $T = 0$ [6], but is reduced at a finite temperature by the factor (20). We derived Eq. (21) for the imaginary part of the optical Hall conductivity and Eq. (24) for the polar Kerr angle, which agrees by the order of magnitude with the recent experimental measurement in Sr_2RuO_4 by Xia *et al.* [1]. Equation (24) predicts that the Kerr angle is proportional to the square of the superconducting energy gap and is inversely proportional to the cube of frequency, which can be verified experimentally. The derivation may be also relevant for the finite-temperature Chern-Simons theories in high-energy physics (see Refs. [20,21]).

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