

Nonreciprocal Current in Noncentrosymmetric Rashba Superconductors

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We study theoretically the nonreciprocal charge transport in two-dimensional noncentrosymmetric superconductors with the Rashba spin-orbit interaction. The resistivity R depends on the current I linearly under the external magnetic field B , i.e., $R = R_0(1 + \gamma BI)$, which is called the magnetochiral anisotropy. It is found that the coefficient γ is gigantically enhanced by the superconducting fluctuation with the components of both spin singlet and triplet pairings, compared with that in the normal state. This finding offers a method to quantitatively estimate the ratio of the pairing interactions between the singlet and triplet channels including its sign.

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The broken inversion symmetry P is expected to result in directional nonreciprocal responses. For example, the propagation of light through matter can be different depending on the direction. However, Onsager's reciprocal theorem [1], which originates from the time-reversal symmetry T of the microscopic dynamics, puts the constraint on the linear response function $K_{\alpha\beta}$ describing the response of α to the input β as

$$K_{\alpha\beta}(q, \omega, B) = \varepsilon_\alpha \varepsilon_\beta K_{\beta\alpha}(-q, \omega, -B), \quad (1)$$

where B is the magnetic field representing the T breaking, $\varepsilon_\alpha (= \pm 1)$ (ε_β) is the even or odd nature of α (β) with respect to T , and q and ω are the wave vector and frequency, respectively, of the physical quantities α and β . An example is the case where α and β are the same component of the current and $K_{\alpha\beta}$ describes the diagonal element of the dielectric function. Therefore, only with the broken T symmetry, asymmetric propagation of light between q and $-q$, called directional dichroism, becomes possible [2].

On the other hand, the nonlinear nonreciprocal responses in the transport phenomena are characterized by the current-dependent resistivity R expressed as

$$R = R_0(1 + \gamma BI), \quad (2)$$

where I is the current, B is the magnetic field, and γ represents the nonreciprocity [3–8]. This means the nonlinear $I - V$ characteristics are asymmetric between the positive and negative sign of I , i.e., the directional resistivity, called magnetochiral anisotropy (MCA) [3]. When the T symmetry is intact, the electronic dispersion $\varepsilon_\sigma(k)$ (k , crystal momentum; σ , spin) has the symmetry between k and $-k$, i.e., $\varepsilon_\sigma(k) = \varepsilon_{-\sigma}(-k)$. Therefore, the magnetic field B , which breaks T symmetry, is necessary in

addition to P breaking to induce the asymmetric energy dispersion and hence $I - V$ characteristics as shown in Eq. (2). Microscopically, both the spin-orbit interaction λ and the Zeeman effect $\mu_B B$ are needed to make the energy dispersion asymmetric, which are usually small perturbations compared with the kinetic energy of electrons, i.e., the Fermi energy E_F . Therefore, the strength γ of MCA in Eq. (2) is usually very tiny, because its expression contains the small factors of λ/E_F and $\mu_B B/E_F$.

The superconductivity in noncentrosymmetric systems changes this situation. The superconductivity changes the transport phenomena within the narrow low-energy window below the superconducting gap Δ or at low temperatures around and below the mean field transition temperature $T_c \cong \Delta/k_B$. The conductivity due to the fluctuating superconducting order parameter, i.e., paraconductivity, above T_c shows enhanced nonreciprocal transport as shown experimentally in MoS₂ [9]. It has been analyzed theoretically in terms of the time-dependent Ginzburg-Landau (GL) theory, and the enhancement of γ compared with that in the normal state γ_N is estimated as $\gamma/\gamma_N \sim (E_F/\Delta)^3$ [9]. This means that MCA provides useful information about the electronic states and superconductivity of noncentrosymmetric materials. In the case of MoS₂ with the out-of-plane magnetic field, the trigonal warping of the band structure leads to the third-order terms in the wave number of the order parameter, which is identified as the main mechanism of MCA.

In this Letter, we study the MCA of two-dimensional superconductors with the Rashba spin-orbit interaction in the temperature regime slightly above T_c , where the current is mainly carried by a thermal fluctuation of the superconducting order parameter [10,11]. The most essential aspect of the noncentrosymmetric superconductivity is the mixing of the spin-singlet even parity and spin-triplet odd parity pairings [12–16]. One of the consequences of this is

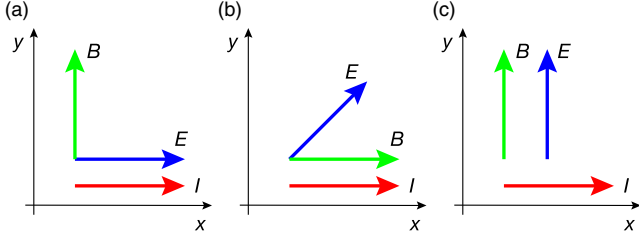


FIG. 1. The three field configurations which correspond to (a) σ_{xyxx} , (b) σ_{xxxy} , and (c) σ_{xyyy} . B , E , and I in the figures represent the electric field, magnetic field, and nonreciprocal current, respectively.

the very large upper critical magnetic field B_{c2} beyond the Pauli limit. As we will show below, the noncentrosymmetric superconductors with the Rashba spin-orbit interaction as given by Eq. (3) show nonreciprocal charge transport, which is very sensitive to the pairing interactions for singlet and triplet channels as shown in Eq. (19). Namely, once the Fermi energy E_F , the strength of the Rashba interaction, and T_c are known, one can estimate the ratio of the pairing interactions between the singlet and triplet channels, i.e., r_s or r_t , including its sign. Note also here that the third-order terms in the wave number of the order parameter, which was the main origin of γ for MoS₂, are estimated to be much smaller in the present case as indicated by Eq. (22) below. We also show that the nonreciprocal current has a unique electric and magnetic field angle dependence due to the symmetry constraints for the higher rank response tensor as shown in Figs. 1 and 2.

Before defining the Hamiltonian, we discuss the general form of the spin-orbit interaction in time-reversal preserved systems. If we express the spin-orbit interaction as $\mathbf{g}_k \cdot \boldsymbol{\sigma}$, time-reversal symmetry requires $\mathbf{g}_k = -\mathbf{g}_{-k}$. In this Letter, we assume the simplest Rashba spin-orbit interaction, although the other forms will give the qualitatively same conclusions.

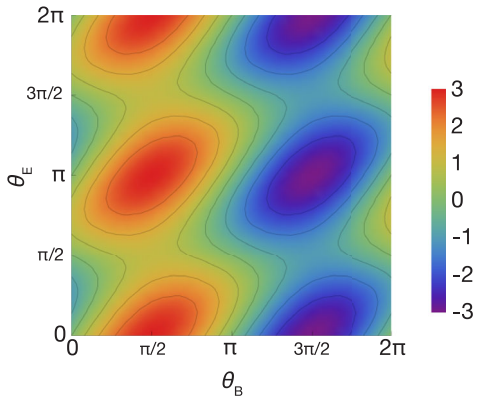


FIG. 2. The electric and magnetic field angle dependence of $j^{(2)}$. θ_B (θ_E) represents the angle between the magnetic (electric) field and the nonreciprocal current. The amplitude is normalized by $\sigma_{xyyy}BE^2$.

We start with the Rashba Hamiltonian, which is given by [8]

$$H_k = \xi_k + \alpha(k_x\sigma_y - k_y\sigma_x) - \mu_B \mathbf{B} \cdot \boldsymbol{\sigma}, \quad (3)$$

where $\xi_k = [(\hbar^2 k^2)/2m] - E_F$ is the dispersion without the spin-orbit interaction, α is the Rashba parameter, \mathbf{B} is the in-plane magnetic field, and $\boldsymbol{\sigma}$ is the Pauli matrix. We have assumed that the g factor is 2. Its eigenenergies are

$$\xi_{\pm k} = \xi_k \pm \sqrt{(\alpha k_y + B_x)^2 + (\alpha k_x - B_y)^2}. \quad (4)$$

Now we consider the superconductivity in the presence of the Rashba interaction [12,16]. For an even parity attractive interaction, we assume the standard BCS-type on-site attractive interaction:

$$H_{\text{int}} = -V^g \sum_{kk'} c_{k'\uparrow}^\dagger c_{-k\downarrow}^\dagger c_{-k'\downarrow} c_{k'\uparrow}, \quad (5)$$

with $c_{k\sigma}^\dagger$ and $c_{k\sigma}$ being the creation and annihilation operators, respectively, of the electron with momentum \mathbf{k} and spin σ . In general, the odd parity part is

$$-\sum_{kk'} V_{ij}^u(\mathbf{k}, \mathbf{k}') (i\sigma_i \sigma_2)_{\alpha\beta} (i\sigma_j \sigma_2)_{\gamma\delta} c_{k\alpha}^\dagger c_{-k\beta}^\dagger c_{-k'\gamma} c_{k'\delta}, \quad (6)$$

with $V_{ij}^u(\mathbf{k}, \mathbf{k}')$ being an odd function with respect to \mathbf{k} and \mathbf{k}' and invariant under the crystal symmetry transformations. For simplicity, we assume the simplest case $V_{ij}^u(\mathbf{k}, \mathbf{k}') = V^u \hat{\gamma}_i(\mathbf{k}) \hat{\gamma}_j(\mathbf{k}')$ with $\hat{\gamma}(\mathbf{k}) = \frac{1}{k}(-k_y, k_x)$ in the Rashba system. We assume that the Rashba splitting is much larger than the critical temperature ($E_R \gg T_c$) and the interband pairings can be neglected. Then, the interaction Hamiltonian in the band basis reads

$$H_{\text{int}} = -\sum_{kk'\lambda\lambda'} t_{k\lambda} t_{k'\lambda'}^* \hat{g}_{\lambda\lambda'} \Psi_{k\lambda}^\dagger \Psi_{-k\lambda}^\dagger \Psi_{-k'\lambda'} \Psi_{k'\lambda'}, \quad (7)$$

where $\Psi_{k\lambda}^\dagger$ and $\Psi_{k\lambda}$ are the creation and annihilation operators, respectively, with the band index $\lambda = \pm$ and $t_{k\lambda} = \lambda i e^{i\phi_k}$ with $\phi_k = \arg \mathbf{k}$. The \mathbf{k} -independent matrix \hat{g} is

$$\hat{g} = \begin{pmatrix} g_1 & g_2 \\ g_2 & g_1 \end{pmatrix}, \quad (8)$$

with $g_1 = (V^g + V^u)/4$ (> 0) and $g_2 = (V^g - V^u)/4$. In this Letter, we focus on two limiting cases. (i) $|V^u| \ll |V^g|$ case.—We calculate the first-order terms in the small parameter $r_t = [(2V^u)/(V^g + V^u)]$, which is proportional to the triplet channel interaction V^u . (ii) $|V^u| \gg |V^g|$ case.—We are interested in the first-order terms in the small parameter $r_s = [(2V^g)/(V^g + V^u)]$, which is proportional to the singlet channel interaction. In most of the cases, we expect that the singlet interaction V^g is larger than

the triplet interaction V^u , and hence we briefly mention the amplitude of r_t . If we assume that V^g and V^u correspond to the on-site and nearest-neighbor interactions, respectively, their amplitudes can be roughly estimated as e^2/a_0 and e^2/a with a_0 being the Bohr radius and a being the lattice constant. Therefore, $r_t \sim 0.1$ is a reasonable value. Although we cannot apply the same argument for r_s , we consider the other limit $r_s \ll 1$ to see the global behavior of γ .

In order to calculate the superconducting fluctuation current slightly above the mean field critical temperature, it is convenient to employ the GL theory. The free energy quadratic with respect to the order parameters can be obtained by the equation [12]

$$F = \int \frac{d^2\mathbf{q}}{(2\pi)^2} \left(\sum_{\lambda\lambda'} \Psi_{\lambda\mathbf{q}}^* (\hat{g}^{-1})_{\lambda\lambda'} \Psi_{\lambda'\mathbf{q}} - \sum_{\lambda} T \sum_{\omega_n} \int \frac{d^2\mathbf{k}}{(2\pi)^2} G_{\lambda}(\mathbf{k}, i\omega_n) \times G_{\lambda}(-\mathbf{k} + \mathbf{q}, -i\omega_n) |\Psi_{\lambda\mathbf{q}}|^2 \right), \quad (9)$$

where $\Psi_{\lambda\mathbf{q}}$ is the order parameter and $G_{\lambda}(\mathbf{k}, i\omega_n) = (i\omega_n - \xi_{\lambda\mathbf{k}})^{-1}$ is the noninteracting normal Green's function. We set the Boltzmann constant $k_B = 1$.

First, we assume $E_F > 0$, and we will soon show that the nonreciprocal current vanishes for $E_F < 0$ in Eq. (18) below. After some calculations (see Supplemental Material [17]), we obtain

$$F = \int \frac{d^2\mathbf{k}}{(2\pi)^2} \sum_{\lambda\lambda'} \Psi_{\lambda}^* [(\hat{g}^{-1})_{\lambda\lambda'} + \delta_{\lambda\lambda'} N_{\lambda} (S_1 - L_{\lambda\mathbf{k}})] \Psi_{\lambda'}, \quad (10)$$

$$L_{\lambda\mathbf{k}} = K_{\lambda} \mathbf{k}^2 - \lambda R_{\lambda} (B_y k_x - B_x k_y), \quad (11)$$

$$S_1 = \log \frac{2e^{\gamma_E} E_c}{\pi T}, \quad (12)$$

with $\delta_{\lambda\lambda'}$, γ_E , and E_c being the Kronecker delta, Euler constant, and cutoff energy, respectively. The density of states N_{λ} and the other coefficients K_{λ} and R_{λ} are given in Supplemental Material [17]. The critical temperatures are obtained by solving

$$\det[\hat{g}^{-1} - \hat{N}S_1(T_c)] = 0, \quad (13)$$

with $\hat{N}_{\lambda\lambda'} = \delta_{\lambda\lambda'} N_{\lambda}$. It results in

$$\frac{1}{S_1(T_c)} = \frac{g_1(N_- + N_+)}{2} \pm \sqrt{\left(\frac{g_1(N_- - N_+)}{2}\right)^2 + g_2^2 N_- N_+}. \quad (14)$$

Because of the form of the interaction ($g_1 \approx g_2$ for the singlet dominant case and $g_1 \approx -g_2$ for the triplet dominant case), the solution with the plus sign has a much higher critical temperature. Hence, we can ignore the order parameter with the lower critical temperature when we calculate the fluctuation current.

The fluctuation current can be obtained by evaluating the equation [9,18]

$$\mathbf{j} = -T \sum_{\mathbf{k}} C \frac{\partial \eta(\mathbf{k} + 2e\mathbf{A})}{\partial \mathbf{A}} \Big|_{\mathbf{A}=0} \times \int_{-\infty}^0 du \exp \left[-C \int_u^0 dt \eta(\mathbf{k} - 2e\mathbf{E}t) \right], \quad (15)$$

where η is the eigenvalue of the matrix in Eq. (10) with the higher critical temperature and $C = \{(32T_c)/[\pi\hbar(N_- + N_+)]\} + O(r_{t,s})$. We expand the eigenvalue up to $O(r_{t,s})$, because we can show that the coefficient of the second-order term is half of the coefficient of the linear term; hence, the higher-order terms can be neglected. It is noted that the factor C should contain an $r_{t,s}$ -dependent correction from the relaxation time of order parameters in the time-dependent GL theory. However, we ignore it, because it does not affect the γ value in the lowest order of $r_{t,s}$. As in the case of the normal state, we assume that the electric and magnetic fields are applied along the x and y directions, respectively, and evaluate the current along the x direction up to $O(B_y E_x^2)$. We will discuss the case of general field configurations later. After the integration in Eq. (15) is carried out (we employed *Mathematica*), the relation Eq. (14) is used to simplify the equation. The result is

$$j_x = \sigma^{(1)} E_x + \sigma^{(2)} E_x^2, \quad (16)$$

$$\sigma^{(1)} = \frac{e^2}{16\hbar\varepsilon}, \quad (17)$$

$$\sigma^{(2)} = \frac{\pi e^3 B_y r_{t,s}}{128\hbar\varepsilon^2} \times \frac{N_- N_+ (K_- N_- - K_+ N_+) (K_- R_+ + K_+ R_-)}{S_1(T_c) T_c (N_- + N_+) (K_- N_- + K_+ N_+)^2}, \quad (18)$$

in the lowest order of $r_{t,s}$. Here, we have defined the reduced temperature $\varepsilon = [(T - T_c)/T_c]$. The linear coefficient $\sigma^{(1)}$ is the conventional form of the fluctuation conductivity in two-dimensional superconductors. The nonlinear coefficient $\sigma^{(2)}$ grows faster than $\sigma^{(1)}$ toward the critical temperature as in the case of MoS₂ [9]. It is noted that the parity mixing is essential for the nonreciprocal current, which vanishes for $r_{t,s} = 0$.

We mention the case when the Fermi energy is below the crossing point of the bands ($E_F < 0$). In this case, because the density of states from the upper band is zero, the

nonreciprocal current vanishes, whereas the normal current contribution exists [8].

For $E_F > 0$, the γ value expressed with the microscopic parameters is

$$W\gamma_S = \frac{\sigma^{(2)}}{B_y(\sigma^{(1)})^2} = \frac{\pi\mu_B\hbar S_3 E_F \alpha r_{l,s}}{eS_1 T_c (2E_F + E_R)}, \quad (19)$$

with W being the sample width and $E_R = [(m\alpha^2)/\hbar^2]$ being the energy splitting at the shifted momentum due to the Rashba spin-orbit interaction, and $S_3 = \{[7\zeta(3)]/(4\pi^2 T_c^2)\}$. We have used the relation between $\sigma^{(1)}$, $\sigma^{(2)}$, and γ as shown in Ref. [9]. More precisely, $\sigma^{(1)}$ and $\sigma^{(2)}$ in Eq. (19) should contain normal state contributions. Therefore, the γ value approaches the value in Eq. (19) when the fluctuation contribution exceeds the normal contribution. Explicitly, according to the Drude formula for the normal conductivity, the γ value develops below $\varepsilon^* = [m/(16\hbar n\tau)]$, with n being the electron density and τ being the relaxation time.

It should also be noted that the sign of Eq. (19) depends on the sign of $r_{l,s}$, i.e., the sign of V^u for the singlet dominant case and V^g for the triplet dominant case. Therefore, we can determine whether the interaction is repulsive or attractive by MCA measurement.

Here, we consider the nonreciprocal current which relies on the third-order term with respect to the wave number as transition metal dichalcogenides [9]. Although the Rashba Hamiltonian possesses rotational symmetry, the cubic term of the wave number of the order parameter appears in the GL free energy in the presence of the in-plane magnetic field. The detailed calculation of the γ value is shown in Supplemental Material [17], and the result is

$$W\tilde{\gamma}_S^{\text{pos}} = \frac{3\pi\mu_B\hbar\alpha}{2eT_c(2E_F + E_R)^2} \quad (E_F > 0), \quad (20)$$

$$W\tilde{\gamma}_S^{\text{neg}} = \frac{15\pi\mu_B\hbar\alpha}{4eT_c E_R (2E_F + E_R)} \quad (E_F < 0). \quad (21)$$

The ratio between the γ values from the parity mixing and the cubic term mechanisms is

$$\frac{\gamma_S}{\tilde{\gamma}_S^{\text{pos}}} \sim \frac{r_{l,s} E_F (2E_F + E_R)}{S_1 T_c^2}. \quad (22)$$

Therefore, the MCA from the cubic term is negligible compared to that from the parity mixing.

Next, we compare the γ values in the normal state and the superconducting fluctuating regime [Eq. (19)]. In Ref. [8], it has been concluded that the MCA exists if the Fermi energy is below the crossing point of the bands ($E_F < 0$). The amplitude of the MCA is

$$W\gamma_N = \frac{3\pi\mu_B\hbar\alpha}{2e[E_R(E_R - 2|E_F|)]^{3/2}}. \quad (23)$$

We assume that the strength of the spin-orbit interaction is comparable with the Fermi energy ($E_R \approx |E_F|$), because it is difficult to realize $E_F < 0$ with a small E_R . Then, we obtain

$$W\gamma_N \sim \frac{\mu_B\hbar^2}{e\sqrt{m}} \frac{1}{|E_F|^{5/2}}. \quad (24)$$

In the superconducting fluctuation regime, the nonreciprocal fluctuation current exists in the case of $E_F > 0$, which is opposite to the normal state. With the same assumption for the normal state, we obtain

$$W\gamma_S \sim \frac{\mu_B\hbar^2}{e\sqrt{m}} \frac{r_{l,s} E_F^{1/2}}{S_1 T_c^3}. \quad (25)$$

From Eqs. (24) and (25), we conclude that the MCA is drastically enhanced in the superconducting fluctuation regime because of the huge energy scale difference between the Fermi energy E_F and the critical temperature T_c . This is similar to the proceeding results for MoS₂ [9].

We finally mention the electric and magnetic field angle dependence of the nonreciprocal current. If we express the second-order current as $j_i = \sigma_{ijkl} B_j E_k E_l$, the coefficient σ_{ijkl} is the pseudotensor consistent with the crystal symmetry. Our model Eq. (3) possesses C_∞ symmetry and arbitrary in-plane mirror symmetries, which impose the restrictions that, among σ_{ijkl} , only σ_{xxxy} ($= \sigma_{xxyx}$), σ_{xyxx} , and σ_{xyyy} can be finite (corresponding configurations are shown in Fig. 1), and $\sigma_{xyyy} = 2\sigma_{xxxy} + \sigma_{xxyx}$ and $\sigma_{yijk} = -\sigma_{xjkl}$ are satisfied. According to calculations the same as that for σ_{xxyx} above, we obtain $\sigma_{xxxy} = -\frac{1}{3}\sigma_{xxyx}$ and $\sigma_{xyyy} = \frac{1}{3}\sigma_{xxyx}$, which satisfy the above conditions. If we define the angle between the current and magnetic (electric) field as θ_B (θ_E), the nonreciprocal current is

$$j^{(2)} = \sigma_{xyyy}[2\sin\theta_B + \sin(\theta_B - 2\theta_E)]BE^2, \quad (26)$$

whose dependence in the (θ_B, θ_E) plane is shown in Fig. 2. It is noted that the normal state has the same angle dependence, although it has not been discussed in the previous paper [8]. Realistic materials do not have such high symmetries; however, the above discussion should be applicable if the Fermi surface is almost circular.

We have investigated the MCA of the Rashba system in the superconducting fluctuation regime. The main result is the explicit form of the γ value shown in Eq. (19). Here, we estimate the γ value for the LaAlO₃/SrTiO₃ interface, where two-dimensional Rashba superconductivity is realized [19–22]. Its superconducting critical temperature is $T_c \sim 100$ mK. We mentioned that the γ value approaches the value in Eq. (19) when the fluctuation current exceeds the normal state contribution. The normal state sheet resistance is

$R_N \sim 1\text{k}\Omega$ [19–22], and the linear part of the fluctuation conductivity is shown in Eq. (17). By comparing them, we obtain $T - T_c \sim 1.5$ mK, below which Eq. (19) is applicable. The carrier density is $n \sim 10^{13}$ cm $^{-2}$, the spin-orbit field is $B_{\text{SO}} \sim 1$ T, and the Debye temperature is $T_D \sim 400$ K. If we assume $r_t = 0.1$, we obtain $W\gamma_S \sim 8 \times 10^{-2}$ T $^{-1}$ A $^{-1}$ m. With the typical sample width $W = 10^{-6}$, we obtain $\gamma_S \sim 8 \times 10^4$ T $^{-1}$ A $^{-1}$, which is a very large value compared with the previously known systems [2–6].

Such a huge enhancement of the MCA originates from the energy scale difference between the Fermi energy E_F and the critical temperature T_c as indicated in Eqs. (24) and (25). This phenomenon is similar to the case of superconducting MoS $_2$ [9], in which the large MCA stems from the trigonal warping term due to its threefold rotational symmetry. However, the MCA originates from the parity mixing of the order parameter in the present case.

We have also shown the unique field angle dependence of the nonreciprocal current, which is summarized in Fig. 1. It originates from the symmetry constraints of the higher rank response tensor. Especially, if the Fermi surface is almost circular and well approximated by our model, the field angle dependence is given in Eq. (26) and shown in Fig. 2.

In addition to LaAlO $_3$ /SrTiO $_3$ as we have discussed, normal Rashba systems can be used with the aid of the superconducting proximity effect. BiTeX ($X = \text{I, Br, Cl}$) [23,24], the surface of Au(111) [25], or Bi/Ag(111) alloy [26] will work well.

Experimentally, the nonreciprocal current can be observed simply by measuring the second-order harmonic voltage drop under a fixed ac current. With such a simple method, we can observe the nontrivial second-order response which reflects the crystal symmetry or the Hall response of the nonlinear current shown in Fig. 1(c). It is also possible to determine the sign of α from the sign of the γ value. Moreover, we may estimate the amplitude of $r_{t,s}$, which is the ratio between the even and odd parity attractive interactions, by using the measured γ_S value.

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