

Hidden anomalous Hall effect in Sr_2RuO_4 with chiral superconductivity dominated by the Ru d_{xy} orbital

Jia-Long Zhang ^{1,2}, Yu Li,¹ Wen Huang ^{2,*} and Fu-Chun Zhang^{1,3,†}

¹Kavli Institute for Theoretical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China

²Shenzhen Institute for Quantum Science and Engineering & Guangdong Provincial Key Laboratory of Quantum Science and Engineering, Southern University of Science and Technology, Shenzhen 518055, Guangdong, China

³Center for Excellence for Topological Quantum Computation, Chinese Academy of Sciences, Beijing 100190, China



(Received 19 September 2020; revised 25 October 2020; accepted 27 October 2020; published 24 November 2020)

The polar Kerr effect in superconducting Sr_2RuO_4 implies finite ac anomalous Hall conductivity. Since intrinsic anomalous Hall effect (AHE) is not expected for a chiral superconducting pairing developed on the single Ru d_{xy} orbital, multiorbital chiral pairing actively involving the Ru d_{xz} and d_{yz} orbitals has been proposed as a potential mechanism. Here we propose that AHE could still arise even if the chiral superconductivity is predominantly driven by the d_{xy} orbital. This is demonstrated through two separate models which take into account subdominant orbitals in the Cooper pairing, one involving the oxygen p_x and p_y orbitals in the RuO_2 plane, and another the d_{xz} and d_{yz} orbitals. In both models, finite orbital mixing between the dominant d_{xy} and the other orbitals may induce interorbital pairing between them, and the resultant states support intrinsic AHE, with Kerr rotation angles that could potentially reconcile with the experimental observation. Our proposal therefore sheds new light on the microscopic pairing in Sr_2RuO_4 . We also show that intrinsic Hall effect is generally absent for nonchiral states such as $S + iD$, $D + iP$, and $D + iG$, which provides a clear constraint on the symmetry of the superconducting order in this material.

DOI: [10.1103/PhysRevB.102.180509](https://doi.org/10.1103/PhysRevB.102.180509)

Introduction. The nature of the unconventional superconducting pairing in Sr_2RuO_4 is an outstanding open question in condensed matter physics. Despite tremendous efforts on various fronts, it remains difficult and controversial to interpret all of the key experimental observations in a consistent theory [1–9]. A number of measurements point to time-reversal symmetry breaking (TRSB) pairing—indicative of the condensation of multiple superconducting order parameters [10–12], most likely in the two-dimensional irreducible representations (irrep) of the underlying crystalline D_{4h} group [3]. This makes chiral pairings, including chiral p wave ($P_x + iP_y$) and chiral d wave ($D_{xz} + iD_{yz}$), promising candidates, although mixed-representation states, such as the chiral d wave with $D_{x^2-y^2} + iD_{xy}$, and the nonchiral states the likes of $S + iD$ and $D + iP$, etc., cannot be definitively ruled out. The chiral pairings support chiral edge modes, and the p -wave state may further support Majorana zero modes that could be utilized for topological quantum computing [13,14].

One important evidence for TRSB pairing in this material is the Kerr rotation, i.e., a circularly polarized light normally incident on a superconducting sample is reflected with a rotated polarization [11]. To date, the origin of the Kerr effect, or that of the closely related ac anomalous Hall effect (AHE) in superconducting Sr_2RuO_4 , remains controversial. It has often been inquired alongside the question about the primary superconducting orbital(s) in this material. To begin

with, a single-orbital chiral pairing, as would be the case if superconductivity is solely associated with the quasi-two-dimensional (2D) Ru d_{xy} orbital, is not expected to generate anomalous Hall response as a consequence of Galilean invariance [15–18], except in the presence of impurities [19–22]. However, such an extrinsic mechanism may not be sufficient to explain the Kerr rotation in measurements on high-quality crystals [4,19,20]. Recent discussions about possible active pairing on the two quasi-one-dimensional (1D) Ru d_{xz} and d_{yz} orbitals have stimulated an alternative interpretation, that intrinsic AHE is not forbidden in such a multiorbital chiral superconductor [23–25].

The Kerr effect bears special significance for understanding the microscopic Cooper pairing in Sr_2RuO_4 , including its driving superconducting orbital(s) and the symmetry of its superconducting order parameter. Our Rapid Communication contributes new insights into both of these highly contentious issues.

On the one hand, we disclose a “hidden” AHE in pristine Sr_2RuO_4 even when its chiral pairing is dominated by the single Ru d_{xy} orbital, i.e., the quasi-2D γ band. We are motivated by the observation of substantial mixing between the d_{xy} orbital and the oxygen p_x and p_y orbitals in the RuO_2 plane. Although they locate relatively far from the Fermi energy in the atomic limit and are thus customarily ignored in most theoretical studies, the oxygen orbitals in fact contribute significantly to the γ -band density of states [26,27]. We will show that, despite having only one band crossing the Fermi energy, such system with a hidden multiorbital character exhibits intrinsic Hall response and will hence

*huangw3@sustech.edu.cn

†fuchun@ucas.ac.cn

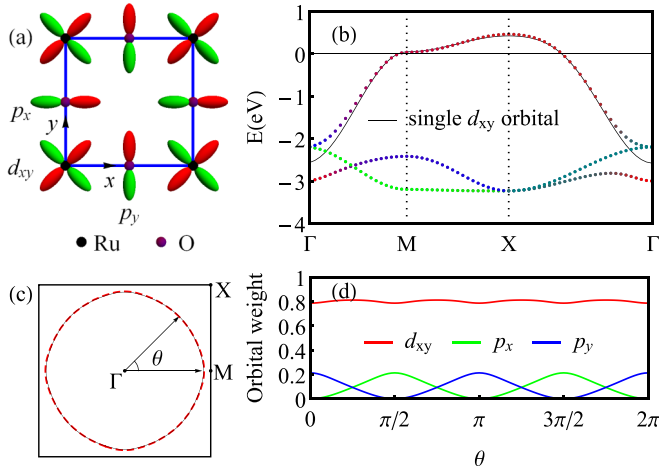


FIG. 1. Lattice and electronic structure of the dpp model. (a) Sketch of the dpp model with Ru- d_{xy} and O- p_x and p_y orbitals on the square lattice of the RuO₂ plane. (b) Band structure of the dpp model (dotted lines) and the single-orbital model with d_{xy} orbital (solid line). We choose a set of tight-binding parameters to match with the first-principle calculations [26]. The color gradient in dotted lines encrypts the variation of the weights of the three orbitals in the electronic states. The color codes are shown in (d). (c) Fermi surfaces of the dpp (red dashed) and the single- d_{xy} -orbital model (black solid). (d) Angular dependence of the individual orbital weights across the Fermi surface.

generate Kerr rotation. The effect is found to rely crucially on the induced interorbital pairing between the Ru- d and O- p orbitals. The conclusion applies to all chiral superconducting states, including chiral p wave, as well as chiral d wave with $\mathcal{D}_{xz} + i\mathcal{D}_{yz}$ and $\mathcal{D}_{x^2-y^2} + i\mathcal{D}_{xy}$ pairings. In a simple generalization, a qualitatively similar Hall response is obtained in another model containing the three t_{2g} orbitals, where finite orbital mixing between the dominant d_{xy} and the subdominant d_{xz}/d_{yz} orbitals may similarly induce interorbital pairings.

On the other hand, our study also places strong constraints on the possible superconducting pairing symmetry in this material. In particular, we show that nonchiral TRSB states such as $\mathcal{S} + i\mathcal{D}$, $\mathcal{D} + i\mathcal{P}$, and $\mathcal{D} + i\mathcal{G}$ generally do not support intrinsic Hall response, irrespective of the microscopic model details.

Chiral superconductivity in dpp model. We start by constructing a tight-binding model consisting of the Ru d_{xy} and O p_x and p_y orbitals in the RuO₂ plane (Fig. 1), which we name the dpp model. The normal-state Hamiltonian is given by $H_0 = \sum_{\vec{k}\sigma} \psi_{\vec{k}\sigma}^\dagger \hat{H}_{0\vec{k}} \psi_{\vec{k}\sigma}$, where $\sigma = \uparrow, \downarrow$ is the spin index, $\psi_{\vec{k}\sigma} = (c_{d\vec{k}\sigma}, c_{p_x\vec{k}\sigma}, c_{p_y\vec{k}\sigma})^\top$ represents the fermionic spinor, and

$$\hat{H}_{0\vec{k}} = \begin{pmatrix} \epsilon_{d\vec{k}} & it_{dp} \sin \frac{k_y}{2} & it_{dp} \sin \frac{k_x}{2} \\ -it_{dp} \sin \frac{k_y}{2} & \epsilon_{p_x\vec{k}} & t_{pp} \sin \frac{k_x}{2} \sin \frac{k_y}{2} \\ -it_{dp} \sin \frac{k_x}{2} & t_{pp} \sin \frac{k_x}{2} \sin \frac{k_y}{2} & \epsilon_{p_y\vec{k}} \end{pmatrix}. \quad (1)$$

Here $\epsilon_{d\vec{k}} = -2t_d(\cos k_x + \cos k_y) - 4t'_d \cos k_x \cos k_y - \mu_d$, $\epsilon_{p_{x(y)}\vec{k}} = -2t_{p\parallel(\perp)} \cos k_x - 2t_{p\perp(\parallel)} \cos k_y - \mu_p$. Here t_d and t'_d stand, respectively, for the first and second neighbor hoppings

between d_{xy} orbitals, and $t_{\parallel(\perp)}$ denotes the first neighbor hopping of the p orbitals parallel (perpendicular) to the orbital's lobe direction. It is worth noting that, due to spatial proximity, the d - p mixing t_{dp} is among the largest hopping integrals in the model.

Throughout this section, we employ the following set of parameters $(t_d, t'_d, t_{p\parallel}, t_{p\perp}, t_{dp}, t_{pp}, \mu_d, \mu_p) = (0.35, 0.14, -0.25, 0.074, 1, 0.33, 1.04, 2.55)$ in units of eV, which leads to the band structure and Fermi surface as shown in Figs. 1(b) and 1(c). This band structure, with a band inversion between the d and p orbitals at the Γ point, shows good agreement with the one obtained from first-principle calculations [26]. Due to this very band inversion, the resultant γ band defies an effective single-Wannier-orbital construction. Crucial to our argument, the p orbitals are found to feature prominently at the Fermi energy [Fig. 1(d)], in total representing roughly 20% of the electronic density of states. An early band structure calculation also found similar-size contribution from the p orbitals [27]. This observation would otherwise raise an interesting question about the role played by the oxygen orbitals in the microscopic theories of the superconductivity in Sr₂RuO₄.

In the following, we illustrate the construction of the gap functions of the dpp model with the example of the spin-triplet chiral p -wave pairing. A similar construction for several other pairing states is presented in the Supplemental Material [28]. The chiral p -wave order parameter belongs to the E_u irrep of the D_{4h} point group, and as per our assumption, the pairing is dominated by the d_{xy} orbital, with $\Delta_{d\vec{k}} = \Delta_1(\sin k_x + i \sin k_y)$. Although the p orbitals are distant from the Fermi energy in the unhybridized limit and thus likely do not exhibit intrinsic Cooper instability [29], the strong d - p mixing may still induce some interorbital pairing under appropriate circumstances. Note that, for simplicity, throughout the Rapid Communication the gap functions are presumed to have the forms of the simplest lattice harmonics. No qualitative feature is lost due to this simplification. Furthermore, since the model in Eq. (1) ignores spin-orbit coupling (SOC), components of the E_u irrep with $\sin k_z$ -like pairings [30] are ignored.

Before turning to the forms of the interorbital pairing, an important remark about the gap classification is in order. While the usual intraorbital pairing is fully classified according to the spin exchange statistics and the spatial parity of the Cooper pair wave function, interorbital pairings are also characterized by the parity number under orbital exchange (orbital singlet vs orbital-triplet). Furthermore, in analyzing the symmetry of the pairing, the spatial parity of the individual electron orbitals constituting the Cooper pair also matters. This is because symmetry operations act on both the Cooper pair wave function and the orbital wave functions of the two constituent electrons [31–33]. This additional degree of freedom adds a layer of complexity when pairing takes place between electron orbitals of opposite parities, such as the d and $p_{x(y)}$ orbitals in the present study. In short, the superconducting gap functions could acquire forms that differ considerably from the lattice generalizations of $k_x + ik_y$. The same holds true for all other irreps [31–33]. As a consequence, each irrep may permit multiple coexisting gap functions coupled by orbital mixing [31].

Since the orbitals d_{xy} and (p_x, p_y) belong respectively to the B_{2g} and E_u irreps, the pair creation (or annihilation) operators $c_d^\dagger c_p^\dagger$ and $c_d^\dagger c_{p_x}^\dagger$ jointly transform according to the E_u irrep. Denoting the interorbital pairing function between the d_{xy} and $p_{x(y)}$ orbitals $\Delta_{dp_{x(y)\vec{k}}}$, their lowest order basis functions are then $1 + ak_x^2 + bk_y^2$ and $1 + bk_x^2 + ak_y^2$, where a and b are real constants. Accounting for the lattice structure in Fig. 1(a), one may take $\Delta_{dp_{x(y)\vec{k}}} = \Delta_2 \cos \frac{k_{y(x)}}{2}$. In contrast to the intraorbital (spatial) odd-parity gap function $\Delta_{d\vec{k}} = \Delta_1 (\sin k_x + i \sin k_y)$, the interorbital odd-parity pairing features even-parity gap functions, while its oddness is encoded instead in the electron orbital manifold, i.e., the product of d and p orbital wave functions being odd under inversion.

The full pairing term of our model then follows as $\sum_{\vec{k}, \sigma \neq \bar{\sigma}} \psi_{\vec{k}\sigma}^\dagger \hat{\Delta}_{\vec{k}} (\psi_{-\vec{k}, \bar{\sigma}}^\dagger)^\dagger + \text{h.c.}$, where

$$\hat{\Delta}_{\vec{k}} = \begin{pmatrix} \Delta_{d\vec{k}} & e^{i\alpha} \Delta_{dp_x\vec{k}} & e^{i\beta} \Delta_{dp_y\vec{k}} \\ -e^{i\alpha} \Delta_{dp_x\vec{k}} & 0 & 0 \\ -e^{i\beta} \Delta_{dp_y\vec{k}} & 0 & 0 \end{pmatrix}. \quad (2)$$

Here the gap amplitudes Δ_1 and Δ_2 are taken to be real and positive, and the phases α and β remain to be determined. Notice that the interorbital pairings are orbital singlet. To understand how interorbital mixing couples Δ_1 and Δ_2 and fixes α and β , we evaluate the second-order term in the standard free energy expansion, $f^{2\text{nd}} = T \sum_l \sum_{\vec{k}, \omega_m} \text{Tr}[\hat{g}(i\omega_m, \vec{k}) \hat{\Delta}_{\vec{k}} \hat{g}(i\omega_m, \vec{k}) \hat{\Delta}_{\vec{k}}^\dagger]^{2l} / (2l)$, where $\hat{g}(i\omega_m, \vec{k}) = (i\omega_m - \hat{H}_{0\vec{k}})^{-1}$ and $\hat{g}(i\omega_m, \vec{k}) = (i\omega_m + \hat{H}_{0,-\vec{k}}^*)^{-1}$ are the respective electron and hole components of the Gorkov Green's function, T is the temperature, and $\omega_m = (2m+1)\pi T$ is the fermionic Matsubara frequency. This returns the following coupling term in the Ginzburg-Landau free energy,

$$f_{\text{coupling}}^{2\text{nd}} \approx \rho t_{dp} (\cos \alpha - \sin \beta) \Delta_1 \Delta_2, \quad (3)$$

where ρ is a real constant. The effect of the interorbital mixing becomes obvious: the energetically favorable choice would be $(\alpha, \beta) = (0, -\pi/2)$ if $\rho t_{dp} < 0$, and $(\alpha, \beta) = (\pi, \pi/2)$ if $\rho t_{dp} > 0$.

Hall conductivity and Kerr angle. We now proceed to compute the Hall conductance of our model. Within linear response theory, it is given by the antisymmetric part of the current-current correlation function $\pi_{xy}(\vec{q}, \omega)$,

$$\sigma_H(\omega) = \frac{i}{2\omega} \lim_{\vec{q} \rightarrow 0} [\pi_{xy}(\vec{q}, \omega) - \pi_{yx}(\vec{q}, \omega)], \quad (4)$$

where, at the one-loop approximation,

$$\pi_{xy}(\vec{q} = 0, i\omega_n) = T \sum_{\vec{k}, \omega_m} \text{Tr}[\hat{v}_{x\vec{k}} \hat{G}(\vec{k}, i\omega_m) \times \hat{v}_{y\vec{k}} \hat{G}(\vec{k}, i\omega_m + i\omega_n)], \quad (5)$$

where $\omega_n = 2n\pi T$ represents the bosonic Matsubara frequency, $\hat{v}_{x(y)\vec{k}}$ stands for the $x(y)$ component of the velocity operator, and $\hat{G}(\vec{k}, i\omega_m) = (i\omega_m - \hat{H}_{\vec{k}}^{\text{BdG}})^{-1}$ the full Green's function of the corresponding Bogoliubov-de Gennes Hamiltonian associated with Eqs. (1) and (3). In actual calculations

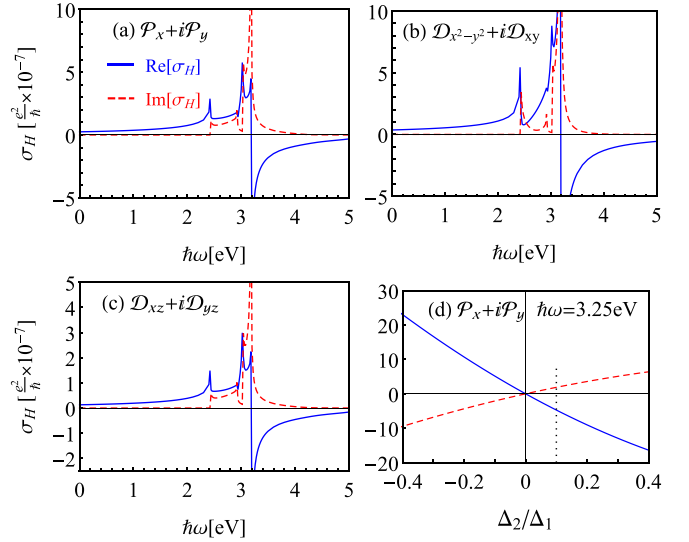


FIG. 2. The real (blue solid line) and imaginary (red dashed line) part of the Hall conductivity for various chiral superconducting states at $T = 0$: (a) $\mathcal{P}_x + i\mathcal{P}_y$, (b) $\mathcal{D}_{x^2-y^2} + i\mathcal{D}_{xy}$, (c) $\mathcal{D}_{xz} + i\mathcal{D}_{yz}$. The interorbital pairing between the d_{xy} and the p orbitals is set at one-tenth of the intraorbital pairing on d_{xy} . The gap functions of the latter two states are presented in the Supplemental Material [28]. (d) The Δ_2 dependence of the Hall conductance at fixed $\Delta_1 = 0.35$ meV and $\hbar\omega = 3.25$ eV.

we transform the Green's function into its spectral representation, and obtain an alternative form of Eq. (5) that has also been employed in Ref. [34]. The Hall conductivity is evaluated by using an analytical continuation to real frequencies $i\omega_n \rightarrow \omega + i\delta$, where δ is taken to be 10^{-5} throughout this study.

Figure 2 presents the representative numerical results for three different chiral states. In accord with a recent experimental estimate [35], we took $\Delta_1 = 0.35$ meV. Without loss of generality, in Figs. 2(a)–2(c) we have taken interorbital pairings that are one-tenth of the intraorbital pairing on the d_{xy} orbital. Given the substantial d - p mixing, such a modest assumption may not be entirely unreasonable, as we substantiate in the Supplemental Material [28]. Overall, the Hall response of the three chiral states shows no qualitative difference. Quantitatively, it is interesting to note that the Hall conductivity in $\mathcal{P}_x + i\mathcal{P}_y$ is approximately twice as large as that in $\mathcal{D}_{xz} + i\mathcal{D}_{yz}$. The factor of 1/2 arises from a k_z integration involving the square of $\sin k_z$ associated with the latter's gap function. In contrast, these two states feature the same thermal Hall conductance [36] and similar spontaneous surface current [37].

As in previous multiorbital models [23,24,34,38,39], the intensity of $\text{Im}[\sigma_H]$ originates from the transitions between pairs of the states belonging to different branches of the Bogoliubov bands, one with positive energy E_1 and the other with negative $-E_2$ ($E_1 \neq E_2$ and $E_1, E_2 > 0$). The lower cutoff frequency ω_c at which $\text{Im}[\sigma_H]$ becomes nonzero is determined by the details of the model. For the specific set of parameters we use, $\hbar\omega_c \approx 2.5$ eV and the corresponding onset intensity is associated with transitions near the M point of the Brillouin zone.

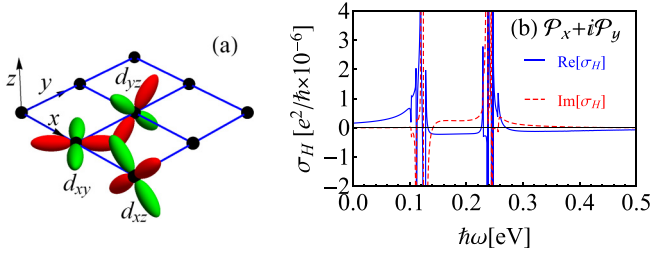


FIG. 3. (a) Lattice structure of the d^3 model. Each site of the square lattice hosts all three Ru t_{2g} orbitals. (b) Real (blue solid line) and imaginary (red dashed line) parts of the Hall conductivity for a chiral p -wave state in the d^3 model. Details of the model are provided in the Supplemental Material [28].

It is worth stressing that a finite interorbital pairing is essential for the emergence of AHE for the pairing model given in Eq. (2). Exemplified by the chiral p -wave state as shown in Fig. 2(d), both real and imaginary parts of σ_H drop to zero linearly as the interorbital pairing decreases. To gain a better understanding, we derive the Hall conductivity of the reduced, and more analytically tractable models containing only d_{xy} and p_x (or d_{xy} and p_y) orbitals. As shown in the Supplemental Material [28], the reduced models have approximately $\sigma_H \propto \Delta_2$ for the chiral p -wave state in the limit of small Δ_2/Δ_1 , consistent with the above numerical results.

Finally, to connect with the optical Kerr measurement, we evaluate the Kerr rotation angle given by

$$\theta_K = \frac{4\pi}{\omega l} \text{Im} \left[\frac{\sigma_H(\omega)}{n(n^2 - 1)} \right], \quad (6)$$

where l stands for the interlayer spacing between the RuO_2 planes and n the frequency-dependent refractive index. Following the estimate in Ref. [23], at the experimental photon energy $\hbar\omega = 0.8$ eV we find $\theta_K \approx 27$, 40, and 14 nrad for the three respective chiral pairings in Fig. 2. They are not far off from the measured value at low temperatures [11]. Nonetheless, caution is needed when using these estimates at face value, as we lack an accurate prediction for the interorbital pairing strength Δ_2 .

d^3 model. In the presence of finite SOC and interlayer coupling, the d_{xy} orbital also mixes with the d_{xz} and d_{yz} orbitals. These couplings are much weaker than t_{dp} [40–42]. However, the two quasi-1D d orbitals in fact lie closer in energy to d_{xy} than the p orbitals do in the dpp model. Moreover, according to a recent theoretical calculation [43], sizable interorbital pairing between the d orbitals is not entirely impossible. It is therefore sensible to consider a model containing all three t_{2g} orbitals and with induced interorbital pairing between the dominant d_{xy} and the other two orbitals (which we refer to as the d^3 model). Note that our motivation differs from a previous study which had assumed comparable pairing instabilities on all three t_{2g} orbitals [34].

Figure 3(b) presents the Hall conductance of a chiral p -wave state (see Supplemental Material [28] for details). In distinction to the dpp model, the characteristic peaks of $\sigma_H(\omega)$ emerge at rather different frequencies due to a very different low-energy quasiparticle spectrum. Notably, at $\hbar\omega = 0.8$ eV, we obtain $\theta_K \approx 63.6$ nrad under the modest assump-

tion of $\Delta_2 = \Delta_1/10$. This is again close to the experimental observation, and we expect similar qualitative behavior for other chiral states.

Nonchiral states. There have been frequent discussions of nonchiral TRSB orders in Sr_2RuO_4 [44–50]. These states condense multiple superconducting order parameters belonging to distinct 1D irreps. However, we find that they most likely do not exhibit intrinsic Hall effect. Some of these states preserve certain vertical mirror symmetries. For this class, σ_H as given by Eqs. (4) and (5) exactly vanishes due to mutually canceled contributions at any pair of \vec{k} 's related by the corresponding mirror reflections. This follows naturally from the symmetry property of the velocity operators under mirror reflections. Some examples are pairings of the forms $A_{1g} + iB_{1g}$ and $B_{1g} + iA_{2g}$ —typically referred to as $\mathcal{S} + i\mathcal{D}_{x^2-y^2}$ and $\mathcal{D}_{x^2-y^2} + i\mathcal{G}_{xy(x^2-y^2)}$, respectively. Some mixed-parity states, for instance $A_{1g} + iA_{1u}$ (a mixture of s -wave and helical p -wave pairings, or simply $\mathcal{S} + i\mathcal{P}$), break all underlying vertical mirror symmetries. However, as we verify numerically, $\hat{v}_{x\vec{k}} \hat{G}(\vec{k}) \hat{v}_{y\vec{k}} \hat{G}(\vec{k}) - (x \leftrightarrow y)$ is zero at every \vec{k} , irrespective of the details of the underlying microscopic model conceivable for this material. Hence these states shall also exhibit vanishing Hall conductance. In short, the presence of a chiral superconducting order parameter appears to be critical for a Hall response to arise in pristine Sr_2RuO_4 . This is consistent with the intuitive expectation by an analogy with quantum Hall insulators.

Concluding remarks. While polar Kerr effect in ultraclean Sr_2RuO_4 may rule against nonchiral states, it cannot reliably discriminate the various chiral states, as our study suggests. A final identification must then be made in conjunction with other key observations. For example, except in rare fine-tuned cases, the $\mathcal{P}_x + i\mathcal{P}_y$ and $\mathcal{D}_{xz} + i\mathcal{D}_{yz}$ states, or more precisely the chiral states in the E_u and E_g irreps, generically support finite spontaneous edge current [37,51], which, however, has eluded experimental detection [52–54]. The $\mathcal{D}_{x^2-y^2} + i\mathcal{D}_{xy}$ state, on the other hand, is understood to produce vanishingly small surface current [55,56]. However, this state, formally classified as $B_{1g} + iB_{2g}$, shall typically exhibit two separate superconducting transitions, whereas experiments have only identified one [57,58]; symmetry analysis would also rule out discontinuities in all shear elastic moduli at the lower superconducting transition, yet a discontinuity was reported in the modulus c_{66} [59]. A final conclusion therefore still seems somewhat distant, and much experimental progress is being made lately [35,44,49,59–62].

In summary, although multiple electron orbitals and a pairing with chirality appear to be critical for superconducting Sr_2RuO_4 to produce intrinsic AHE and Kerr rotation, the Ru quasi-1D d_{xz} and d_{yz} orbitals need not proactively participate in the Cooper pairing. Even when the pairing is driven solely by the Ru d_{xy} orbital, interorbital pairing between this and other orbitals may emerge due to orbital mixing, which then leads to an intrinsic Hall effect. We have demonstrated this for two separate models, one taking into account the O p_x and p_y orbitals in the RuO_2 plane, and another the Ru d_{xz} and d_{yz} orbitals. We evaluated the corresponding Kerr rotation angle and made a connection with the experimental measurement. In this light, it seems worthwhile to reassess the

microscopic theories of the Cooper pairing and the question about the driving superconducting orbital(s) in this material [43,45–47,50,63–89]. Finally, our proposal may be readily generalized to other TRSB superconductors where Kerr rotation has also been reported, including UPt₃, URu₂Si₂, PrOs₄Sb₁₂, and UTe₂ [90–93].

Acknowledgments. We are grateful to Catherine Kallin, Aline Ramires, and Zhiqiang Wang for their critical reading and comments on an earlier version of the manuscript. We also acknowledge fruitful discussions with Thomas Scaffidi, Manfred Sgrist, Qiang-Hua Wang, and Fan Yang. This work

is supported by NSFC under Grants No. 11674278 (F.-C.Z.), and No. 11904155 (W.H. and J.-L.Z.), the strategic priority research program of CAS Grant No. XDB28000000 (F.-C.Z.), and Beijing Municipal Science and Technology Commission Project No. Z181100004218001 (F.-C.Z.), the Guangdong Provincial Key Laboratory under Grant No. 2019B121203002 (W.H.), and the China Postdoctoral Science Foundation under Grant No. 2020M670422 (Y.L.). Computing resources are provided by the Center for Computational Science and Engineering at Southern University of Science and Technology.

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