Supplemental Material for "Possibility of mixed helical p-wave pairing in Sr_2RuO_4"

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CALCULATION OF THE ANOMALOUS HALL CONDUCTIVITY

In the absence of SOC, the two spin blocks decouple. We can calculate $\sigma_{\mathrm{H}}^{\uparrow}$ and $\sigma_{\mathrm{H}}^{\downarrow}$ separately.

Spin up block

The model we consider consists of the two quasi-1d orbitals, d_{xz} and d_{yz}, of Sr_2RuO_4. Written in the basis $\{c_{d_{xz}}(\mathbf{k}), c_{d_{yz}}(\mathbf{k})\}$ the spin up block of the normal state Hamiltonian reads

$$\hat{H}_{\mathrm{N}}(\mathbf{k}) \equiv \begin{pmatrix} \xi_{xz}(\mathbf{k}) & \lambda(\mathbf{k}) \\ \lambda(\mathbf{k}) & \xi_{yz}(\mathbf{k}) \end{pmatrix} = \begin{pmatrix} -2t\cos k_x - 2\tilde{t}\cos k_y - \mu & 4t'\sin k_x \sin k_y \\ 4t'\sin k_x \sin k_y & -2t\cos k_y - 2\tilde{t}\cos k_x - \mu \end{pmatrix}. \tag{1}$$

The corresponding BdG Hamiltonian is

$$\hat{H}^{\uparrow}_{\mathrm{BdG}}(\mathbf{k}) = \begin{pmatrix} \hat{H}_{\mathrm{N}}(\mathbf{k}) & \hat{\Delta}^{\uparrow\uparrow}(\mathbf{k}) \\ [\hat{\Delta}^{\uparrow\uparrow}]^{\dagger}(\mathbf{k}) & -[\hat{H}_{\mathrm{N}}]^{T}(-\mathbf{k}) \end{pmatrix} \tag{2}$$

with

$$\hat{\Delta}^{\uparrow\uparrow}(\mathbf{k}) = -(\Delta_{A_{1u}} - \Delta_{A_{2u}}) \begin{pmatrix} \sin k_x & 0 \\ 0 & -i\sin k_y \end{pmatrix}, \tag{3}$$

which is effectively a (spinless) chiral $p_x - ip_y$ order parameter. For simplicity we have considered only intra-orbital pairing while neglected any inter-orbital one. The d_{xz} (d_{yz}) intraorbital component $\Delta_{11}^{\uparrow\uparrow}$ ($\Delta_{22}^{\uparrow\uparrow}$) transforms like k_x (k_y) under the spatial $D_{4\mathrm{h}}$ group. Hence, $\Delta_{11}^{\uparrow\uparrow}(\mathbf{k}) = \left[\mathbf{d}_{\mathbf{k}}^{(x)} \cdot \boldsymbol{\sigma} i\sigma_2\right]_{\uparrow\uparrow} = -(\Delta_{A_{1u}} - \Delta_{A_{2u}})\sin k_x$, where $\mathbf{d}_{\mathbf{k}}^{(x)}$ denotes the k_x-like part of $\mathbf{d}_{\mathbf{k}}$. Note that $\mathbf{d}_{\mathbf{k}} = \Delta_{A_{1u}}(\hat{x}\sin k_x + \hat{y}\sin k_y) + \Delta_{A_{2u}}(\hat{x}\sin k_y - \hat{y}\sin k_x)$. Similarly, $\Delta_{22}^{\uparrow\uparrow}(\mathbf{k}) = \left[\mathbf{d}_{\mathbf{k}}^{(y)} \cdot \boldsymbol{\sigma} i\sigma_2\right]_{\uparrow\uparrow}$. From H_{BdG} we can define the electric current velocity operator

$$\hat{\mathbf{v}}(\mathbf{k}) = \begin{pmatrix} \nabla_{\mathbf{k}}\xi_{xz}(\mathbf{k}) & \nabla_{\mathbf{k}}\lambda(\mathbf{k}) \\ \nabla_{\mathbf{k}}\lambda(\mathbf{k}) & \nabla_{\mathbf{k}}\xi_{yz}(\mathbf{k}) \end{pmatrix} \otimes 1_{2\times 2}, \tag{4}$$

where $1_{2\times 2}$ is the identity matrix in the Nambu particle-hole space.

We calculate $\sigma_{\mathrm{H}}^{\uparrow}$ using the standard one-loop Kubo formula

$$\sigma_{\mathrm{H}}^{\uparrow}(\omega) = \frac{i}{\omega}\frac{\pi_{xy}(\mathbf{q}=0, \omega + i0^+) - \pi_{yx}(\mathbf{q}=0, \omega + i0^+)}{2}, \tag{5}$$

where $\pi_{xy}(\mathbf{q}, i\nu_m)$ is the electric current-current density correlator, given by

$$\pi_{xy}(0, i\nu_m) = e^2 T \sum_n \sum_{\mathbf{k}} \mathrm{Tr}[\hat{v}_x(\mathbf{k})\hat{G}(\mathbf{k}, i\omega_n)\hat{v}_y(\mathbf{k})\hat{G}(\mathbf{k}, i\omega_n + i\nu_m)]. \tag{6}$$

In this equation $\hat{G}(\mathbf{k}, i\omega_n) \equiv [i\omega_n - \hat{H}^{\uparrow}_{\mathrm{BdG}}(\mathbf{k})]^{-1}$ is the Green's function. $\omega_n = (2n+1)\pi T$ and $\nu_m = 2m\pi T$ are Fermionic and Bosonic Matsubara frequencies. The trace Tr is with respect to both the Nambu particle-hole and orbital spaces. e is the charge of an electron. Using Eq. (2) in the expression of $\pi_{xy}(0, i\nu_m)$ in Eq. (6), completing the Matsubara sum over ω_n, and then making the analytical continuation, $i\nu_m \to \omega + i\delta$, we obtain

$$\frac{\sigma_{\mathrm{H}}^{\uparrow}(\omega)}{e^2/\hbar} = \sum_{\mathbf{k}} \frac{\mathcal{F}_{\mathbf{k}}^{\uparrow}}{E_{+}^{\uparrow}(\mathbf{k})E_{-}^{\uparrow}(\mathbf{k})[E_{+}^{\uparrow}(\mathbf{k}) + E_{-}^{\uparrow}(\mathbf{k})][(E_{+}^{\uparrow}(\mathbf{k}) + E_{-}^{\uparrow}(\mathbf{k}))^2 - (\omega + i\delta)^2]}, \tag{7}$$

where

$$\mathcal{F}^{\uparrow}_{\mathbf{k}} = 32\ (t-\tilde{t})(t')^2(\Delta_1-\Delta_2)^2(\sin^2 k_x \cos k_y + \sin^2 k_y \cos k_x)\sin^2 k_x \sin^2 k_y. \tag{8}$$

$E^{\uparrow}_{\pm}(\mathbf{k})$ are eigenvalues of $\hat{H}^{\uparrow}_{\text{BdG}}(\mathbf{k})$.

$$E^{\uparrow}_{\pm} = \sqrt{\frac{-\alpha \pm \sqrt{\alpha^2-4\beta}}{2}}, \tag{9a}$$

$$\alpha = -(\xi_{xz}^2 + |\Delta^{\uparrow\uparrow}_{11}|^2 + \xi_{yz}^2 + |\Delta^{\uparrow\uparrow}_{22}|^2 + 2\lambda^2), \tag{9b}$$

$$\beta = (\xi_{xz}^2 + |\Delta^{\uparrow\uparrow}_{11}|^2)(\xi_{yz}^2 + |\Delta^{\uparrow\uparrow}_{22}|^2) + \lambda^4 + \lambda^2([\Delta^{\uparrow\uparrow}_{11}]^*\Delta^{\uparrow\uparrow}_{22} + \Delta^{\uparrow\uparrow}_{11}[\Delta^{\uparrow\uparrow}_{22}]^*) - 2\lambda^2\xi_{xz}\xi_{yz}. \tag{9c}$$

For brevity we have suppressed the $\mathbf{k}$ dependence in these equations.

Written out explicitly,

$$\text{Re}[\sigma^{\uparrow}_{\text{H}}](\omega) = \frac{e^2}{\hbar}\sum_{\mathbf{k}} \frac{\mathcal{F}^{\uparrow}_{\mathbf{k}}}{E^{\uparrow}_{+}E^{\uparrow}_{-}(E^{\uparrow}_{+}+E^{\uparrow}_{-})[(E^{\uparrow}_{+}+E^{\uparrow}_{-})^2-\omega^2]}, \tag{10}$$

$$\text{Im}[\sigma^{\uparrow}_{\text{H}}](\omega) = \frac{e^2}{\hbar}\sum_{\mathbf{k}} \frac{\mathcal{F}^{\uparrow}_{\mathbf{k}}}{E^{\uparrow}_{+}E^{\uparrow}_{-}}\frac{\pi}{2\omega^2}\Big[\delta(\omega-(E^{\uparrow}_{+}+E^{\uparrow}_{-})) - \delta(\omega+(E^{\uparrow}_{+}+E^{\uparrow}_{-}))\Big]. \tag{11}$$

Spin down block

The derivation of $\sigma^{\downarrow}_{\text{H}}$ is almost identical. The only difference is that in the definition of the BdG Hamiltonian, $\hat{\Delta}^{\uparrow\uparrow}$ is now replaced by

$$\hat{\Delta}^{\downarrow\downarrow}(\mathbf{k}) = (\Delta_{A_{1u}} + \Delta_{A_{2u}})\begin{pmatrix}\sin k_x & 0\\ 0 & i\sin k_y\end{pmatrix}. \tag{12}$$

Correspondingly,

$$\frac{\sigma^{\downarrow}_{\text{H}}(\omega)}{e^2/\hbar} = \sum_{\mathbf{k}} \frac{\mathcal{F}^{\downarrow}_{\mathbf{k}}}{E^{\downarrow}_{+}E^{\downarrow}_{-}(E^{\downarrow}_{+}+E^{\downarrow}_{-})[(E^{\downarrow}_{+}+E^{\downarrow}_{-})^2-(\omega+i\delta)^2]} \tag{13}$$

with

$$\mathcal{F}^{\downarrow}_{\mathbf{k}} = -32(t-\tilde{t})(t')^2(\Delta_1+\Delta_2)^2(\sin^2 k_x \cos k_y + \sin^2 k_y \cos k_x)\sin^2 k_x \sin^2 k_y. \tag{14}$$

$E^{\downarrow}_{\pm}$ differs from $E^{\uparrow}_{\pm}$ in Eq. (9) by replacing $\{\Delta^{\uparrow\uparrow}_{11}, \Delta^{\uparrow\uparrow}_{22}\}$ with $\{\Delta^{\downarrow\downarrow}_{11}, \Delta^{\downarrow\downarrow}_{22}\}$. Notice that although the gap magnitudes can be quite different for spin up and down, $\min\{E^{\uparrow}_{+}(\mathbf{k}) + E^{\uparrow}_{-}(-\mathbf{k})\}$, which determines the onset frequency of $\text{Im}[\sigma^{\uparrow}_{\text{H}}](\omega)$ (see Eq. (11)), is actually almost the same as $\min\{E^{\downarrow}_{+}(\mathbf{k}) + E^{\downarrow}_{-}(-\mathbf{k})\}$, because both of them are governed by the orbital-hybridization parameter t' [1], instead of the pairing gap magnitudes. This explains why, in Fig. 1, $\text{Im}[\sigma^{\uparrow}_{\text{H}}](\omega)$ and $\text{Im}[\sigma^{\downarrow}_{\text{H}}](\omega)$ become nonzero essentially at the same frequency.

Comparing $\mathcal{F}^{\downarrow}_{\mathbf{k}}$ to $\mathcal{F}^{\uparrow}_{\mathbf{k}}$ in Eq. (8) we see that they carry opposite signs, leading to a partial cancellation between $\sigma^{\downarrow}_{\text{H}}$ and $\sigma^{\uparrow}_{\text{H}}$ (see Fig. 1). This sign difference comes from $\hat{\Delta}^{\downarrow\downarrow}$ having a chirality opposite to that of $\hat{\Delta}^{\uparrow\uparrow}$. Also, under a relative sign change between $\Delta_{A_{1u}}$ and $\Delta_{A_{2u}}$, $\sigma_{\text{H}} \to -\sigma_{\text{H}}$, because that relative sign determines which chirality of the pairing, $p_x - ip_y$ (associated with spin-up) or $p_x + ip_y$ (with spin-down), dominates. This can be seen from Fig. 2, where we plot $\sigma_{\text{H}}(\omega)$ for a given frequency as a function of $4\Delta_{A_{1u}}\Delta_{A_{2u}}/(|\Delta_{A_{1u}}| + |\Delta_{A_{2u}}|)^2$. Fig. 2 also shows that $\sigma_{\text{H}} = 0$ if $\Delta_{A_{1u}} = 0$ or $\Delta_{A_{2u}} = 0$. This is expected since in this case our mixed helical pairing state is reduced to one of the two single-representation helical states, either A_{1u} or A_{2u}, for which the two chiral components completely compensate each other.

ESTIMATION OF THE KERR ANGLE

From the calculated $\sigma_{\text{H}}(\omega)$ we can estimate the Kerr angle for $\hbar\omega = 0.8$eV using

$$\theta_{\text{K}}(\omega) = \frac{4\pi}{\omega d}\text{Im}[\frac{\sigma_{\text{H}}(\omega)}{n(n^2-1)}], \tag{15}$$

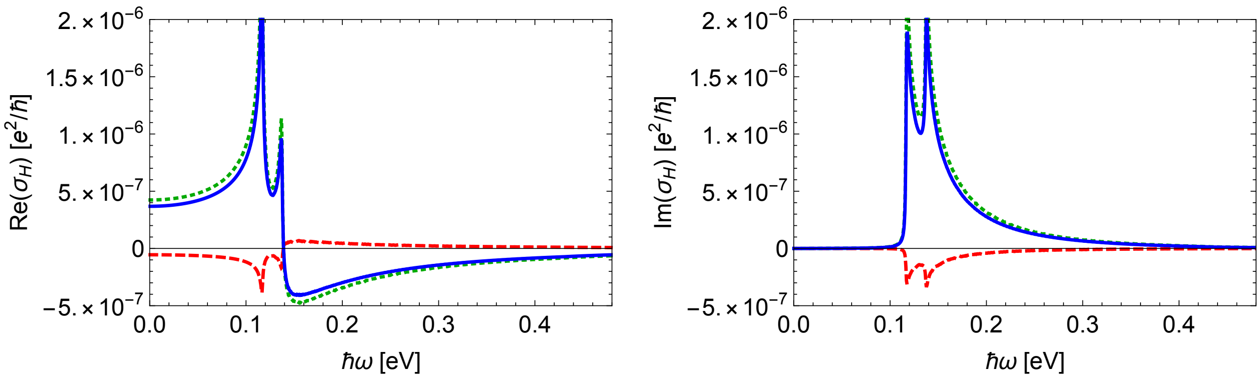

FIG. 1. Spin $\uparrow$ (red dashed line), $\downarrow$ (dark green dotted line), and the total (blue solid line) contributions to the $T = 0$ Hall conductivity. Left: Re[σ_{H}]; Right: Im[σ_{H}]. Band parameters used are [1]: $t = \mu = 0.4\,\mathrm{eV}$, $\tilde{t} = 0.1t$, and $t' = 0.05t$. $\Delta_{A_{1u}} = 2\Delta_{A_{2u}} = \frac{2}{3}\Delta_{\max}$ with $\Delta_{\max} = 0.23\,\mathrm{meV}$ [1].

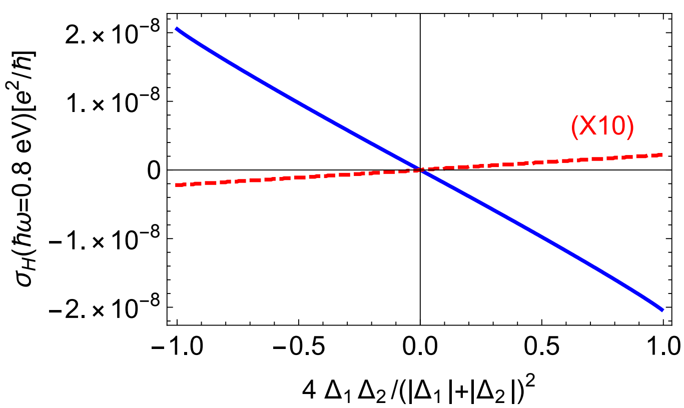

FIG. 2. Real (blue solid line) and imaginary (red dashed line) parts of the zero temperature $\sigma_{\mathrm{H}}(\omega)$ as a function of $4\Delta_{A_{1u}}\Delta_{A_{2u}}/(|\Delta_{A_{1u}}| + |\Delta_{A_{2u}}|)^2$. $\hbar\omega = 0.8\,\mathrm{eV}$. $\Delta_{\max} = |\Delta_{A_{1u}}| + |\Delta_{A_{2u}}| = 0.23\,\mathrm{meV}$ is kept a constant in this calculation. The plot shows that σ_{H} is roughly linear in $4\Delta_{A_{1u}}\Delta_{A_{2u}}/(|\Delta_{A_{1u}}|+|\Delta_{A_{2u}}|)^2$. From the plot we see that σ_{H} is odd in the relative sign between $\Delta_{A_{1u}}$ and $\Delta_{A_{2u}}$, but even under the interchange $|\Delta_{A_{1u}}| \leftrightarrow |\Delta_{A_{2u}}|$.

where $n = n(\omega)$ is the complex index of refraction, given by

$$n = \sqrt{\epsilon_{ab}(\omega)}, \tag{16}$$

$$\epsilon_{ab}(\omega) = \epsilon_\infty + \frac{4\pi i}{\omega}\sigma(\omega). \tag{17}$$

$\epsilon_{ab}(\omega)$ is the permeability tensor in the ab-plane. $\epsilon_\infty = 10$ [1] is the background permeability. $d = 6.8\mathring{A}$ is the inter-layer spacing along the c-axis. $\sigma(\omega)$ is the optical longitudinal conductivity. Following Ref. [1] we use a simple Drude model for $\sigma(\omega)$

$$\sigma(\omega) = -\frac{\omega_{\mathrm{pl}}^2}{4\pi i(\omega + i\Gamma)}, \tag{18}$$

where $\omega_{\mathrm{pl}} = 2.9\,\mathrm{eV}$ is the the plasma frequency and $\Gamma = 0.4\,\mathrm{eV}$ is an elastic scattering rate. At $\hbar\omega = 0.8\,\mathrm{eV}$, $\sigma(\omega) = 0.33 + i\,0.67$, $\epsilon_{ab} = -0.52 + i\,5.20$, and $n = 1.53 + i\,1.69$. Plugging this n value into Eq. (15) and using the $\sigma_{\mathrm{H}}(\hbar\omega = 0.8\mathrm{eV})$ value from Fig. 2 we get $\theta_{\mathrm{K}} \approx 20\,\mathrm{nrad}$. The whole frequency dependence of θ_{K} is shown in Fig. 3.

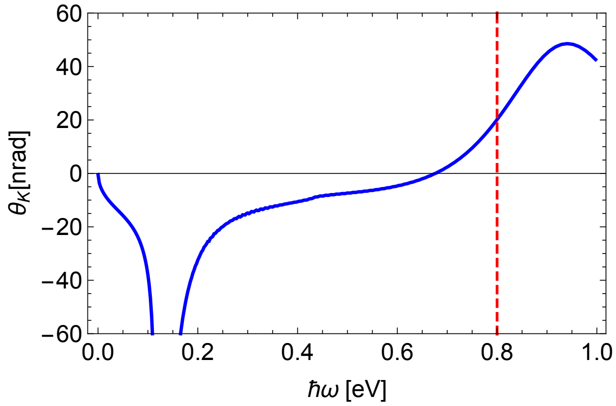

FIG. 3. (color online) Kerr angle as a function of $\hbar\omega$. Band parameters and the values of $\{\Delta_{A_{1u}}, \Delta_{A_{2u}}\}$ are the same as in Fig. 1.

RESULTS FOR CHIRAL P-WAVE PAIRING

For easy comparison, in this section, we reproduce some of the results of $\sigma_{\rm H}$ and $\theta_{\rm K}$ obtained in Ref. [1] for the chiral $p_x + ip_y$ pairing. In this case the pairing is between spin up and down with the nonzero order parameter components given by

$$\hat{\Delta}^{\uparrow\downarrow}(\mathbf{k}) = \Delta_0 \begin{pmatrix} \sin k_x & 0 \\ 0 & i\sin k_y \end{pmatrix}, \hat{\Delta}^{\downarrow\uparrow}(\mathbf{k}) = \hat{\Delta}^{\uparrow\downarrow}(\mathbf{k}). \tag{19}$$

In the absence of SOC, the BdG Hamiltonian can be again decomposed into two decoupled blocks. One of them is

$$\hat{H}^{\uparrow}_{\rm BdG}(\mathbf{k}) = \begin{pmatrix} \hat{H}_{\rm N}(\mathbf{k}) & \hat{\Delta}^{\uparrow\downarrow}(\mathbf{k}) \\ [\hat{\Delta}^{\uparrow\downarrow}]^{\dagger}(\mathbf{k}) & -[\hat{H}_{\rm N}]^T(-\mathbf{k}) \end{pmatrix}, \tag{20}$$

which is written in the basis $\{c^{\dagger}_{d_{xz},\uparrow}(\mathbf{k}), c^{\dagger}_{d_{yz},\uparrow}(\mathbf{k}), c_{d_{xz},\downarrow}(-\mathbf{k}), c_{d_{yz},\downarrow}(-\mathbf{k})\}$. The other block is for the same basis but with opposite spin, and we denote it by $\hat{H}^{\downarrow}_{\rm BdG}(\mathbf{k})$. Because $\hat{\Delta}^{\downarrow\uparrow} = \hat{\Delta}^{\uparrow\downarrow}$, $\hat{H}^{\downarrow}_{\rm BdG} = \hat{H}^{\uparrow}_{\rm BdG}$. Since the expression of $\hat{H}^{\uparrow}_{\rm BdG}$ in Eq. (20) is almost identical to that in Eq. (2) we can immediately write down the expression of $\sigma^{\uparrow}_{\rm H}$,

$$\frac{\sigma^{\uparrow}_{\rm H}(\omega)}{e^2/\hbar} = \sum_{\mathbf{k}} \frac{\mathcal{F}_{\mathbf{k}}}{E_+(\mathbf{k})E_-(\mathbf{k})[E_+(\mathbf{k}) + E_-(\mathbf{k})][(E_+(\mathbf{k}) + E_-(\mathbf{k}))^2 - (\omega + i\delta)^2]}, \tag{21}$$

where

$$\mathcal{F}_{\mathbf{k}} = -32\,(t - \tilde{t})(t')^2\Delta_0^2(\sin^2 k_x \cos k_y + \sin^2 k_y \cos k_x)\sin^2 k_x \sin^2 k_y \tag{22a}$$

$$E_{\pm} = \sqrt{\frac{-\alpha \pm \sqrt{\alpha^2 - 4\beta}}{2}}, \tag{22b}$$

$$\alpha = -(\xi_{xz}^2 + 2|\Delta_0|^2 + \xi_{yz}^2 + 2\lambda^2), \tag{22c}$$

$$\beta = (\xi_{xz}^2 + |\Delta_0|^2)(\xi_{yz}^2 + |\Delta_0|^2) + \lambda^4 - 2\lambda^2\xi_{xz}\xi_{yz}. \tag{22d}$$

As a consequence of $\hat{H}^{\downarrow}_{\rm BdG} = \hat{H}^{\uparrow}_{\rm BdG}$, $\sigma^{\downarrow}_{\rm H} = \sigma^{\uparrow}_{\rm H}$ for the chiral p-wave pairing such that $\sigma_{\rm H} = \sigma^{\downarrow}_{\rm H} + \sigma^{\uparrow}_{\rm H} = 2\sigma^{\uparrow}_{\rm H}$. This should be contrasted with the previous $A_{1u} + iA_{2u}$ pairing case where $\sigma^{\downarrow}_{\rm H}$ and $\sigma^{\uparrow}_{\rm H}$ carry opposite signs.

Fig. 4 shows the corresponding numerical results of $\sigma_{\rm H}$ and $\theta_{\rm K}$. Here, $\theta_{\rm K}$ is evaluated from $\sigma_{\rm H}$ using Eq. (15) with the same index of refraction $n(\omega)$. From Fig. 4 we see that $\theta_{\rm K}(\hbar\omega = 0.8\,{\rm eV}) \approx 46\,{\rm nrad}$, which is about twice larger than the estimated value for the $A_{1u} + iA_{2u}$ pairing (see Fig. 3). The larger $\theta_{\rm K}$ in the current case comes from the absence of cancellation, i. e. $\sigma^{\downarrow}_{\rm H}$ and $\sigma^{\uparrow}_{\rm H}$ carry the same sign and the two add together instead of cancel each other.

ABSENCE OF CROSS-GRADIENT TERMS IN THE GINZBURG-LANDAU FREE ENERGY

Using the $A_{1u} + iA_{2u}$ state as an example, we argue that there are no cross-gradient terms in the free energy that are quadratic in the order parameter components. There are in total four possible such terms that involve both order parameter components as

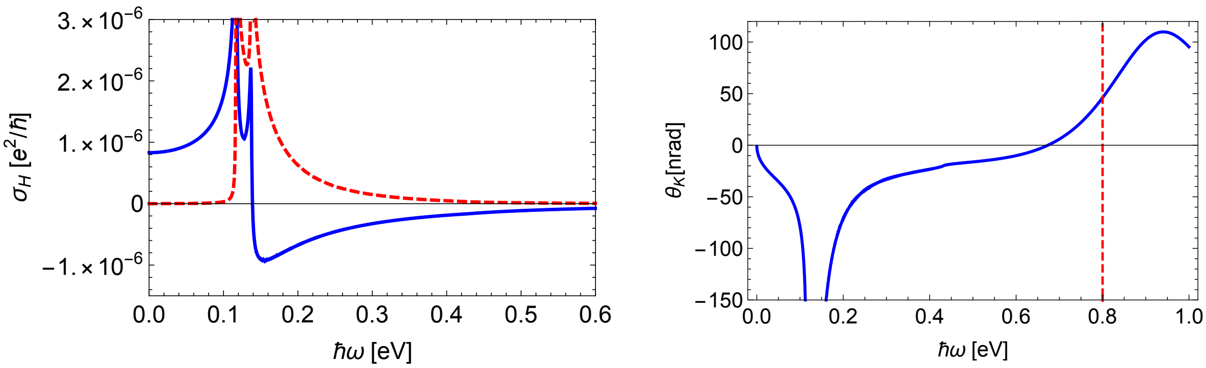

FIG. 4. Results of $\sigma_{\rm H}$ and $\theta_{\rm K}$ for the chiral p-wave pairing. For this calculation the normal state band parameters used are the same as in Fig. 1; $\Delta_0 = 0.23\,$meV. In the left plot, the blue solid (red dashed) line is for the real (imaginary) part of $\sigma_{\rm H}$. In the right plot, the dashed line shows that $\theta_{\rm K}(\hbar\omega = 0.8\,{\rm eV}) \approx 46\,$nrad.

well as gradients in both x and y directions:

$$a\partial_x\Delta^*_{A_{1u}}\partial_y\Delta_{A_{2u}} + b\partial_y\Delta^*_{A_{1u}}\partial_x\Delta_{A_{2u}} + c\partial_x\Delta^*_{A_{2u}}\partial_y\Delta_{A_{1u}} + d\partial_y\Delta^*_{A_{2u}}\partial_x\Delta_{A_{1u}} \tag{23}$$

where the coefficients a, b, c and d assume the values of 1 or -1 and we have dropped an overall Ginzburg-Landau coefficient. The general requirement is that the free energy be real and invariant under all D_{4h} point group symmetry transformations. Bearing in mind the symmetry properties of the two individual components, one can check that no single set of $\{a, b, c, d\}$ ensures the invariance of the above free energy under all symmetry operations. One thus concludes that these cross-gradient terms must be absent. Same argument applies to the $B_{1u} + iB_{2u}$ state.

[1] E. Taylor, and C. Kallin, Intrinsic Hall Effect in a Multiband Chiral Superconductor in the Absence of an External Magnetic Field, Phys. Rev. Lett. 108, 157001 (2012).