

# Odd-frequency pairing and Kerr effect in the heavy-fermion superconductor $\text{UPt}_3$

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We study the emergence of odd-frequency superconducting pairing in  $\text{UPt}_3$ . Starting from a tight-binding model accounting for the nonsymmorphic crystal symmetry of  $\text{UPt}_3$  and assuming an order parameter in the  $E_{2u}$  representation, we demonstrate that odd-frequency pairing arises very generally, as soon as intersublattice hopping or spin-orbit coupling is present. We also show that in the low temperature superconducting  $B$  phase, the presence of a chiral order parameter together with spin-orbit coupling, leads to additional odd-frequency pair amplitudes not present in the  $A$  or  $C$  phases. Furthermore, we show that a finite Kerr rotation in the  $B$  phase is only present if odd- $\omega$  pairing also exists.

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## I. INTRODUCTION

The heavy-fermion material  $\text{UPt}_3$  has a truly unconventional superconducting phase diagram, possessing two zero-field superconducting phases, the  $A$  phase and the  $B$  phase, with critical temperatures  $T_{c,+} \approx 550$  mK and  $T_{c,-} \approx 500$  mK [1,2], respectively. Additionally, a third phase, the  $C$  phase, emerges at high magnetic field [3]. Knight shift observations point to a spin-triplet superconducting order parameter [4]. Josephson interferometry has revealed the presence of line nodes in the  $A$  phase [5], as well as the onset of a complex order parameter in the  $B$  phase [5,6]. Recently, measurements of the Kerr effect have also demonstrated time-reversal symmetry breaking in the  $B$  phase [7]. These observations appear to be consistent with a gap belonging to the  $E_{2u}$  representation [2,5,6,8]. Furthermore, recent work considering the nonsymmorphic crystal structure [9,10] suggested that the measured value of the Kerr rotation is related to the presence of a nonunitary linear combination of  $f$ -wave and  $d$ -wave pairing in the  $B$  phase [11].

It is well known that the fermionic nature of electrons tightly constrains the allowed symmetries of the superconducting gap function. Specifically, in the limit of equal-time pairing and a single-component gap, spatially even-parity gap functions (like the  $s$  or  $d$  wave) must correspond to spin-singlet states, while odd-parity gap functions ( $p$  or  $f$  wave) must correspond to spin-triplet states. However, if the electrons comprising the condensate are paired at unequal times the superconducting gap can be odd in time, or equivalently, odd in frequency (odd- $\omega$ ). In that case the condensate can be even in spatial parity and spin-triplet or odd-parity and spin-singlet. This possibility, originally posited for  $^3\text{He}$  by Berezinskii [12] and then later for superconductivity [13–15], is intriguing both because of the unconventional symmetries which it permits and for the fact that it represents a class of hidden order due to the vanishing of all equal time correlations.

While the thermodynamic stability of intrinsically odd- $\omega$  phases is, so far, only discussed as a theoretical possibility [16–23], significant progress has been made understanding systems with conventional superconductors in which odd- $\omega$  pairing can be induced by altering the native superconducting correlations [24–42]. Well-established examples can be found

in ferromagnet-superconductor junctions [24–30] in which experiments have recently observed key signatures of odd- $\omega$  pair correlations [43,44]. For a modern review of this field see Ref. [45].

Another intriguing possibility for odd- $\omega$  superconductivity can be found in multiband superconductors in which it has been shown that odd- $\omega$  pairing is ubiquitously induced in the presence of interband hybridization [46–49]. As an illustrative example, if we consider a generic two-band model:  $H = \xi_1 \psi_1^\dagger \psi_1 + \xi_2 \psi_2^\dagger \psi_2 + \Delta_1 \psi_1^\dagger \psi_1^\dagger + \Delta_2 \psi_2^\dagger \psi_2^\dagger + \text{H.c.}$ , the addition of any finite hybridization term of the form  $\Gamma \psi_1^\dagger \psi_2$  induces odd- $\omega$  interband pairing proportional to the difference of the two gaps,  $\sim \omega \Gamma (\Delta_1 - \Delta_2)$  [46,49]. An interband hybridization of this form is intrinsic to the superconductor whenever there is a mismatch between the orbital character of the Cooper pairs and that of the quasiparticles of the normal state, or alternatively, it can arise from scattering processes in the presence of disorder [47,48]. In contrast to other known mechanisms for realizing odd- $\omega$  pairing which employ superconductor heterostructures, multiband superconductors allow for the generation of odd- $\omega$  pair amplitudes in the bulk, without breaking either spatial translation or time-reversal symmetry. With many known multiband superconductors with highly unconventional features, such as  $\text{Sr}_2\text{RuO}_4$ , [50,51] iron-based superconductors [52,53–56],  $\text{MgB}_2$  [57–61], and  $\text{UPt}_3$  [1–3,5,6], it remains a very interesting question how much odd- $\omega$  superconductivity contributes to their physical properties.

It was recently shown that the multiband superconductor  $\text{Sr}_2\text{RuO}_4$  hosts odd- $\omega$  pairing due to the finite hybridization between the different orbitals in the normal state [48]. At the same time it was shown that the conditions leading to the observation of a finite Kerr rotation angle also guarantee the emergence of odd- $\omega$  pairing. Since the Kerr effect has been observed in  $\text{Sr}_2\text{RuO}_4$  [62], this directly implies that  $\text{Sr}_2\text{RuO}_4$  hosts odd- $\omega$  pair amplitudes. The Kerr effect has also been observed in the multiband superconductor  $\text{UPt}_3$  [7]; however, while  $\text{Sr}_2\text{RuO}_4$  is a relatively simple system assumed to possess a  $p$ -wave order parameter, the gap structure in  $\text{UPt}_3$  is thought to be primarily  $f$  wave and likely with additional character. Given that the Kerr rotation angle is known to be highly sensitive to system-specific details, such as the existence

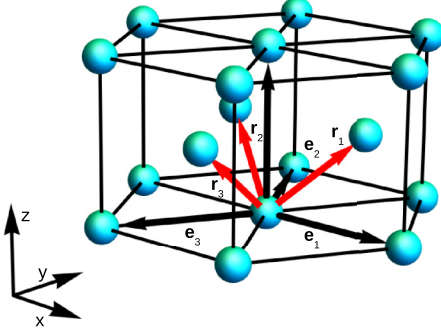


FIG. 1. Schematic depiction of the locations of the U atoms in the  $AB$ -stacked hexagonal lattice of  $UPt_3$  with vectors in the basal plane labeled  $\mathbf{e}_i$  and intersublattice vectors labeled  $\mathbf{r}_i$ .

of interband transitions [63–65] or the presence of disorder [66] the observation of a finite Kerr rotation angle in  $UPt_3$  cannot simply be presumed to imply the presence of odd- $\omega$  pairing also in this material. The purpose of this work is therefore to elucidate the possibility of odd- $\omega$  pairing channels in  $UPt_3$  and, if possible, connect these to the Kerr effect found in the  $B$  phase of  $UPt_3$ .

In this work we use a tight-binding model, which, while simple enough to allow for analytical results, captures the  $5f$  states on the U atoms, the main Fermi surfaces, and explicitly takes into account the nonsymmorphic symmetry of the lattice. Adhering to the bulk of experimental results we further assume spin-triplet pairing within the  $E_{2u}$  irreducible representation. By calculating the full anomalous Green's function we are able to extract all possible odd- $\omega$  pairing amplitudes in  $UPt_3$ . We find that as soon as intersublattice hopping is present, i.e., out-of-plane nearest neighbor U-U hopping, intrasublattice odd- $\omega$  pairing emerges in all three superconducting phases of  $UPt_3$ . For finite spin-orbit coupling, we find that intersublattice odd- $\omega$  pairing is also always present in all three phases. In the  $B$  phase we find additional inter and intrasublattice odd- $\omega$  pairing due to spin-orbit coupling and the nonunitary order parameter. We furthermore compare the criteria for the existence of odd- $\omega$  pairing and finite Kerr effect, as experimentally measured in the  $B$  phase of  $UPt_3$ . We are able to show that the conditions needed for a finite Kerr rotation angle automatically lead to odd-frequency pairing. Thus, the finite Kerr effect measured in the  $B$  phase serves as experimental evidence for odd- $\omega$  superconductivity in  $UPt_3$ .

The remainder of this article is organized as follows. In Sec. II we introduce the model used to describe the electronic properties of  $UPt_3$  and discuss the assumptions used to solve it analytically. In Sec. III we perform our analysis of the symmetries exhibited by the anomalous Green's functions, finding the conditions under which odd- $\omega$  pairing is expected to emerge in  $UPt_3$ . In Sec. IV we compare these conditions to previous theoretical work characterizing the Kerr effect in  $UPt_3$ . In Sec. V we conclude our study.

## II. MODEL

In Fig. 1 we show the three-dimensional (3D) crystal structure of  $UPt_3$  with the U ions (blue spheres) forming  $AB$ -stacked layered triangular lattices with basal plane lattice vectors

given by:  $\mathbf{e}_1 = (1, 0, 0)$ ,  $\mathbf{e}_2 = (-\frac{1}{2}, \frac{\sqrt{3}}{2}, 0)$ ,  $\mathbf{e}_3 = (-\frac{1}{2}, -\frac{\sqrt{3}}{2}, 0)$  and intersublattice vectors given by:  $\mathbf{r}_1 = (\frac{1}{2}, \frac{1}{2\sqrt{3}}, \frac{1}{2})$ ,  $\mathbf{r}_2 = (-\frac{1}{2}, \frac{1}{2\sqrt{3}}, \frac{1}{2})$ , and  $\mathbf{r}_3 = (0, -\frac{1}{\sqrt{3}}, \frac{1}{2})$ . The Pt ions (omitted for simplicity) are located between each of the nearest-neighbor U ions. To model the electronic properties of  $UPt_3$  near the Fermi level we focus on the itinerant  $5f$  electrons originating from the U atoms. Motivated by previous work we assume the same tight-binding Bogoliubov–de Gennes Hamiltonian [9–11]

$$\begin{aligned} \mathcal{H} = & \sum_{\mathbf{k}, m, \sigma} \xi_{\mathbf{k}} c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}m\sigma} + \sum_{\mathbf{k}, \sigma} [\epsilon_{\mathbf{k}} c_{\mathbf{k}1\sigma}^\dagger c_{\mathbf{k}2\sigma} + \text{H.c.}] \\ & + \sum_{\mathbf{k}, m, m', \sigma, \sigma'} g_{\mathbf{k}} \tau_{mm'}^z \otimes \sigma_{\sigma\sigma'}^z c_{\mathbf{k}m\sigma}^\dagger c_{\mathbf{k}m'\sigma'} \\ & + \frac{1}{2} \sum_{\mathbf{k}, m, m', \sigma, \sigma'} [\Delta_{mm', \sigma\sigma'}(\mathbf{k}) c_{\mathbf{k}m\sigma}^\dagger c_{-\mathbf{k}m'\sigma'}^\dagger + \text{H.c.}], \quad (1) \end{aligned}$$

where  $c_{\mathbf{k}m\sigma}^\dagger$  ( $c_{\mathbf{k}m\sigma}$ ) creates (annihilates) a fermionic quasiparticle with crystal momentum  $\mathbf{k}$ , on sublattice  $m = \{1, 2\}$ , and with spin  $\sigma = \{\uparrow, \downarrow\}$ . The intersublattice hopping terms are given by  $\xi_{\mathbf{k}} = 2t \sum_i \cos \mathbf{k}_{\parallel} \cdot \mathbf{e}_i + 2t_z \cos k_z - \mu$ , with  $\mathbf{k}_{\parallel} \equiv (k_x, k_y, 0)$ , which is manifestly even in  $\mathbf{k}$ . The intersublattice hopping terms are given by  $\epsilon_{\mathbf{k}} = 2t' \cos \frac{k_z}{2} \sum_i e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}_i}$ , which is, in general, complex with  $\text{Re}\{\epsilon_{\mathbf{k}}\}$  even in  $\mathbf{k}$ , while  $\text{Im}\{\epsilon_{\mathbf{k}}\}$  is odd in  $\mathbf{k}$ . The Kane-Mele-like spin-orbit coupling is described by  $g_{\mathbf{k}} = \alpha \sum_i \sin \mathbf{k}_{\parallel} \cdot \mathbf{e}_i$  which is clearly odd in  $\mathbf{k}$ . Following the same conventions, a general superconducting order parameter is written as  $\Delta_{mm', \sigma\sigma'}(\mathbf{k})$ . As is widely done, we assume an order parameter within the  $E_{2u}$  irreducible representation and with spin-triplet  $m_z = 0$  pairing [2,9,67].

Quantum oscillation measurements employing the de Haas–van Alphen effect [68,69] combined with first-principles calculations [8,67,70–72] revealed several Fermi surfaces in  $UPt_3$ : two Fermi surfaces at the  $A$  point, the so-called “starfish” and “octopus”; three Fermi surfaces at the  $\Gamma$ -point, the “oyster,” “mussel,” and “pearl”; and also relatively small Fermi surfaces at the  $K$ -points, the “urchins.” The Hamiltonian in Eq. (1) can reproduce the topology of the Fermi surfaces appearing at the  $\Gamma$  point using the parameters [9]  $(t, t_z, t', \alpha, \mu) = (1, 4, 1, 0, 16)$ , see Figs. 2(a) and 2(b). This same model can also reproduce the topology of the “starfish” Fermi surface appearing at the  $A$  point, using a different set of parameters [9]  $(t, t_z, t', \alpha, \mu) = (1, -4, 1, 2, 12)$ , see Figs. 2(c) and 2(d). Therefore, the intersublattice hybridization as described by the Hamiltonian in Eq. (1) is necessarily appreciable at both  $\Gamma$  and  $A$ , while the spin-orbit coupling is only relevant at  $A$  due to its  $\mathbf{k}$  dependence. In what follows, we derive general results without assuming a particular set of values for these parameters and proceed to discuss the implications considering the sets of parameters associated with the Fermi surfaces appearing at  $A$  and  $\Gamma$  separately.

In general, a superconducting order parameter belonging to the  $E_{2u}$  irreducible representation, as widely assumed for  $UPt_3$ , may be written as  $\hat{\Delta}(\mathbf{k}) = \eta_1 \hat{\Gamma}_1(\mathbf{k}) + \eta_2 \hat{\Gamma}_2(\mathbf{k})$  where  $\hat{\Gamma}_i(\mathbf{k})$  are basis functions and  $\eta_i$  are complex numbers parameterizing the phase diagram [2,8,9,67]. Previous analyses using Ginzburg–Landau theory demonstrated that the minimum energy solution may be parameterized by a single real number,  $\eta$ , such that

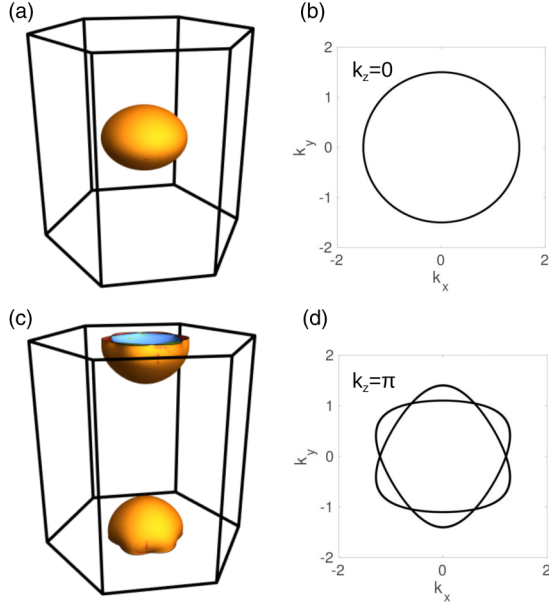


FIG. 2. (a) Fermi surface of  $\text{UPt}_3$  shown within the 3D Brillouin zone (hexagonal frame) plotted using the Hamiltonian in Eq. (1) with parameters  $(t, t_z, t', \alpha, \mu) = (1, 4, 1, 0, 16)$  to reproduce the topology of the Fermi surface near the  $\Gamma$  point. (b) A 2D cross-section of the Fermi surface shown in (a) for  $k_z = 0$ . (c) Same as (a) except using different parameters,  $(t, t_z, t', \alpha, \mu) = (1, -4, 1, 2, 12)$ , to reproduce the topology of the Fermi surface at the  $A$  point. (d) A 2D cross-section of the Fermi surface shown in (c) for  $k_z = \pi$ .

$(\eta_1, \eta_2) = \Delta_0(1, i\eta)/\sqrt{1 + \eta^2}$ , where in the  $A$  phase  $\eta = \infty$ , in the  $B$  phase  $0 < \eta < \infty$ , and in the  $C$  phase  $\eta = 0$  [2,9,67]. Following recent work [8,9,11], we use basis functions that explicitly account for the symmetries of the lattice, which have been shown [9] to give rise to a linear combination of  $p$ -wave,  $d$ -wave, and  $f$ -wave symmetries:

$$\begin{aligned} \hat{\Gamma}_1(\mathbf{k}) &= [\delta\{p_x(\mathbf{k})\sigma^x \otimes \tau^0 - p_y(\mathbf{k})\sigma^y \otimes \tau^0\} \\ &\quad + f_{(x^2-y^2)z}(\mathbf{k})\sigma^z \otimes \tau^x - d_{yz}(\mathbf{k})\sigma^z \otimes \tau^y]i\sigma^y, \\ \hat{\Gamma}_2(\mathbf{k}) &= [\delta\{p_y(\mathbf{k})\sigma^x \otimes \tau^0 + p_x(\mathbf{k})\sigma^y \otimes \tau^0\} \\ &\quad + f_{xyz}(\mathbf{k})\sigma^z \otimes \tau^x - d_{xz}(\mathbf{k})\sigma^z \otimes \tau^y]i\sigma^y, \end{aligned} \quad (2)$$

where  $\delta$  is a small parameter associated with a weak  $p$ -wave component [9], while  $\sigma^i$  and  $\tau^i$  are Pauli matrices in the spin and sublattice spaces, respectively. Notice the unusual combination of spin-triplet  $f$ -wave terms being odd in spatial parity and spin-triplet  $d$ -wave terms being even in parity. This combination is caused by the nonsymmorphic lattice symmetry. Note that these terms still satisfy the constraints imposed by Fermi-Dirac statistics on the Cooper pairs since the  $f$ -wave terms are even in the sublattice index while the  $d$ -wave terms are odd in the sublattice index. From Eq. (2) we can see that, in the absence of the  $p$ -wave component, the order parameter is completely off-diagonal in sublattice space.

To reduce the complexity of the problem we first neglect the  $p$ -wave component by setting  $\delta = 0$ . This term has by far the smallest contribution and as an intrasublattice term we do not expect it to interfere significantly with any potential odd- $\omega$  pairing originating from the intersublattice channels. This

also allows us to make direct contact with previously derived expressions for the Kerr effect in  $\text{UPt}_3$  which also used  $\delta = 0$ . In this case, the Hamiltonian in Eq. (1) breaks down into the following two decoupled sectors [11]

$$\begin{aligned} \mathcal{H} &= \mathcal{H}_a + \mathcal{H}_b \\ &= \frac{1}{2} \sum_{\mathbf{k}} [\Psi_{a,\mathbf{k}}^\dagger \hat{\mathcal{H}}_a(\mathbf{k}) \Psi_{a,\mathbf{k}} + \Psi_{b,\mathbf{k}}^\dagger \hat{\mathcal{H}}_b(\mathbf{k}) \Psi_{b,\mathbf{k}}], \end{aligned} \quad (3)$$

where

$$\hat{\mathcal{H}}_a(\mathbf{k}) = \begin{pmatrix} \xi_{\mathbf{k}} + g_{\mathbf{k}} & \epsilon_{\mathbf{k}} & 0 & \Delta_{12}(\mathbf{k}) \\ \epsilon_{\mathbf{k}}^* & \xi_{\mathbf{k}} - g_{\mathbf{k}} & \Delta_{21}(\mathbf{k}) & 0 \\ 0 & \Delta_{21}^*(\mathbf{k}) & -\xi_{\mathbf{k}} - g_{\mathbf{k}} & -\epsilon_{\mathbf{k}} \\ \Delta_{12}^*(\mathbf{k}) & 0 & -\epsilon_{\mathbf{k}}^* & -\xi_{\mathbf{k}} + g_{\mathbf{k}} \end{pmatrix}, \quad (4)$$

$$\hat{\mathcal{H}}_b(\mathbf{k}) = \begin{pmatrix} \xi_{\mathbf{k}} - g_{\mathbf{k}} & \epsilon_{\mathbf{k}} & 0 & \Delta_{12}(\mathbf{k}) \\ \epsilon_{\mathbf{k}}^* & \xi_{\mathbf{k}} + g_{\mathbf{k}} & \Delta_{21}(\mathbf{k}) & 0 \\ 0 & \Delta_{21}^*(\mathbf{k}) & -\xi_{\mathbf{k}} + g_{\mathbf{k}} & -\epsilon_{\mathbf{k}} \\ \Delta_{12}^*(\mathbf{k}) & 0 & -\epsilon_{\mathbf{k}}^* & -\xi_{\mathbf{k}} - g_{\mathbf{k}} \end{pmatrix} \quad (5)$$

and the gap functions take the form

$$\Delta_{12}(\mathbf{k}) = f_{\mathbf{k}} + id_{\mathbf{k}}, \quad \Delta_{21}(\mathbf{k}) = f_{\mathbf{k}} - id_{\mathbf{k}}, \quad (6)$$

with

$$\begin{aligned} f_{\mathbf{k}} &= \eta_1 f_{(x^2-y^2)z}(\mathbf{k}) + \eta_2 f_{xyz}(\mathbf{k}), \\ d_{\mathbf{k}} &= \eta_1 d_{yz}(\mathbf{k}) + \eta_2 d_{xz}(\mathbf{k}), \end{aligned} \quad (7)$$

and all expressed with the bases defined by

$$\begin{aligned} \Psi_{a,\mathbf{k}}^\dagger &= (c_{\mathbf{k}1\uparrow}^\dagger \ c_{\mathbf{k}2\uparrow}^\dagger \ c_{-\mathbf{k}1\downarrow} \ c_{-\mathbf{k}2\downarrow}), \\ \Psi_{b,\mathbf{k}}^\dagger &= (c_{\mathbf{k}1\downarrow}^\dagger \ c_{\mathbf{k}2\downarrow}^\dagger \ c_{-\mathbf{k}1\uparrow} \ c_{-\mathbf{k}2\uparrow}). \end{aligned} \quad (8)$$

Notice that the two  $4 \times 4$  Hamiltonian matrices are nearly identical, they are related by simply changing the sign of the spin-orbit coupling  $g_{\mathbf{k}}$ . Therefore, without loss of generality, for the remainder of this article, we focus on just  $\mathcal{H}_a$  and note that the results for  $\mathcal{H}_b$  may be obtained by taking  $\alpha \rightarrow -\alpha$ .

### III. PAIR SYMMETRY CLASSIFICATION

We begin our discussion by defining both the normal Green's function  $G$  and the anomalous Green's function  $F$  in terms of the creation and annihilation operators in Eq. (1):

$$\begin{aligned} G_{mm',\sigma\sigma'}(\mathbf{k}; \tau) &= -\langle T_\tau c_{\mathbf{k}m\sigma}(\tau) c_{\mathbf{k}m'\sigma'}^\dagger(0) \rangle, \\ F_{mm',\sigma\sigma'}(\mathbf{k}; \tau) &= -\langle T_\tau c_{\mathbf{k}m\sigma}(\tau) c_{-\mathbf{k}m'\sigma'}(0) \rangle, \end{aligned} \quad (9)$$

where  $\tau$  is the imaginary time and  $T_\tau$  is the usual  $\tau$ -ordering operator for fermions. The pair symmetry of  $\text{UPt}_3$  can be determined by studying the anomalous Green's function  $F$ .

Using the Hamiltonian matrix in Eq. (4) it is straightforward to find the Matsubara representation of the total Green's functions from

$$\hat{\mathcal{G}}_a(\mathbf{k}; i\omega_n) = [i\omega_n - \hat{\mathcal{H}}_a(\mathbf{k})]^{-1}, \quad (10)$$

where  $\omega_n = \pi(2n + 1)/\beta$  is a Matsubara frequency for inverse temperature  $\beta$ . Here  $\hat{\mathcal{G}}_a(\mathbf{k}; i\omega_n)$  is a  $4 \times 4$  matrix with components given by

$$\hat{\mathcal{G}}_a = \begin{pmatrix} G_{11,\uparrow\uparrow} & G_{12,\uparrow\uparrow} & F_{11,\uparrow\downarrow} & F_{12,\uparrow\downarrow} \\ G_{21,\uparrow\uparrow} & G_{22,\uparrow\uparrow} & F_{21,\uparrow\downarrow} & F_{22,\uparrow\downarrow} \\ -F_{11,\downarrow\uparrow}^* & -F_{12,\downarrow\uparrow}^* & -G_{11,\downarrow\downarrow}^* & -G_{12,\downarrow\downarrow}^* \\ -F_{21,\downarrow\uparrow}^* & -F_{22,\downarrow\uparrow}^* & -G_{21,\downarrow\downarrow}^* & -G_{22,\downarrow\downarrow}^* \end{pmatrix}, \quad (11)$$

where we suppressed the dependence on crystal momentum  $\mathbf{k}$  and Matsubara frequency  $\omega_n$  for brevity. Notice that  $\hat{\mathcal{G}}_a$  is comprized of four  $2 \times 2$  blocks in sublattice space, where we can identify the upper off-diagonal block as the anomalous Green's function  $\hat{F}_a \equiv \hat{F}_{\uparrow\downarrow}$ .

To isolate  $\hat{F}_a$ , we start by noting that the Hamiltonian  $\hat{\mathcal{H}}_a$  in Eq. (4) has the form

$$\hat{\mathcal{H}}_a = \begin{pmatrix} \hat{h}_{\mathbf{k}} & \hat{\Delta}(\mathbf{k}) \\ \hat{\Delta}^\dagger(\mathbf{k}) & -\hat{h}_{\mathbf{k}} \end{pmatrix}. \quad (12)$$

Combining Eq. (12) with Eq. (10) it can be shown that the anomalous Green's function associated with  $\hat{\mathcal{H}}_a$  is given by

$$\hat{F}_a(\mathbf{k}; i\omega_n) = [(i\omega_n + \hat{h}_{\mathbf{k}})\hat{\Delta}^{-1}(\mathbf{k})(i\omega_n - \hat{h}_{\mathbf{k}}) - \hat{\Delta}^\dagger(\mathbf{k})]^{-1}. \quad (13)$$

The remainder of this section is dedicated to understanding the symmetries of Eq. (13). However, before we study the general expressions for the full model appearing in Eq. (4) we turn our attention to three limiting cases: (i) no spin-orbit coupling or intersublattice hopping,  $\alpha = t' = 0$ ; (ii) no spin-orbit coupling but finite intersublattice hopping,  $\alpha = 0$ ,  $t' \neq 0$ ; and (iii) finite spin-orbit coupling but no intersublattice hopping,  $\alpha \neq 0$ ,  $t' = 0$ .

#### A. Case (i): $\alpha = t' = 0$

In the limit of no spin-orbit coupling and no intersublattice hopping,  $\alpha = t' = 0$ , all terms proportional to  $g_{\mathbf{k}}$  or  $\epsilon_{\mathbf{k}}$  vanish in  $\hat{\mathcal{H}}_a$ . In this case Eq. (13) takes on a relatively simple form:

$$\hat{F}_a(\mathbf{k}; i\omega_n) = A_{\mathbf{k}; i\omega_n} \begin{pmatrix} 0 & \Delta_{12}(\mathbf{k})[(i\omega_n)^2 - E_{21}(\mathbf{k})^2] \\ \Delta_{21}(\mathbf{k})[(i\omega_n)^2 - E_{12}(\mathbf{k})^2] & 0 \end{pmatrix} \quad (14)$$

where we define

$$A_{\mathbf{k}; i\omega_n}^{-1} = \{(i\omega_n)^4 - (i\omega_n)^2[E_{12}^2(\mathbf{k}) + E_{21}^2(\mathbf{k})] + E_{12}^2(\mathbf{k})E_{21}^2(\mathbf{k})\}, \quad E_{ij} = \sqrt{\xi_{\mathbf{k}}^2 + |\Delta_{ij}(\mathbf{k})|^2}. \quad (15)$$

This trivial case demonstrates that without any mixing of the sublattice or spin degrees of freedom the anomalous Green's function has exactly the same symmetries as the underlying gap functions,  $\Delta_{ij}(\mathbf{k})$ . It is thus clear that there are no odd- $\omega$  pair amplitudes in this case. This result is consistent with the previous results in multiband superconductors showing a required mixing of degrees of freedom for odd- $\omega$  pairing [46].

#### B. Case (ii): $\alpha = 0$ , $t' \neq 0$

In the limit of no spin-orbit coupling but finite intersublattice hopping,  $\alpha = 0$ ,  $t' \neq 0$ , we neglect all terms proportional to  $g_{\mathbf{k}}$  appearing in Eq. (4); however, we must keep track of the intersublattice hopping terms,  $\epsilon_{\mathbf{k}}$ , which are, in general, complex numbers. In this case, the anomalous Green's function becomes

$$\hat{F}_a(\mathbf{k}; i\omega_n) = B_{\mathbf{k}; i\omega_n} \begin{pmatrix} i\omega_n a_{-}(\mathbf{k}) + \xi_{\mathbf{k}} a_{+}(\mathbf{k}) & \Delta_{12}(\mathbf{k})[(i\omega_n)^2 - E_{21}(\mathbf{k})^2] - \Delta_{21}(\mathbf{k})\epsilon_{\mathbf{k}}^2 \\ \Delta_{21}(\mathbf{k})[(i\omega_n)^2 - E_{12}(\mathbf{k})^2] - \Delta_{12}(\mathbf{k})\epsilon_{\mathbf{k}}^{*2} & -i\omega_n a_{-}(\mathbf{k}) + \xi_{\mathbf{k}} a_{+}(\mathbf{k}) \end{pmatrix} \quad (16)$$

where we define

$$B_{\mathbf{k}; i\omega_n}^{-1} = \{(i\omega_n)^4 - (i\omega_n)^2[E_{12}^2(\mathbf{k}) + E_{21}^2(\mathbf{k}) + 2|\epsilon_{\mathbf{k}}|^2] + E_{12}^2(\mathbf{k})E_{21}^2(\mathbf{k}) + |\epsilon_{\mathbf{k}}|^4 - 2\xi_{\mathbf{k}}^2|\epsilon_{\mathbf{k}}|^2 + 2\text{Re}[\Delta_{12}(\mathbf{k})\Delta_{21}^*(\mathbf{k})\epsilon_{\mathbf{k}}^{*2}]\},$$

$$a_{\pm} = \epsilon_{\mathbf{k}}\Delta_{21}(\mathbf{k}) \pm \epsilon_{\mathbf{k}}^*\Delta_{12}(\mathbf{k}), \quad E_{ij} = \sqrt{\xi_{\mathbf{k}}^2 + |\Delta_{ij}(\mathbf{k})|^2}. \quad (17)$$

By inspecting Eqs. (16) and (17) it is straightforward to show that the intersublattice hopping terms generate one odd- $\omega$  pairing channel, given by

$$\hat{F}_{a,\text{odd}}(\mathbf{k}; i\omega_n) = B_{\mathbf{k}; i\omega_n} \begin{pmatrix} 2\omega_n[d_{\mathbf{k}}\text{Re}\{\epsilon_{\mathbf{k}}\} - f_{\mathbf{k}}\text{Im}\{\epsilon_{\mathbf{k}}\}] & 0 \\ 0 & -2\omega_n[d_{\mathbf{k}}\text{Re}\{\epsilon_{\mathbf{k}}\} - f_{\mathbf{k}}\text{Im}\{\epsilon_{\mathbf{k}}\}] \end{pmatrix}. \quad (18)$$

Note that this odd- $\omega$  pair amplitude is proportional to the sublattice hybridization but that it is strictly diagonal in the sublattice index, i.e., it is intrasublattice pairing. Hence, starting from the initial state with only intersublattice pairing, Eq. (4), the addition of intersublattice hopping terms acts to mix the electronic degrees of freedom on the two sublattices allowing for the emergence of a finite intrasublattice pair amplitude. From the form of Eq. (18) we see that, generically, for any finite intersublattice hopping amplitude  $\epsilon_{\mathbf{k}}$  this odd- $\omega$  intrasublattice pair amplitude will be nonzero. Furthermore, we see that, because  $\text{Re}\{\epsilon_{\mathbf{k}}\} = \text{Re}\{\epsilon_{-\mathbf{k}}\}$  and  $\text{Im}\{\epsilon_{\mathbf{k}}\} = -\text{Im}\{\epsilon_{-\mathbf{k}}\}$  the symmetry constraints imposed by Fermi-Dirac statistics are always satisfied since the odd- $\omega$  state is spin-triplet and has even spatial parity.



Notice that, in the absence of the  $d$ -wave component, the two gaps are equal,  $\Delta_{12}(\mathbf{k}) = \Delta_{21}(\mathbf{k})$ . In this case, for purely real values of  $\epsilon_{\mathbf{k}}$  the odd- $\omega$  term vanishes. This is consistent with the expectation that emergent odd- $\omega$  pairing in a multiband superconductor is proportional to the difference between the two gaps [46–49]. However, the addition of an imaginary component to the interband hybridization allows the generation of odd- $\omega$  pairing in multiband systems even when the two gaps are equal. This is distinct from all other forms of odd- $\omega$  pairing that were previously discussed in multiband superconductors [46–49].

### C. Case (iii): $\alpha \neq 0, t' = 0$

In the limit of finite spin-orbit coupling but no intersublattice hopping,  $\alpha \neq 0, t' = 0$ , we neglect all terms proportional to  $\epsilon_{\mathbf{k}}$  appearing in Eq. (4); however, we must keep track of all terms proportional to  $g_{\mathbf{k}}$ , which is a real-valued and odd function of momentum. In this case, the anomalous Green's function becomes

$$\hat{F}_a(\mathbf{k}; i\omega_n) = C_{\mathbf{k}; i\omega_n} \begin{pmatrix} 0 & \Delta_{12}(\mathbf{k})[(i\omega_n + g_{\mathbf{k}})^2 - E_{21}(\mathbf{k})^2] \\ \Delta_{21}(\mathbf{k})[(i\omega_n - g_{\mathbf{k}})^2 - E_{12}(\mathbf{k})^2] & 0 \end{pmatrix} \quad (19)$$

where we define

$$C_{\mathbf{k}; i\omega_n}^{-1} = \{(i\omega_n)^4 - (i\omega_n)^2[E_{12}^2(\mathbf{k}) + E_{21}^2(\mathbf{k}) + 2g_{\mathbf{k}}^2] + E_{12}^2(\mathbf{k})E_{21}^2(\mathbf{k}) + 4g_{\mathbf{k}}\omega_n[d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*] + g_{\mathbf{k}}^2[g_{\mathbf{k}}^2 - E_{12}^2(\mathbf{k}) - E_{21}^2(\mathbf{k})]\},$$

$$E_{ij} = \sqrt{\xi_{\mathbf{k}}^2 + |\Delta_{ij}(\mathbf{k})|^2}. \quad (20)$$

From Eq. (19) we see immediately that the addition of spin-orbit coupling induces an odd- $\omega$  pair amplitude proportional to  $g_{\mathbf{k}}$ . Careful inspection of Eqs. (19) and (20) reveals that the addition of spin-orbit coupling in fact gives rise to two different terms contributing to odd- $\omega$  pairing, one from the numerator

$$\hat{F}_{a,\text{odd}}^{(1)}(\mathbf{k}; i\omega_n) = C_{\mathbf{k}; i\omega_n}^{(1)} \begin{pmatrix} 0 & 2i\omega_n g_{\mathbf{k}}(f_{\mathbf{k}} + id_{\mathbf{k}}) \\ -2i\omega_n g_{\mathbf{k}}(f_{\mathbf{k}} - id_{\mathbf{k}}) & 0 \end{pmatrix}, \quad (21)$$

and one coming from an odd- $\omega$  term in the denominator

$$\hat{F}_{a,\text{odd}}^{(2)}(\mathbf{k}; i\omega_n) = -4g_{\mathbf{k}}\omega_n[d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*]\hat{F}_{a,\text{even}}(\mathbf{k}; i\omega_n), \quad (22)$$

where we define

$$\hat{F}_{a,\text{even}}(\mathbf{k}; i\omega_n) = C_{\mathbf{k}; i\omega_n}^{(2)} \begin{pmatrix} 0 & \Delta_{12}(\mathbf{k})[(i\omega_n)^2 + g_{\mathbf{k}}^2 - E_{21}(\mathbf{k})^2] \\ \Delta_{21}(\mathbf{k})[(i\omega_n)^2 + g_{\mathbf{k}}^2 - E_{12}(\mathbf{k})^2] & 0 \end{pmatrix} \quad (23)$$

where  $C_{\mathbf{k}; i\omega_n}^{(1)}$  and  $C_{\mathbf{k}; i\omega_n}^{(2)}$  are both strictly even functions of the frequency  $\omega_n$  and can thus be ignored in the symmetry analysis. Notice that the spin-orbit coupling cannot modify the pairing in the sublattice index and thus we find that all pairing remains off-diagonal in the sublattice index, just as in case (i).

Recalling that  $g_{\mathbf{k}} = -g_{-\mathbf{k}}$  we can check that the constraints imposed by Fermi-Dirac statistics are satisfied in Eqs. (21) and (22). Inspection of Eq. (21) shows that the  $f$ -wave term is converted from: sublattice-even to sublattice-odd; parity-odd to parity-even; and from even- $\omega$  to odd- $\omega$ . While this appears to violate the statistics we must recall that these results only apply for the  $a$  sector. To obtain the results for the  $b$  sector we take  $g_{\mathbf{k}} \rightarrow -g_{\mathbf{k}}$  and  $\uparrow \leftrightarrow \downarrow$ . Therefore, we see that the odd- $\omega$  terms in Eqs. (21) and (22) are actually spin-singlet and hence Fermi-Dirac statistics are satisfied for both the  $f_{\mathbf{k}}$  and  $d_{\mathbf{k}}$  components.

The first type of odd- $\omega$  pair amplitude  $\hat{F}_{a,\text{odd}}^{(1)}$  has its origin in the numerators appearing in Eq. (19). The emergence of this pair amplitude can be understood by noting that the spin-orbit coupling acts as a momentum-dependent exchange field modifying the spin-symmetry of the superconducting correlations and thereby generating terms in the anomalous Green's function that are odd in frequency. This is similar in spirit to previous analyses of heterostructures incorporating superconductors and materials with different kinds of spin-orbit coupling [41].

The second type of odd- $\omega$  pair amplitude  $\hat{F}_{a,\text{odd}}^{(2)}$  has its origin in the denominator appearing in Eq. (20) and is proportional to  $g_{\mathbf{k}}\omega_n[d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*]$ . While the contribution from the numerator  $\hat{F}_{a,\text{odd}}^{(1)}$  is, in general, nonzero for any finite spin-orbit coupling, the term coming from the denominator  $\hat{F}_{a,\text{odd}}^{(2)}$  requires both a finite spin-orbit coupling and a gap such that  $d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^* \neq 0$ . Considering the form of  $f_{\mathbf{k}}$  and  $d_{\mathbf{k}}$  given in Eq. (7), we conclude that this term is only nonzero in the nonunitary  $B$  phase of UPt<sub>3</sub>, where both  $\eta_1$  and  $\eta_2$  take on finite values.

### D. General case: $\alpha, t' \neq 0$

Finally, we turn our attention to the most general case, both finite spin-orbit coupling and intersublattice hopping  $\alpha, t' \neq 0$ . In this case the anomalous Green's function is given by

$$\hat{F}_a(\mathbf{k}; i\omega_n) = D_{\mathbf{k}; i\omega_n} \begin{pmatrix} i\omega_n a_{-}(\mathbf{k}) + (\xi_{\mathbf{k}} - g_{\mathbf{k}})a_{+}(\mathbf{k}) & \Delta_{12}(\mathbf{k})[(i\omega_n + g_{\mathbf{k}})^2 - E_{21}(\mathbf{k})^2] - \Delta_{21}(\mathbf{k})\epsilon_{\mathbf{k}}^2 \\ \Delta_{21}(\mathbf{k})[(i\omega_n - g_{\mathbf{k}})^2 - E_{12}(\mathbf{k})^2] - \Delta_{12}(\mathbf{k})\epsilon_{\mathbf{k}}^{*2} & -i\omega_n a_{-}(\mathbf{k}) + (\xi_{\mathbf{k}} + g_{\mathbf{k}})a_{+}(\mathbf{k}) \end{pmatrix} \quad (24)$$

where we define

$$D_{\mathbf{k};i\omega_n}^{-1} = \{(i\omega_n)^4 - 2(i\omega_n)^2[\xi_{\mathbf{k}}^2 + |\epsilon_{\mathbf{k}}|^2 + g_{\mathbf{k}}^2 + |f_{\mathbf{k}}|^2 + |d_{\mathbf{k}}|^2] + 4g_{\mathbf{k}}\omega_n[d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*] + \xi_{\mathbf{k}}^4 + |\epsilon_{\mathbf{k}}|^4 + g_{\mathbf{k}}^4 - 2g_{\mathbf{k}}^2\xi_{\mathbf{k}}^2 + 2(\xi_{\mathbf{k}}^2 - g_{\mathbf{k}}^2)[|f_{\mathbf{k}}|^2 + |d_{\mathbf{k}}|^2 - |\epsilon_{\mathbf{k}}|^2] + |\Delta_{12}(\mathbf{k})|^2|\Delta_{21}(\mathbf{k})|^2 + 2\text{Re}[\Delta_{12}(\mathbf{k})\Delta_{21}^*(\mathbf{k})\epsilon_{\mathbf{k}}^{*2}]\},$$

$$a_{\pm} = \epsilon_{\mathbf{k}}\Delta_{21}(\mathbf{k}) \pm \epsilon_{\mathbf{k}}^*\Delta_{12}(\mathbf{k}), \quad E_{ij} = \sqrt{\xi_{\mathbf{k}}^2 + |\Delta_{ij}(\mathbf{k})|^2}. \quad (25)$$

By inspecting Eqs. (24) and (25), once again, we see the emergence of two distinct kinds of odd- $\omega$  pairing amplitudes, one from the numerator

$$\hat{F}_{a,\text{odd}}^{(1)}(\mathbf{k}; i\omega_n) = D_{\mathbf{k};i\omega_n}^{(1)} \begin{pmatrix} 2\omega_n[d_{\mathbf{k}}\text{Re}\{\epsilon_{\mathbf{k}}\} - f_{\mathbf{k}}\text{Im}\{\epsilon_{\mathbf{k}}\}] & 2i\omega_n g_{\mathbf{k}}(f_{\mathbf{k}} + id_{\mathbf{k}}) \\ -2i\omega_n g_{\mathbf{k}}(f_{\mathbf{k}} - id_{\mathbf{k}}) & -2\omega_n[d_{\mathbf{k}}\text{Re}\{\epsilon_{\mathbf{k}}\} - f_{\mathbf{k}}\text{Im}\{\epsilon_{\mathbf{k}}\}] \end{pmatrix}, \quad (26)$$

and one from the denominator

$$\hat{F}_{a,\text{odd}}^{(2)}(\mathbf{k}; i\omega_n) = -4g_{\mathbf{k}}\omega_n[d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*]\hat{F}_{a,\text{even}}(\mathbf{k}; i\omega_n), \quad (27)$$

where we define

$$\hat{F}_{a,\text{even}}(\mathbf{k}; i\omega_n) = D_{\mathbf{k};i\omega_n}^{(2)} \begin{pmatrix} (\xi_{\mathbf{k}} - g_{\mathbf{k}})[\epsilon_{\mathbf{k}}\Delta_{21}(\mathbf{k}) + \epsilon_{\mathbf{k}}^*\Delta_{12}(\mathbf{k})] & \Delta_{12}(\mathbf{k})[(i\omega_n)^2 + g_{\mathbf{k}}^2 - E_{21}(\mathbf{k})^2] - \Delta_{21}(\mathbf{k})\epsilon_{\mathbf{k}}^2 \\ \Delta_{21}(\mathbf{k})[(i\omega_n)^2 + g_{\mathbf{k}}^2 - E_{12}(\mathbf{k})^2] - \Delta_{12}(\mathbf{k})\epsilon_{\mathbf{k}}^{*2} & (\xi_{\mathbf{k}} + g_{\mathbf{k}})[\epsilon_{\mathbf{k}}\Delta_{21}(\mathbf{k}) + \epsilon_{\mathbf{k}}^*\Delta_{12}(\mathbf{k})] \end{pmatrix} \quad (28)$$

where the functions  $D_{\mathbf{k};i\omega_n}^{(1)}$  and  $D_{\mathbf{k};i\omega_n}^{(2)}$  are strictly even functions of frequency  $\omega_n$ . Overall, the relative sizes of these odd- $\omega$  terms depend on the parameters: the spin-orbit coupling term  $g_{\mathbf{k}}$ , the band hybridization  $\epsilon_{\mathbf{k}}$ , and gap symmetry parameterized by  $\eta_1$  and  $\eta_2$ . This general result is a combination of cases (ii) and (iii) with additional terms appearing in  $\hat{F}_{a,\text{even}}$  due to  $g_{\mathbf{k}}$  and  $\epsilon_{\mathbf{k}}$  being nonzero simultaneously.

From Eqs. (26) and (27) we can deduce the general criteria for obtaining finite odd- $\omega$  pair amplitudes in UPT<sub>3</sub>. To obtain finite odd- $\omega$  *intersublattice* pair amplitudes we only need  $g_{\mathbf{k}} \neq 0$ . Additional terms are present when  $d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^* \neq 0$ ; however, these latter terms do not lead to distinct channels in the sublattice index or in spatial parity. To obtain finite odd- $\omega$  *intrasublattice* pair amplitudes only a finite intersublattice hopping term is necessary  $\epsilon_{\mathbf{k}} \neq 0$ . This odd- $\omega$  intrasublattice channel is predominantly  $f$  wave when  $\epsilon_{\mathbf{k}}$  is purely imaginary and  $d$  wave when  $\epsilon_{\mathbf{k}}$  is purely real. It also has additional contributions when both  $d_{\mathbf{k}}f_{\mathbf{k}}^* - f_{\mathbf{k}}d_{\mathbf{k}}^*$  and  $g_{\mathbf{k}}$  are nonzero.

Having derived the general criteria for odd- $\omega$  pairing we finally focus on the specific Fermi surfaces. First we recall that the Fermi surface of UPT<sub>3</sub> around the  $\Gamma$  point is describable with parameters:  $(t, t_z, t', \alpha, \mu) = (1, 4, 1, 0, 16)$ . Therefore, we conclude that near the  $\Gamma$  point in UPT<sub>3</sub> we expect to find odd- $\omega$  pairing proportional to  $\epsilon_{\mathbf{k}}$ , and only in the intrasublattice channel since  $g_{\mathbf{k}} = 0$ . Furthermore, these amplitudes are present in all three phases,  $A$ ,  $B$ , and  $C$  since they appear very generally for any finite values of  $\eta_1, \eta_2$  in Eq. (7). Next, recall the set of parameters describing UPT<sub>3</sub> around the  $A$  point:  $(t, t_z, t', \alpha, \mu) = (1, -4, 1, 2, 12)$ . From this parameter set we see that near the  $A$  point in UPT<sub>3</sub> both the odd- $\omega$  intrasublattice channel and odd- $\omega$  intersublattice channel are finite in all three phases. Furthermore, while both the  $A$  phase and  $C$  phase are unitary according to our model, the  $B$  phase is not. This results in the  $B$  phase receiving additional contributions of the form given in Eq. (27) in both the odd- $\omega$  intrasublattice and odd- $\omega$  intersublattice channels. These additional contributions, originating from a term in the denominator being odd in frequency, are absent in the  $A$  and  $C$  phases.

It should be noted that, while the crystal symmetry of UPT<sub>3</sub> was originally recorded to be the close-packed hexagonal symmetry of space group  $P6_3/mmc$  [73,74] there has been an indication in measurements combining x-ray diffraction and transition electron microscopy that the actual symmetry

may be that of the trigonal space group  $P\bar{3}m1$  [74]. In this case, the lattice will be distorted from the picture appearing in Fig. 1 leading to a layer dimerization that will introduce an asymmetry in the intersublattice hybridization along the  $z$ -axis. In the presence of this asymmetry the intersublattice hopping term  $\epsilon_{\mathbf{k}}$  becomes [10]

$$\tilde{\epsilon}_{\mathbf{k}} = t'[(1+d)e^{i\frac{k_z}{2}} + (1-d)e^{-i\frac{k_z}{2}}] \sum_i e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}_i}$$

$$= 2t' \left[ \cos \frac{k_z}{2} + id \sin \frac{k_z}{2} \right] \sum_i e^{i\mathbf{k}_{\parallel} \cdot \mathbf{r}_i}, \quad (29)$$

where  $d$  parameterizes the magnitude of the layer dimerization. From this expression we note that, just as with  $\epsilon_{\mathbf{k}}$ ,  $\text{Re}\{\tilde{\epsilon}_{\mathbf{k}}\}$  is strictly even in  $\mathbf{k}$ , while  $\text{Im}\{\tilde{\epsilon}_{\mathbf{k}}\}$  is strictly odd in  $\mathbf{k}$ . Hence, while the precise momentum dependence of the odd- $\omega$  pairing will be affected by the crystal distortion described by Eq. (29), the qualitative features discussed above for the odd- $\omega$  pair amplitudes emerging in the absence of the crystal distortion will be preserved even in the presence of such a crystal distortion.

#### IV. ODD-FREQUENCY PAIRING AND THE KERR EFFECT

Now that we have shown how odd- $\omega$  superconductivity is ubiquitous in UPT<sub>3</sub>, we turn to investigating its relationship with the Kerr effect, which has been measured within the  $B$  phase. In general, the frequency-dependent rotation angle for the polarization of reflected light, known as the Kerr angle, is given by

$$\theta(\omega) = \frac{4\pi}{\omega} \text{Im} \left[ \frac{\sigma_H(\omega)}{n(n^2 - 1)} \right], \quad (30)$$

where  $\sigma_H(\omega)$  is the anomalous Hall conductivity and  $n$  is the index of refraction for the material. Motivated by observations of a finite Kerr angle in UPT<sub>3</sub> [7], a recent work [11] computed

the value of Eq. (30) employing the same Hamiltonian as used here, Eq. (1). It was shown that  $\theta(\omega)$  is given by a sum over the Brillouin zone of a quantity proportional to  $[f_{\mathbf{k}}d_{\mathbf{k}}^* - f_{\mathbf{k}}^*d_{\mathbf{k}}]$ , where  $f_{\mathbf{k}}, d_{\mathbf{k}}$  are those given by Eq. (7). Furthermore, it was determined that  $\theta(\omega) = 0$  if the intersublattice hopping function  $\epsilon_{\mathbf{k}}$  is real. Therefore, the criteria for observing a finite Kerr rotation angle in  $\text{UPt}_3$  are given by: (1)  $[f_{\mathbf{k}}d_{\mathbf{k}}^* - f_{\mathbf{k}}^*d_{\mathbf{k}}] \neq 0$ , and (2)  $\text{Im}\{\epsilon_{\mathbf{k}}\} \neq 0$ . Additionally, it was found that a second contribution to the Kerr angle arises when  $g_{\mathbf{k}}$  is finite. Comparing these criteria to the results of the previous sections of this work, we see that, while they differ from our general criteria for the emergence of odd- $\omega$  pairing amplitudes in  $\text{UPt}_3$ , there are still strong similarities.

In particular, notice that to observe the Kerr effect the system must possess finite  $\epsilon_{\mathbf{k}}$  and finite contributions from both  $f_{\mathbf{k}}$  and  $d_{\mathbf{k}}$ . From this alone we can conclude that the system must then also possess odd- $\omega$  intrasublattice pairing of the form appearing in Eq. (18). Additionally, we note that when  $g_{\mathbf{k}} \neq 0$ , the Kerr angle picks up an additional contribution, comparable in magnitude to the contribution independent of spin-orbit coupling [11]. Directly correlated with this additional term is the emergence of a finite odd- $\omega$  intersublattice pairing amplitude with finite  $g_{\mathbf{k}}$  given in Eqs. (21) and (22). Also, since this second term in  $\sigma_H(\omega)$  is proportional to both  $f_{\mathbf{k}}d_{\mathbf{k}}^* - f_{\mathbf{k}}^*d_{\mathbf{k}}$  as well as  $g_{\mathbf{k}}$ , we see from Eq. (27) that novel odd- $\omega$  contributions to both the intrasublattice and intersublattice pairing channels emerge, arising from the denominator of the Green's function.

Therefore, we conclude that the observation of the Kerr effect directly implies the presence of odd- $\omega$  intrasublattice pair amplitudes in  $\text{UPt}_3$ . While this observation alone cannot tell us whether or not odd- $\omega$  intersublattice pair amplitudes also exist in  $\text{UPt}_3$ , the same expressions determining the size of the odd- $\omega$  intersublattice pair amplitudes also provide a contribution to the size of the Kerr rotation angle.

## V. CONCLUSION

In this work we study the emergence of odd- $\omega$  pairing in the heavy-fermion superconductor  $\text{UPt}_3$ . Using a tight-binding model describing the electrons associated with the U ions capturing the nonsymmorphic crystal structure, and assuming an order parameter belonging to  $E_{2u}$ , we characterize the emergence of odd- $\omega$  pair amplitudes and their dependence on the underlying parameters. We find that, in the presence of intersublattice hopping, odd- $\omega$  intrasublattice pairing is present in the system in all three superconducting phases of  $\text{UPt}_3$ , A, B, and C. Similarly, in the presence of spin-orbit coupling we found that odd- $\omega$  intersublattice pairing will be present in all three superconducting phases. Furthermore, we find that in the B phase additional odd- $\omega$  contributions emerge. Considering model parameters which faithfully portray the topology of the Fermi surface near the  $\Gamma$  point and A point, we find that near  $\Gamma$  the model predicts finite odd- $\omega$  intrasublattice pairing, while at A both odd- $\omega$  intrasublattice and odd- $\omega$  intersublattice pairing are finite. Additionally, we compare our criteria for the realization of odd- $\omega$  pairing in  $\text{UPt}_3$  to recent calculations of the size of the Kerr effect using the same model [11] and find very strong similarities. Notably, we show that when the Kerr rotation angle is finite, odd- $\omega$  pair amplitudes are always present. Since the Kerr effect was observed in  $\text{UPt}_3$  [7], this strongly suggests the presence of odd- $\omega$  pairing in this heavy-fermion superconductor.

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- [1] G. R. Stewart, Z. Fisk, J. O. Willis, and J. L. Smith, *Phys. Rev. Lett.* **52**, 679 (1984).
  - [2] J. Sauls, *Adv. Phys.* **43**, 113 (1994).
  - [3] S. Adenwalla, S. W. Lin, Q. Z. Ran, Z. Zhao, J. B. Ketterson, J. A. Sauls, L. Taillefer, D. G. Hinks, M. Levy, and B. K. Sarma, *Phys. Rev. Lett.* **65**, 2298 (1990).
  - [4] H. Tou, Y. Kitaoka, K. Asayama, N. Kimura, Y. Ōnuki, E. Yamamoto, and K. Maezawa, *Phys. Rev. Lett.* **77**, 1374 (1996).
  - [5] J. D. Strand, D. J. Bahr, D. J. Van Harlingen, J. P. Davis, W. J. Gannon, and W. P. Halperin, *Science* **328**, 1368 (2010).
  - [6] J. D. Strand, D. J. Van Harlingen, J. B. Kycia, and W. P. Halperin, *Phys. Rev. Lett.* **103**, 197002 (2009).
  - [7] E. R. Schemm, W. J. Gannon, C. M. Wishne, W. P. Halperin, and A. Kapitulnik, *Science* **345**, 190 (2014).
  - [8] T. Nomoto and H. Ikeda, *Phys. Rev. Lett.* **117**, 217002 (2016).
  - [9] Y. Yanase, *Phys. Rev. B* **94**, 174502 (2016).
  - [10] Y. Yanase and K. Shiozaki, *Phys. Rev. B* **95**, 224514 (2017).
  - [11] Z. Wang, J. Berlinsky, G. Zwiczak, and C. Kallin, *Phys. Rev. B* **96**, 174511 (2017).
  - [12] V. L. Berezinskii, *Pis'ma Zh. Eksp. Teor. Fiz.* **20**, 628 (1974).
  - [13] T. R. Kirkpatrick and D. Belitz, *Phys. Rev. Lett.* **66**, 1533 (1991).
  - [14] D. Belitz and T. R. Kirkpatrick, *Phys. Rev. B* **46**, 8393 (1992).
  - [15] A. Balatsky and E. Abrahams, *Phys. Rev. B* **45**, 13125 (1992).
  - [16] P. Coleman, E. Miranda, and A. Tsvelik, *Phys. Rev. Lett.* **70**, 2960 (1993).
  - [17] P. Coleman, E. Miranda, and A. Tsvelik, *Phys. Rev. B* **49**, 8955 (1994).
  - [18] P. Coleman, E. Miranda, and A. Tsvelik, *Phys. Rev. Lett.* **74**, 1653 (1995).
  - [19] R. Heid, *Z. Phys. B* **99**, 15 (1995).
  - [20] D. Belitz and T. R. Kirkpatrick, *Phys. Rev. B* **60**, 3485 (1999).
  - [21] D. Solenov, I. Martin, and D. Mozysky, *Phys. Rev. B* **79**, 132502 (2009).
  - [22] H. Kusunose, Y. Fuseya, and K. Miyake, *J. Phys. Soc. Jpn* **80**, 054702 (2011).
  - [23] Y. V. Fominov, Y. Tanaka, Y. Asano, and M. Eschrig, *Phys. Rev. B* **91**, 144514 (2015).
  - [24] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, *Phys. Rev. Lett.* **86**, 4096 (2001).
  - [25] F. S. Bergeret, A. F. Volkov, and K. B. Efetov, *Rev. Mod. Phys.* **77**, 1321 (2005).

- [26] T. Yokoyama, Y. Tanaka, and A. A. Golubov, *Phys. Rev. B* **75**, 134510 (2007).
- [27] M. Houzet, *Phys. Rev. Lett.* **101**, 057009 (2008).
- [28] M. Eschrig and T. Löfwander, *Nat. Phys.* **4**, 138 (2008).
- [29] J. Linder, T. Yokoyama, and A. Sudbø, *Phys. Rev. B* **77**, 174514 (2008).
- [30] F. Crépin, P. Bursset, and B. Trauzettel, *Phys. Rev. B* **92**, 100507 (2015).
- [31] T. Yokoyama, *Phys. Rev. B* **86**, 075410 (2012).
- [32] A. M. Black-Schaffer and A. V. Balatsky, *Phys. Rev. B* **86**, 144506 (2012).
- [33] A. M. Black-Schaffer and A. V. Balatsky, *Phys. Rev. B* **87**, 220506 (2013).
- [34] C. Triola, E. Rossi, and A. V. Balatsky, *Phys. Rev. B* **89**, 165309 (2014).
- [35] Y. Tanaka and A. A. Golubov, *Phys. Rev. Lett.* **98**, 037003 (2007).
- [36] Y. Tanaka, Y. Tanuma, and A. A. Golubov, *Phys. Rev. B* **76**, 054522 (2007).
- [37] J. Linder, T. Yokoyama, A. Sudbø, and M. Eschrig, *Phys. Rev. Lett.* **102**, 107008 (2009).
- [38] J. Linder, A. Sudbø, T. Yokoyama, R. Grein, and M. Eschrig, *Phys. Rev. B* **81**, 214504 (2010).
- [39] Y. Tanaka, M. Sato, and N. Nagaosa, *J. Phys. Soc. Jpn* **81**, 011013 (2012).
- [40] F. Parhizgar and A. M. Black-Schaffer, *Phys. Rev. B* **90**, 184517 (2014).
- [41] C. Triola, D. M. Badiane, A. V. Balatsky, and E. Rossi, *Phys. Rev. Lett.* **116**, 257001 (2016).
- [42] C. Triola and A. V. Balatsky, *Phys. Rev. B* **94**, 094518 (2016).
- [43] A. Di Bernardo, S. Diesch, Y. Gu, J. Linder, G. Divitini, C. Ducati, E. Scheer, M. G. Blamire, and J. W. Robinson, *Nat. Commun.* **6**, 8053 (2015).
- [44] A. Di Bernardo, Z. Salman, X. L. Wang, M. Amado, M. Egilmez, M. G. Flokstra, A. Suter, S. L. Lee, J. H. Zhao, T. Prokscha, E. Morenzoni, M.G. Blamire, J. Linder, and J. W. A. Robinson, *Phys. Rev. X* **5**, 041021 (2015).
- [45] J. Linder and A. V. Balatsky, [arXiv:1709.03986](https://arxiv.org/abs/1709.03986).
- [46] A. M. Black-Schaffer and A. V. Balatsky, *Phys. Rev. B* **88**, 104514 (2013).
- [47] L. Komendová, A. V. Balatsky, and A. M. Black-Schaffer, *Phys. Rev. B* **92**, 094517 (2015).
- [48] L. Komendová and A. M. Black-Schaffer, *Phys. Rev. Lett.* **119**, 087001 (2017).
- [49] C. Triola and A. V. Balatsky, *Phys. Rev. B* **95**, 224518 (2017).
- [50] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. Bednorz, and F. Lichtenberg, *Nature* **372**, 532 (1994).
- [51] Y. Maeno, S. Kittaka, T. Nomura, S. Yonezawa, and K. Ishida, *J. Phys. Soc. Jpn* **81**, 011009 (2012).
- [52] F. Hunte, J. Jaroszynski, A. Gurevich, D. Larbalestier, R. Jin, A. Sefat, M. A. McGuire, B. C. Sales, D. K. Christen, and D. Mandrus, *Nature* **453**, 903 (2008).
- [53] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *J. Am. Chem. Soc.* **130**, 3296 (2008).
- [54] K. Ishida, Y. Nakai, and H. Hosono, *J. Phys. Soc. Jpn* **78**, 062001 (2009).
- [55] V. Cvetkovic and Z. Tesanovic, *Euro. Phys. Lett.* **85**, 37002 (2009).
- [56] G. Stewart, *Rev. Mod. Phys.* **83**, 1589 (2011).
- [57] J. Nagamatsu, N. Nakagawa, T. Muranaka, Y. Zenitani, and J. Akimitsu, *Nature* **410**, 63 (2001).
- [58] F. Bouquet, R. A. Fisher, N. E. Phillips, D. G. Hinks, and J. D. Jorgensen, *Phys. Rev. Lett.* **87**, 047001 (2001).
- [59] A. Brinkman, A. A. Golubov, H. Rogalla, O. V. Dolgov, J. Kortus, Y. Kong, O. Jepsen, and O. K. Andersen, *Phys. Rev. B* **65**, 180517 (2002).
- [60] A. Golubov, J. Kortus, O. Dolgov, O. Jepsen, Y. Kong, O. Andersen, B. Gibson, K. Ahn, and R. Kremer, *J. Phys.: Condens. Matter* **14**, 1353 (2002).
- [61] M. Iavarone, G. Karapetrov, A. Koshelev, W. Kwok, G. Crabtree, D. Hinks, W. Kang, E.-M. Choi, H. J. Kim, H.-J. Kim, and S. I. Lee, *Phys. Rev. Lett.* **89**, 187002 (2002).
- [62] J. Xia, Y. Maeno, P. T. Beyersdorf, M. M. Fejer, and A. Kapitulnik, *Phys. Rev. Lett.* **97**, 167002 (2006).
- [63] K. I. Wysokiński, J. F. Annett, and B. L. Györfy, *Phys. Rev. Lett.* **108**, 077004 (2012).
- [64] E. Taylor and C. Kallin, *Phys. Rev. Lett.* **108**, 157001 (2012).
- [65] M. Gradhand, K. I. Wysokiński, J. F. Annett, and B. L. Györfy, *Phys. Rev. B* **88**, 094504 (2013).
- [66] R. M. Lutchyn, P. Nagornykh, and V. M. Yakovenko, *Phys. Rev. B* **80**, 104508 (2009).
- [67] R. Joynt and L. Taillefer, *Rev. Mod. Phys.* **74**, 235 (2002).
- [68] L. Taillefer, R. Newbury, G. Lonzarich, Z. Fisk, and J. Smith, *J. Magn. Magn. Mater.* **63-64**, 372 (1987).
- [69] L. Taillefer and G. G. Lonzarich, *Phys. Rev. Lett.* **60**, 1570 (1988).
- [70] C. S. Wang, M. R. Norman, R. C. Albers, A. M. Boring, W. E. Pickett, H. Krakauer, and N. E. Christensen, *Phys. Rev. B* **35**, 7260 (1987).
- [71] M. Norman, R. Albers, A. Boring, and N. Christensen, *Solid State Commun.* **68**, 245 (1988).
- [72] G. McMullan, M. Norman, A. Huxley, N. Doiron-Leyraud, J. Flouquet, G. Lonzarich, A. McCollam, and S. Julian, *New J. Phys.* **10**, 053029 (2008).
- [73] T. J. Heal and G. I. Williams, *Acta Crystallogr.* **8**, 494 (1955).
- [74] D. A. Walko, J.-I. Hong, T. V. Chandrasekhar Rao, Z. Wawrzak, D. N. Seidman, W. P. Halperin, and M. J. Bedzyk, *Phys. Rev. B* **63**, 054522 (2001).