Umklapp scattering in unconventional superconductors: Microwave conductivity shows that κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br is a d_{xy} superconductor

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Microwave conductivity experiments can directly measure the quasiparticle scattering rate in the superconducting state. We show that this, combined with knowledge of the Fermi surface geometry, can distinguish between closely related superconducting order parameters, e.g., $d_{x^2-y^2}$ and d_{xy} superconductivity. We benchmark this method on $YBa_2Cu_3O_{7-\delta}$ and, unsurprisingly, confirm that this is a $d_{x^2-y^2}$ superconductor. We then apply our method to κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, which we discover is a d_{xy} superconductor.

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In many unconventional superconductors, the tunneling experiments that definitively identified the superconducting gap symmetries in cuprate superconductors [1,2] are prohibitively difficult to perform. This presents a significant difficulty in distinguishing the form of the order parameter and therefore in understanding the microscopic origin of superconductivity, which cannot be probed directly in macroscopic experiments. Conventional probes of the superconducting gap rely on the use of extremely low temperature measurements, where the temperature dependence can be used to identify the low energy density of states [3–5]. This then gives insight into the nature of low energy excitations in the superconducting system.

While such methods are useful in distinguishing between nodeless "*s*-wave" superconductivity, gaps with line nodes, and those with point nodes, they are unable to resolve the exact form of the superconducting gap. For example, the temperature dependence of the low temperature heat capacity may identify the presence of line nodes, but in order to determine the location of such nodes on the Fermi surface, more complicated directional probes have been necessary [6,7]. Such experiments with directional resolution are difficult to perform and interpret, which motivates one to discover new probes or learn how to gain more information from existing experiments.

The temperature dependence of the penetration depth is often measured via microwave conductivity experiments and has long been used as a probe of unconventional superconductors. The exponential suppression of low energy quasiparticles in conventional (nodeless) superconductors is evident in an activated exponential temperature dependence of the penetration depth. In contrast, the penetration depth in unconventional (nodal) superconductors exhibit a power-law (often linear) temperature dependence at low temperatures [8–11].

In this article we propose a richer use of the microwave conductivity, as a more detailed probe of the superconducting gap structure. This relies on the ability of the microwave conductivity to accurately determine the relaxation rate of the superconducting quasiparticles, responsible for the screening of the Meissner effect. We show that, in conjunction with the knowledge of the Fermi surface geometry in the normal state, measurements of the quasiparticle relaxation rate via the penetration depth can be used to differentiate between closely related order parameters, e.g., $d_{x^2-y^2}$ and d_{xy}.

To make these ideas concrete, we focus on two families of unconventional superconductor: the high-temperature cuprate superconductors and organic superconductors. The $d_{x^2-y^2}$ pairing symmetry of $YBa_2Cu_3O_{7-\delta}$ (YBCO) is long established [1,2,12]. At low temperatures the quasiparticle scattering rate varies exponentially with temperature [13], which results from umklapp scattering becoming an activated process in the $d_{x^2-y^2}$ superconducting state [14,15]. Here we show that the experimentally observed penetration depth is incompatible with d_{xy} pairing, as this would allow umklapp scattering at arbitrarily low energies. The conclusion that the pairing symmetry is $d_{x^2-y^2}$ in YBCO is not new, but demonstrates the potential additional information available in microwave conductivity experiments.

Despite being one of the most studied organic superconductors, the pairing symmetry of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (κ-Br) remains contested [7,16–34].

Carrington *et al.* [35] reported that the penetration depth in κ-Br has a $T^{3/2}$ power-law temperature dependence at low temperatures, which was interpreted as indicative of nodal superconductivity, but is not expected for any known nodal structure. Importantly, this interpretation of this experiment requires knowledge of the absolute value of in-plane superfluid density, which Carrington *et al.* could not measure. Recently Milbradt *et al.* [36] measured the absolute value from a normal state matching technique and found that is was an order of magnitude larger than estimated by Carrington *et al.*, which dramatically changes this result although otherwise the data are remarkably consistent. Using the measured absolute value of in-plane penetration depth, Milbradt *et al.* found that

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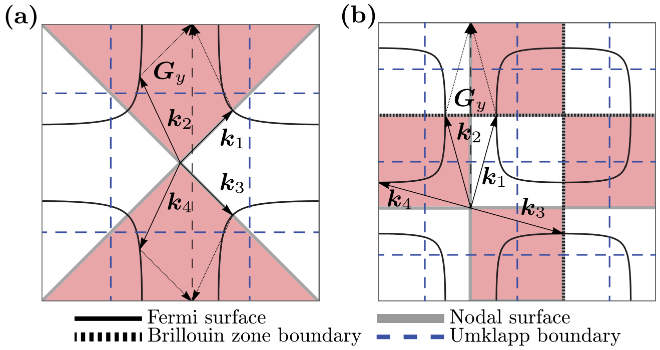

FIG. 1. Typical umklapp scattering process involving nodal quasiparticles in YBCO, assuming (a) $d_{x^2-y^2}$ pairing and (b) d_{xy} pairing, in this panel we plot momenta outside of the FBZ for clarity. In (a) umklapp scattering necessarily involves particles away from the nodes, leading to an exponentially activated umklapp scattering rate. In (b) umklapp scattering is possible involving only nodal quasiparticles, leading to $\tau_u^{-1} \propto T^3$. The activated temperature dependence of the relaxation rate is therefore sufficient to distinguish between these two gap symmetries, independent of other experiments. Dashed arrows indicate the reciprocal lattice vectors, dotted arrows are guides to the eye to show the total momentum before and after the scattering process. The Fermi surface is calculated from the single band model for YBCO [15], and the shading denotes the sign of the order parameter.

the superfluid density varies linearly with temperature, consistent with point nodes on a two-dimensional Fermi surface. Given the dramatic effect of the correct calibration of these experiments, we will focus on Milbradt *et al.*'s data below.

The accurate determination of the penetration depth also allowed high resolution measurements of the quasiparticle relaxation rate in κ-Br, which show a cubic temperature dependence, as opposed to the activated exponential seen in YBCO [36]. This shows that umklapp scattering occurs at arbitrarily low energies in the superconducting state of κ-Br. We show that the only order parameter, of those discussed for κ-Br, consistent with gapless umklapp scattering, is d_{xy} symmetry.

The current is proportional to the total momentum carried by the electronic quasiparticles and therefore cannot be relaxed by elastic processes. Current relaxation due to elastic electron-electron interactions requires the presence of some mechanism for the loss of momentum. The most significant mechanism for such relaxation is umklapp scattering. Where the initial ($\boldsymbol{k}_1, \boldsymbol{k}_2$) and final ($\boldsymbol{k}_3, \boldsymbol{k}_4$) momentum states satisfy

$$\boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_3 - \boldsymbol{k}_4 = \pm \boldsymbol{G}_j, \qquad (1)$$

and $\boldsymbol{G}_j$ is a reciprocal lattice vector. Umklapp scattering transfers momentum to the lattice allowing the current to relax due entirely to elastic electron-electron scattering [14,15,37–42]. For a simple Fermi liquid, with a circular Fermi surface, umklapp scattering requires $k_F \geqslant |\boldsymbol{G}_j/4|$. For nontrivial band structures, it is natural to define an "umklapp boundary" of $|\boldsymbol{k}_j| \geqslant \boldsymbol{G}_j/4$ in the first Brillouin zone (FBZ), see Fig. 1. Umklapp scattering can occur between states on different sides of the umklapp boundary [14,40].

In a normal metal, as long as the umklapp condition Eq. (1) is satisfied for some points on the Fermi surface, the current can relax entirely due to electron-electron scattering, yielding the well-known quadratic temperature dependence of the resistance [37]. The scattering rate due to umklapp processes, which relax the current, and the total electron-electron scattering rate, including elastic processes, differ only by an overall factor, due to the reduced phase space available for umklapp processes [37,40]. For a superconductor with a nodal gap function, however, the structure of the superconducting gap reduces the phase space available for umklapp scattering. The total scattering rate and current relaxation rate can therefore differ more dramatically than in a normal metal.

Walker and Smith [14] first addressed this possibility theoretically, after experiments on YBCO found a current relaxation rate with an activated exponential temperature dependence, rather than the cubic dependence found for the total scattering rate [13]. The central argument of their theory is that, because the nodes of the gap on the Fermi surface in YBCO do not satisfy the umklapp condition [Eq. (1)], any umklapp scattering process must necessarily involve quasiparticles away from the nodes. They showed geometrically that in YBCO with $d_{x^2-y^2}$ superconductivity, no umklapp process is possible involving only nodal quasiparticles [see Fig. 1(a)]. This necessarily leads to a relaxation rate for umklapp processes involving two nodal quasiparticles of $\tau_u^{-1} \propto T^2 f(\Delta_U)\bar{f}(\Delta_U) \propto T^2 \exp(-\Delta_U/k_B T)$ where the umklapp Δ_U is the minimum energy of the states to which non-nodal quasiparticles can umklapp scatter. More generally, we find that Δ_U is the minimum energy of any four states that satisfy Eq. (1). The total scattering rate, in contrast, can be found from power counting of scattering processes involving the nodal quasiparticles to be $\tau^{-1} \propto T^3$. Both scattering rates were later reproduced by numerical calculations based on the random phase approximation (RPA) [15].

The existence of these two distinct temperature dependencies for the total and umklapp scattering rates is in general expected in nodal superconductors, with the energy scale Δ_U set by the geometry of the gap function and the nodal placement. In this article we identify a crucial exception to this rule: if the nodes exactly satisfy the umklapp condition [Eq. (1)], then the umklapp scattering rate will be dominated by the contribution due to the low energy nodal quasiparticles, and will vary cubically, rather than exponentially, with temperature. Crucially, we show that a nodal placement satisfying the umklapp condition is *not* an exotic occurrence requiring fine tuning, rather, this is required for certain combinations of pairing symmetry and Fermi surface geometry. A key example below will be d_{xy} pairing on an open Fermi surface (that crosses the boundary of the FBZ), which allows umklapp scattering between quasiparticles exactly at the nodes. However, this is certainly not the only possible route to quasiparticle scattering without an umklapp gap, other possibilities will briefly be discussed toward the end of this article.

As a demonstrative example, we first consider an alternative d_{xy} state in YBCO. In Fig. 1 we sketch the possible momentum configurations for umklapp scattering involving nodal quasiparticles in a realistic model of YBCO for the two d-wave gaps. For a $d_{x^2-y^2}$ gap, an umklapp scattering process involving quasiparticles at the nodes, inside the umklapp boundary, must also involve states outside the boundary and away from the nodes [Fig. 1(a)], leading to an exponential temperature dependence. In the d_{xy} case, however [Fig. 1(b)], there exists an electron configuration for which the quasiparticles at the nodes (on the FBZ boundary) contribute to the umklapp scattering, with no umklapp gap, giving a cubic temperature dependence. This example suggests the possibility of using such measurements as a more direct probe of the detailed form of the superconducting gap than has been previously considered. Any insight gained from these measurements, however, requires a detailed understanding of the underlying normal state Fermi surface.

κ-Br provides an important opportunity to use the quasiparticle scattering rate as a probe of superconducting gap symmetry. The normal state properties have been studied in great detail [31,43–49], and the quasiparticle relaxation rate has been measured: it varies cubically with temperature [36].

Both YBCO and κ-Br have D_{2h} point group symmetries. In YBCO this is due to a small orthorhombic distortion, which does not change the analysis above in any significant way. In κ-Br the lattice is far from tetragonal and this has important consequences for the analysis of the superconductivity [30] and the quasiparticle scattering rate. Both YBCO and κ-Br form a layered structure: the layers lie in the a-b plane in YBCO and the a-c plane in κ-Br. We will adopt a labeling convention where the x and y axes are considered parallel to the a and c directions, respectively. However, one should note that, particularly in the theoretical literature, superconducting order parameters are often defined in coordinated systems rotated 45° from the crystal axes in κ-Br. In this basis, the d_{xy} and $d_{x^2-y^2}$ labels are reversed. Unlike YBCO, the Fermi surface of κ-Br is strongly anisotropic, and as such the umklapp scattering along each of the two (a and c) crystal axes must be considered independently, as shown in Fig. 2.

Three different gap symmetries have been proposed in κ-Br: a full gapped s-wave order parameter [18]; d_{xy} pairing [Fig. 2(a)], where both the presence and the placement of nodes are required by symmetry [24,25,29,30,50]; and a $d_{x^2-y^2}+s$ order parameter [22,31–33,51] [Fig. 2(b)]. In the D_{2h} point group both the $d_{x^2-y^2}$ and s pairing channels transform according to the trivial (A_{1g}) irreducible representation, therefore any nodes are formally accidental, but can arise provided the $d_{x^2-y^2}$ component is larger than the s-wave component.

An s-wave order parameter necessarily leads to an activated quasiparticle scattering rate, which is inconsistent with the microwave conductivity measurements in κ-Br. We display possible umklapp scattering processes, in both the a and c directions, involving nodal quasiparticles for both d_{xy} and $d_{x^2-y^2}+s$ in Fig. 2. It is clear that umklapp scattering processes involving only nodal quasiparticles exist for a d_{xy} pairing [Fig. 2(b)], for transport in the c direction, but not in the a direction [i.e., only for $\boldsymbol{G}_j = \boldsymbol{G}_c$ in Eq. (1)]. Nevertheless, at low temperatures contribution to quasiparticle scattering in the c direction shorts out the contribution in the a direction, leading to a cubic temperature dependence expected for d_{xy} pairing, consistent with experiment [36]. In contrast for a $d_{x^2-y^2}+s$ gap there is no set of nodes that satisfies the umklapp condition [Eq. (1)], Fig. 2(a). Thus, for $d_{x^2-y^2}+s$ pairing one expects the quasiparticle scattering rate to be exponentially activated—in clear contradiction to experiment [36].

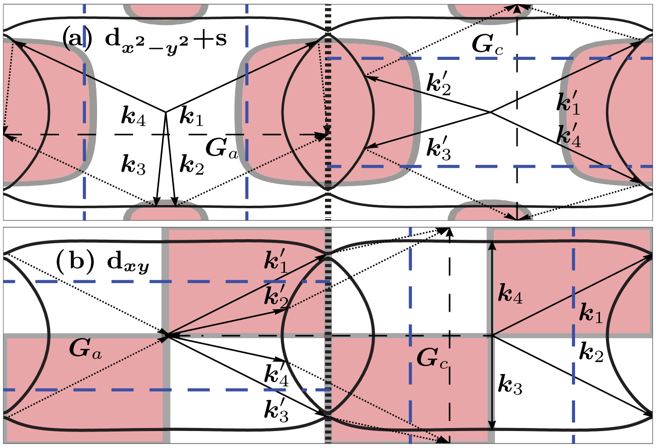

FIG. 2. Momentum configurations for umklapp processes involving nodal quasiparticles in κ-Br for (a) $d_{x^2-y^2}+s$ pairing and (b) d_{xy} pairing. For $d_{x^2-y^2}+s$ pairing umklapp scattering must involve particles away from the nodes, leading to an exponentially activated umklapp scattering rate. For d_{xy} pairing umklapp scattering involving only nodal quasiparticles is possible in the c direction, but not the a direction. Therefore at low temperatures umklapp scattering in the c direction dominates leading to $\tau_u^{-1} \propto T^3$. The activated temperature dependence of the relaxation rate is therefore sufficient to distinguish between these two gap symmetries, independent of other experiments. The gap in (b) uses the parametrization of Guterding *et al.* [31,32]. The Fermi surface is calculated from the "monomer" model of κ-Br [48], and the shading denotes the sign of the order parameter. Only the umklapp boundary relevant to scattering in a given direction is shown.

This analysis strongly suggests that the superconductivity in κ-Br occurs in the d_{xy} channel. But a little care is required. The temperature dependencies discussed so far apply only as $T \to 0$. Therefore, before reaching a firm conclusion, one needs to understand how large Δ_U is and hence how the quasiparticle scattering behaves at higher temperatures. To investigate this we numerically calculated the Fermi golden rule scattering rate,

$$\tau^{-1}(\boldsymbol{k}_1) = \sum_{\boldsymbol{k}_2,\boldsymbol{k}_3,\boldsymbol{k}_4,j} |\tilde{V}_{\{\boldsymbol{k}_i\}}|^2 f(E_{\boldsymbol{k}_2}) \bar{f}(E_{\boldsymbol{k}_3}) \bar{f}(E_{\boldsymbol{k}_4}) \times \delta(E_{\boldsymbol{k}_1} + E_{\boldsymbol{k}_2} - E_{\boldsymbol{k}_3} - E_{\boldsymbol{k}_4}) \times \delta(\boldsymbol{k}_1 + \boldsymbol{k}_2 - \boldsymbol{k}_3 - \boldsymbol{k}_4 + n_j \boldsymbol{G}_j), \quad (2)$$

where $E_{\boldsymbol{k}} = \sqrt{\xi_{\boldsymbol{k}}^2 + \Delta_{\boldsymbol{k}}^2}$ is the quasiparticle energy, defined in terms of the electron dispersion $\xi_{\boldsymbol{k}}$, and the superconducting gap $\Delta_{\boldsymbol{k}}$, $f(E)$ is the Fermi-Dirac distribution function [$\bar{f}(E) = 1 - f(E)$], the scattering potential $\tilde{V}_{\{\boldsymbol{k}_i\}}$ takes quasiparticle coherence factors into account, and $n_j = 1$ ($n_j = 0$) for umklapp (normal) scattering in the j direction. We take the scattering potential potential to be of the RPA form, which has been shown to accurately reproduce the measured quasiparticle scattering rate in YBCO [15]. The temperature dependence of the superconducting gap is assumed to follow a strong-coupling BCS form: $\Delta(T) = (5k_B T_c/2)\tanh[3\sqrt{(T_c/T) - 1}]$, with the parameters governing the overall magnitude and temperature dependence determined from experiment [36].

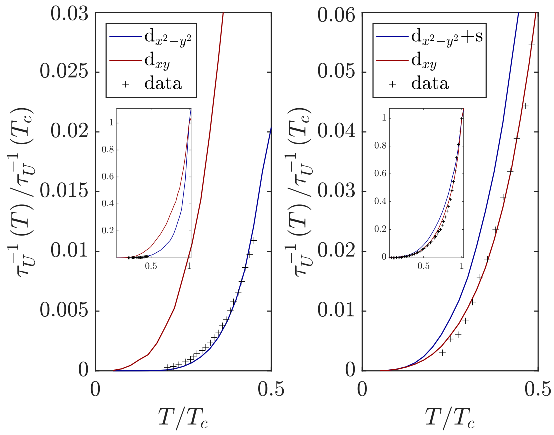

FIG. 3. Comparison of the calculated quasiparticle relaxation rate for the different order parameters to experimental data for (a) YBCO [13] and (b) κ-Br [36]. In both cases the experimental data are described accurately by only one of the order parameters: $d_{x^2-y^2}$ for YBCO and d_{xy} for κ-Br. A temperature-independent constant, resulting from residual impurity scattering, has been subtracted from each set of experimental data, equal to 5.0×10^{10} s^{-1} in (a) and 4.7×10^{10} s^{-1} in (b). In both cases, the constant term is on the order of $\lesssim 0.01\tau_U^{-1}(T_c)$.

For concreteness we consider only the simplest d_{xy} order parameter: $\Delta_{\boldsymbol{k}} \propto \sin(k_a)\sin(k_c)$, where k_a and k_c are the crystal momentum components along the a and c crystal axes. $d_{x^2-y^2}+s$ order parameters have an additional freedom, the degree of mixing of the $d_{x^2-y^2}$ and s components. To avoid treating this as a free parameter, we take the recent parametrization from fits to scanning tunneling spectra, which also includes an extended-s component: $\Delta_{\boldsymbol{k}} \propto c_{s_1}[\cos(k_a)+\cos(k_c)] + c_{s_2}\cos(k_a)\cos(k_c) + c_d[\cos(k_a)-\cos(k_c)]$ [32].

In order to efficiently evaluate the integrals required to calculate the relaxation rate involving low energy quasiparticles, the quasiparticle energy E_k was first calculated on a two-dimensional grid with 2×10^5 sites per dimension. For each temperature a Monte Carlo approach was used to select a subset of these points. We calculated $\partial f(\omega)/\partial\omega$ which is peaked at low energies with a temperature-dependent width, and retain only those points for which an appropriately normalized random number was less than $\partial f(\omega)/\partial\omega$. The resulting adaptive mesh was then used to perform the integrals required to calculate the scattering rate as described in [15]. All calculations have been performed using $\omega = 0.005t$ and broadening the δ functions to Lorentzians of width $0.0005t$. Our results vary only weakly with the bare interaction strength U, and we report here only the results in the weakly interacting limit.

We compare the calculated umklapp scattering rates for each pairing symmetry with the Fermi surfaces for YBCO and κ-Br to the relevant experimental data [13,36] in Fig. 3. In YBCO the temperature dependence of the relaxation rate strongly differentiates between the two gap functions, with a clear cubic temperature dependence seen for the d_{xy} gap, while the $d_{x^2-y^2}$ gap shows the exponential temperature dependence observed experimentally. Thus, the uncontroversial conclusion that this is a $d_{x^2-y^2}$ superconductor follows safely from this experiment alone. For κ-Br the Δ_U for $d_{x^2-y^2}+s$ pairing is somewhat smaller than the umklapp gap in YBCO. Therefore, the results are less distinct at higher temperatures, but as $T \to 0$ it is clear that the cubic temperature dependence arising from the d_{xy} gap is in much better agreement with experiment than the exponential temperature dependence from $d_{x^2-y^2}+s$ pairing. This is compelling evidence that κ-Br is a d_{xy} superconductor.

An important distinction should be made between the $d_{x^2-y^2}$ gap in YBCO and the $d_{x^2-y^2}+s$ gap in κ-Br. In both cases, the umklapp processes are exponentially suppressed, but in the latter case the predicted scattering rate exceeds the T^3 rate for the d_{xy} gap at experimentally relevant temperatures. This is due in part to the significantly smaller Δ_U for the $d_{x^2-y^2}+$s gap in κ-Br. But, more importantly, the nodes are accidental in κ-Br, rather than symmetry required as in YBCO gaps. In this case, the variation of the gap in the direction perpendicular to the Fermi surface is nonzero, resulting in a scattering rate that varies as $T^3\exp(-\Delta_U/k_BT)$, rather than $T^2\exp(-\Delta_U/k_BT)$. Finally, a large nonuniversal prefactor also enhances scattering in the $d_{x^2-y^2}+s$ gap in κ-Br.

We must also note a technical loophole in the above argument. For a superconducting gap with accidental nodes, there is no restriction on the nodal placement on the Fermi surface. It is possible to fine tune the gap $d_{x^2-y^2}+s$ case to give a nodal placement satisfying Eq. (1). We find that the best fit to the experimental scattering rate data is given if the s-wave component is negligibly small. For a pure $d_{x^2-y^2}$ gap ($\Delta_{\mathbf{k}} \propto \cos k_a - \cos k_c$) the calculated umklapp scattering rate is numerically indistinguishable from the d_{xy} case. However, such a gap is theoretically extremely unlikely as, if allowed by symmetry, the system should always be able to lower its energy by including an s-wave admixture to the gap. This conclusion is supported by numerous microscopic calculations [31–33,51], which find a significant s-wave component. Similarly, previous interpretations of other experiments in terms of a $d_{x^2-y^2}+s$ gap require a sizable s-wave component. Finally, the lack of a Hebel-Slichter peak in the nuclear magnetic resonance [20] suggests that accidental nodes are unlikely [52].

So far we have considered only models with open, two-dimensional Fermi surfaces. The analysis of pairing symmetry on the basis of quasiparticle scattering rates is by no means limited to such cases, though they represent a clear and relevant example.

For three-dimensional Fermi surfaces, the efficacy of this approach is enhanced by the possibility of measuring the penetration depth perpendicular to different surfaces. This allows one to determine the quasiparticle scattering rates in different planes, and therefore to determine the scattering rate in each crystallographic direction. Given some knowledge of the Fermi surface, the temperature dependence of the three scattering rates obtained should often provide a powerful method to allow one to distinguish between different superconducting gaps. For example, if the Fermi surface is open in one or more planes, it immediately follows from the analysis above that a cubic temperature dependence of the scattering in that direction arises for some gaps but not others, allowing them to be distinguished.

If the Fermi surface is closed, gapless umklapp scattering requires fine tuning of the Fermi surface for any given gap function (as the Fermi surface at the nodal points must be separated by half of a reciprocal lattice vector). Therefore, one does not, generically, expect qualitative differences in the quasiparticle scattering rate for different gap symmetries. However, the magnitude of the exponential correction to the scattering rate, along with any anisotropy, will generally be quite different for different gaps. Thus, the quasiparticle scattering rate may be sufficient to distinguish between different gap structures if the models are sufficiently well defined.

It is also worth briefly discussing alternative scattering mechanisms that may lead to current relaxation. Particularly, we make note of the mechanism of Baber scattering [53], in which quasiparticles of different effective masses scatter, a process which conserves momentum but not velocity. Baber scattering has been discussed previously as a possible mechanism for current relaxation in both normal metals and superconductors [40–42,54]. The contributions of Baber scattering have been found to be less significant than umklapp scattering in models of d-wave superconductors [40], though they may be of greater importance in materials with small Fermi surfaces, where umklapp scattering is not possible. As the masses difference between the two sheets in κ-Br is a band structure effect [47] and not due to electronic correlations, Baber scattering is included in the calculations shown in Fig. 3(b). Thus we see that the presence of Baber scattering does not change our conclusions.

In κ-Br, the anisotropy of the relaxation rate could provide a further test of the superconducting gap. For d_{xy} pairing, the overall relaxation rate is dominated by the contribution in the a direction, as it is only in this direction that there is umklapp scattering involving only nodal quasiparticles. Thus, we predict that a directional measurement of the relaxation rate will show an exponential temperature dependence along the c direction and a cubic dependence in the a direction. As discussed above, such a measurement could be accomplished by measuring the penetration depth perpendicular to three different surfaces of the crystal, which in the case of κ-Br will also involve the much larger interplane scattering rate.

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