Superfluid density and microwave conductivity of FeSe superconductor: ultra-long-lived quasiparticles and extended s-wave energy gap

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Keywords: FeSe, superfluid density, superconductivity, microwave conductivity

Communicated by Peter Hirschfeld

Abstract

FeSe is an iron-based superconductor of immense current interest due to the large enhancements of $T_c$ that occur when it is pressurised or grown as a single layer on an insulating substrate. Here we report precision measurements of its superconducting electrodynamics, at frequencies of 202 and 658 MHz and at temperatures down to 0.1 K. The quasiparticle conductivity reveals a rapid collapse in scattering on entering the superconducting state that is strongly reminiscent of unconventional superconductors such as cuprates, organics and the heavy fermion material CeCoIn$_5$. At the lowest temperatures the quasiparticle mean free path exceeds 50 $\mu$m, a record for a compound superconductor. From the superfluid response we confirm the importance of multiband superconductivity and reveal strong evidence for a non-zero energy-gap minimum.

1. Introduction

A recurring theme in correlated electron research is the sensitivity of such materials to small perturbations, which, when applied, can tune the material through a range of distinct electronic ground states [1–6]. At the heart of this behaviour is a delicate balance between kinetic energy and potential energy, with kinetic energy favouring delocalised electrons and potential energy promoting various types of electronic order. This balance can be tipped in one direction or the other by the application of magnetic field [7] and hydrostatic pressure [8], and by making small changes in chemical composition [9, 10]. In the iron-based superconductor FeSe, the first indication of this sensitivity occurs at around 90 K when the material changes from tetragonal to orthorhombic as it enters a nematic phase [11, 12], lowering rotational symmetry without breaking translational symmetry. At lower temperatures FeSe becomes a superconductor, at $T_c \approx 9$ K under ambient conditions [13], with $T_c$ rapidly increasing by a factor of 4 under hydrostatic pressure [14]. Even more dramatic enhancements occur in single-layer FeSe grown on insulating and semi-insulating substrates such as SrTiO$_3$, with reported values of $T_c$ up to 100 K [15–18]. A major effort is now underway to understand this fascinating new example of high temperature superconductivity.

FeSe is one of the simplest iron-based materials, superconducting at its stoichiometric composition and available as high quality single crystals grown using vapour transport methods [19, 20]. A crucial question is to understand what the bulk, ambient superconductor is holding in reserve, that it can respond so strongly when pressurised [21] or placed in contact with a substrate [22]. Central to addressing this issue is the identification of the superconducting gap structure and, ultimately, the pairing glue [21–27]. On the question of gap structure, a number of measurements emphasise the importance of multiband superconductivity [28–33], but differ on whether or not the energy gap has nodes. The main evidence for nodes appears in a combined study [31] that
High quality single crystals of FeSe were grown by vapour transport using the method described in section 2.1. Samples reported large residual thermal conductivity and power-law penetration depth, λ(T), although these conclusions are by no means universally agreed on [33–35]. A number of scanning tunnelling microscopy (STM) studies show evidence of V-shaped tunneling spectra [31, 36], but the picture here is also complicated. Some STM studies report features that look more like non-zero gaps [32, 37], and there can be a complicated interplay with twin boundaries [37]. In addition, within experimental uncertainty, all STM studies appear to observe zero conductance at zero bias, something that can only occur if there is a single-particle gap at least three times the measurement temperature. This on its own would imply a non-zero gap minimum in the range Δmin/kB ≈ 1.2–4.5 K. Also weighing in in favour of a non-zero gap minimum are a number of bulk probes including heat capacity [29, 32], thermal conductivity [35], μSR [38], lower critical field [34] and penetration depth [33].

In the work reported here we use precision low temperature measurements of the electrodynamic response of FeSe to shed further light on its gap structure and superconducting charge dynamics, in the process revealing extremely long-lived quasiparticles, a testament to the high degree of crystalline order that is possible in this intriguing material.

2. Methods

2.1. Samples

High quality single crystals of FeSe were grown by vapour transport using the method described in [19]. The sample used for the microwave conductivity experiment was a mm-sized platelet, cleaved from a thicker crystal to have a thickness along the c-direction of t = 15 μm.

2.2. Surface impedance and microwave conductivity

Measurements of surface impedance, Zs = R∗ + iX, were made at two frequencies using cavity perturbation [39–43] of a self-resonant coil wound from Nb wire and housed inside a Pb:Sn-coated enclosure mounted below the mixing chamber of an MX40 3He−4He dilution refrigerator [46]. During the experiments, the resonator was maintained at a fixed temperature of 1.5 K, while the sample temperature was scanned between T = 0.1 and 20 K using a silicon hot-finger [47] thermally linked to the mixing chamber. Data were obtained at 202 MHz using the fundamental mode of the coil resonator, and at 658 MHz using its second overtone. In both cases, the sample was positioned at a local maximum of the microwave magnetic field, Hrf, which was applied parallel to the FeSe layers to induce predominantly in-plane screening currents. Measurements were carried out under conditions of constant Hrf, to avoid nonlinearities, and microwave power was kept low enough to prevent self-heating. Temperature-dependent changes in the effective surface impedance, Zs, are obtained directly from shifts in resonator frequency, f0, and resonant bandwidth, fB, using the cavity perturbation approximation ΔZs = Γ(Δf0(T)/2 − iΔf0(T)). Here Γ is a resonator constant determined empirically from the DC resistivity of FeSe samples from the same batch [35], and Δf0 is the change in resonator bandwidth with and without the sample. Due to the thin sample and low measurement frequencies, finite-size effects are important near and above Tk, and are corrected using the 1D finite-size formula Zs = Zs tanh(ωμµατ/2Zs), where Zs is the surface impedance of a semi-infinite sample. The absolute zero-temperature surface reactance, Xs(T = 0) = ωμµαλ0, is set using a previously reported value of the zero-temperature penetration depth, λ0 = 400 nm [31]. The complex microwave conductivity, σ = σ1 − iσ2, is obtained from the surface impedance using the local electrodynamic relation, σ = iωµµα / Zs2.

2.3. Generalised two-fluid model and modified Drude conductivity

For the purposes of extracting relaxation rate Γ and extrapolating superfluid density to the zero-frequency limit we fit conductivity data to a generalised two-fluid model [48, 49]

$$\sigma = \frac{ne^2}{m^*} \left[ \frac{f_f}{1 + \frac{1}{(\omega/\Gamma')^y}} - i\kappa K (\omega/\Gamma', y) \right],$$

(1)

where Γ′ = Γ × y sin(\frac{\pi}{2}) is a convenient scaling that makes the integrated quasiparticle spectral weight independent of y. The imaginary part of the quasiparticle conductivity is obtained using a Kramers–Krönig transform [49] and f_f + f_0 = 1 in order to conserve spectral weight. Away from the Drude limit (y = 2) the frequency exponent y modifies the quasiparticle conductivity to better capture situations with strongly energy-dependent scattering [46, 50]. In FeSe at low temperatures the best-fit value was found to be y = 1.5; at high temperatures, where σ0 has little frequency dependence, the fits are insensitive to the choice of y: the frequency exponent was subsequently fixed at y = 1.5 at all temperatures. The static, London superfluid density is 1/λ2 = f_fµµαne^2/m^*. 
2.4. Two-band extended s-wave superconductor

To model the superfluid density, calculations are made for a clean-limit, two-band superconductor in which the angle dependence of the superconducting order parameter in bands $\nu = 1, 2$ has the form $\Delta_{\nu}(T)\Omega_{\nu}(\phi_{\nu})$, where $\Omega_{\nu}(\phi_{\nu}) = (1 + \sqrt{2}\alpha_{\nu}\cos(2\phi_{\nu}))/(\sqrt{1 + \alpha_{\nu}^2})$ takes an extended $s$-wave form, normalised so that $(\Omega_{\nu}^2(\phi_{\nu}))_{\phi_{\nu}=0} = 1$. Here $\phi_{\nu}$ is a band-specific variable measuring angle around each circular Fermi pocket and $\alpha_{\nu}$ controls the degree of gap anisotropy, with gap nodes appearing for $\alpha_{\nu} \geq 1/\sqrt{2}$. We follow the weak-coupling Eliensmitten method described in [51], modified to incorporate angle-dependent order parameters. Specifically, we solve the self-consistent gap equation

$$\Delta_{\nu} = \sum_{\nu'=1,2} n_{\nu'} \lambda_{\nu\nu'} \Delta_{\nu'} \sum_{\omega_{\nu}>0} \frac{2\pi k_B T}{\Delta_{\nu'}(\Omega_{\nu'}^2(\phi_{\nu'}))]^{1/4}} + \frac{\hbar^2\omega_{\nu}^2}{\Delta_{\nu}(\Omega_{\nu}^2(\phi_{\nu}))_{\phi_{\nu}=0}}$$

\[ (2) \]

where the $\omega_{\nu}$ are the fermionic Matsubara frequencies and the frequency cut-off $\omega_{\nu}$ is set by the measured $T_c$. The relative densities of states $n_{\nu}$ and the coupling constants $\lambda_{\nu\nu'}$ have the same form as in [51] and, subject to the constraints $n_{1} = n_{2} = 1$ and $\lambda_{\nu\nu'} = \lambda_{\nu\nu'}$, are adjustable parameters. The normalised superfluid density, $\rho_{\nu} = \lambda_{\nu}^2(0)/\lambda_{\nu}(T)$, is directly computed from $\Delta_{\nu}(T)$ and $\Delta_{\nu}(T)$ and is a simple linear combination of the contribution from each band: $\rho_{\nu}(T) = \gamma\rho_{1}(T) + (1 - \gamma)\rho_{2}(T)$, where the parameter $\gamma$ controls the relative magnitude of the superfluid weights.

3. Results

In the low frequency limit the electrodynamics of a superconductor are dominated by the superfluid response, giving rise to perfect dc conductivity. Far from being a quiescent state, Cooper pairs are continually being broken apart into quasiparticle excitations then reforming in a phase-coherent manner—such scattering does not degrade a steady current and is important in establishing the equilibrium superfluid density [46]. In order to study the electrodynamic response of the quasiparticles, higher frequencies are needed, ideally comparable to the quasiparticle relaxation rate. Because the superfluid has finite inertia, a high frequency supercurrent is accompanied by an electric field, which in turn couples to the quasiparticle excitations and gives rise to a dissipative response. This results in power absorption that increases as $\omega^2$ and is directly proportional to the real part of the quasiparticle conductivity, $\sigma_{\nu}$.

3.1. Surface impedance

The experimentally accessible quantity is the surface impedance, $Z_{\nu} = R_{\nu} + iX_{\nu}$, with $R_{\nu} \approx \frac{1}{2}\omega^2\rho_{\nu}^2\lambda\sigma_{\nu}$ and $X_{\nu} \approx \omega\mu_{\nu}\lambda$. As we will show below the quasiparticle lifetimes in FeSe are extraordinarily long for a compound superconductor. Experimentally, this means that the GHz-frequency resonators typically used in this type of measurement are too fast, not allowing the quasiparticles sufficient time to relax during the measurement period. That said, surface resistance measurements face significant difficulties at lower frequencies and are rarely attempted [52]: $R_{\nu}$ falls off as $\omega^2$ and the characteristic size of resonators increases like $1/\omega$, making a mm-sized single crystal a negligible perturbation inside a large resonator volume. Our solution has been to employ a special self-resonant coil, of diameter 4 mm and length 10 mm, wound from superconducting Nb wire and operating at $\omega/2\pi = 202$ MHz and $\omega_0/2\pi = 658$ MHz, with empty-coil quality factors of several hundred thousand. Such a resonator has sufficient sensitivity to resolve both the superfluid and quasiparticle response of a mm-sized crystal of FeSe.

Figure 1(a) shows surface resistance data for FeSe at the two frequencies. There is a sharp superconducting transition at $T_c = 9.1$ K, indicative of a homogeneous sample. Surface resistance drops quickly as Meissner screening currents take over from the normal-state skin effect, with $R_{\nu}(T)$ reaching a minimum at $T \approx 7$ K. Below this temperature the surface resistance rises again, with a peak that moves to lower temperature with decreasing frequency. At the lowest temperatures $R_{\nu}$ falls again, decreasing into the $\rho\Omega$ range. The nonmonotonic temperature dependence of $R_{\nu}$ has been observed in only one other material system—ultrahigh purity YBa$_2$Cu$_3$O$_{6+x}$—and is immediately indicative of a system with rapidly collapsing scattering and extremely long-lived quasiparticles [53–56]. The main differences between FeSe and YBa$_2$Cu$_3$O$_{6+x}$ are twofold: the details of the freeze-out of $R_{\nu}(T)$ at low temperatures suggest a non-zero energy gap in FeSe, instead of the symmetry-protected $d$-wave nodes in YBa$_2$Cu$_3$O$_{6+x}$ [57, 58], and in FeSe the nonmonotonic $R_{\nu}(T)$ appears only below about 1 GHz, as compared to about 70 GHz in YBa$_2$Cu$_3$O$_{6.993}$ [55, 56]. This on its own suggests that quasiparticles lifetimes are unusually long in FeSe.
3.2. Microwave conductivity

From the surface impedance we obtain the high frequency conductivity $\sigma = \sigma_1 - i\sigma_2$. The real part of the conductivity is plotted in figure 1(b) for the two measurement frequencies and shows more clearly the underlying quasiparticle dynamics that are responsible for the nonmonotonic form of $R_s(T)$. On cooling through $T_c$, $\sigma_1(T)$ starts to rise. In this temperature range $\sigma_1$ shows no appreciable frequency dependence, consistent with a quasiparticle relaxation rate that is much larger than the measurement frequencies. On further decreasing temperature $\sigma_1(T)$ rises rapidly, eventually peaking at around a third of $T_c$, at a value 40 times higher than $\sigma_1(T_c)$ in the case of the 202 MHz data. As well as in the YBa$_2$Cu$_3$O$_{6+x}$ system [53, 55, 56, 59], qualitatively similar behaviour is observed in the heavy fermion superconductor CeCoIn$_5$ [46, 60], and the organic superconductor $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br [61], although only in Ortho-I YBa$_2$Cu$_3$O$_{6.99}$ is the enhancement as dramatic. In all cases the physics is the same—on passing through $T_c$ there is a sudden collapse in quasiparticle scattering that vastly outpaces the steadier condensation of quasiparticles into the superfluid condensate. Consistent with this conclusion $\sigma_1$ develops substantial frequency dependence below $T_c/2$, indicating that the quasiparticle relaxation rate is becoming comparable to the measurement frequency—in the case of FeSe, a relaxation rate in the sub-GHz range.

3.3. Superfluid density

The imaginary part of the conductivity is dominated by the superfluid term, $\sigma_2 = 1/\omega\mu_0\lambda_L^2$, where $\lambda_L$ is the London penetration depth. We plot the frequency-dependent superfluid density, $1/\omega\mu_0\sigma_2(\omega)$, in figure 1(c). There is some frequency dependence in the lower part of the temperature range, consistent with the presence of slowly relaxing quasiparticles below $T_c/2$ that eventually freeze out at the lowest temperatures. Fits to a generalised two-fluid model (see the methods) are used to extrapolate $\omega\mu_0\sigma_2(\omega)$ to the static limit and obtain the London superfluid density, $1/\lambda_L^2$. The superfluid density shows a strong, approximately linear temperature dependence over most of the temperature range, with upwards curvature in $1/\lambda_L^2(T)$ at around $T_c/3$, which we will discuss below in the context of multiband superconductivity. At the lowest temperatures there is a substantial flattening of $1/\lambda_L^2(T)$, similar to that seen in $R_s(T)$ and $\sigma_1(T)$, that we will show is evidence of small but non-zero gap minima in one of the bands. Broadly similar behaviour in $1/\lambda_L^2(T)$ has very recently been reported in [33].

4. Discussion

To gain further insight into the superconducting charge dynamics we have fit a generalised two-fluid model (see the methods) to the complex conductivity; at each temperature this gives the average quasiparticle relaxation rate, $\Gamma$, and the London superfluid density. The relaxation rate is plotted in figure 2. As expected from the qualitative behaviour of $R_s(\omega, T)$ and $\sigma_1(\omega, T)$ there is a rapid collapse in $\Gamma(T)$ on cooling into the superconducting state, which in the context of the cuprates, organics and CeCoIn$_5$ is interpreted as a strong indication that the fluctuations responsible for inelastic scattering are electronic in origin and are gapped by the onset of superconductivity. Another factor that is relevant to the suppression of $\Gamma(T)$ in these systems is the importance of Umklapp processes in relaxing electrical currents [62, 63]—as we will discuss later, energy.
conservation in Umklapp events becomes difficult to satisfy in multiband superconductors with anisotropic gap. Below 2.5 K $\Gamma(T)$ locks into a linear temperature dependence. Such behaviour is reminiscent of cuprate superconductors in the Born-scattering limit [49, 50, 64, 65], where the linear temperature dependence of $\Gamma$ reflects the linear-in-energy density of states (DOS) of the $d$-wave quasiparticles.

We will argue below that the energy gap in FeSe has an extended $s$-wave form, with deep gap-minima—such a state is accompanied by a linear DOS at energies immediately above the gap minimum and possibly leads to superconductors in the Born-scattering limit [71, 72]. To put these observations on a quantitative footing, as our lowest measurement frequency is of the same order as $\Gamma_{\min}/2\pi \approx 200$ MHz, corresponding to $/k_B \approx 10$ mK in temperature units. Using a value of $v_F \approx 7 \times 10^4$ m s$^{-1}$ for the Fermi velocity [12], the quasiparticle mean free path is $\ell_0 = v_F/\Gamma_{\min} \approx 55 \mu$m. This is the largest value we are aware of for a compound superconductor and is 5 times larger than that of the best YBa$_2$Cu$_3$O$_y$ crystals, where the low temperature scattering rate reaches $\Gamma/2\pi \approx 3.3$ GHz [50]. On the experimental side, we emphasise that this result is on a firm footing, as our lowest mean free path due to these effects, the inferred value of $\ell_0$ is remarkable and suggests that FeSe may provide a novel testing ground for exploring hydrodynamic effects [71, 72] in superconducting transport.

We turn now to the superfluid density, a thermodynamic probe sensitive to the itinerant electronic degrees of freedom, the temperature dependence of which is controlled by the energy dependence of the quasiparticle DOS [73, 74]. The main features of $1/\lambda_2^2$, pointed out above, are its strong temperature dependence across most of the superconducting range, indicating a broad distribution of energy scales in the gap; the upwards curvature of $1/\lambda_2^2(T)$ around $T_G/3$, a hallmark of multiband physics [51]; and its pronounced flattening at low temperatures, indicating the presence of non-zero gap minima instead of true nodes [73, 74]. To put these observations on a quantitative footing we have fit the normalised superfluid density, $\rho_s = \lambda_2^2(0)/\lambda_2^2(T)$, to a two-band model similar to that developed by Kogan, Martin and Prozorov (KMP) in [51]. From initial fitting attempts it became apparent that the small energy scale implied by our data could not be captured by a two-band model with isotropic gaps, motivating us to modify the KMP formalism to include gap anisotropy of an extended $s$-wave form with $\Delta(q) \propto (1 + \sqrt{2} \alpha \cos(2q))$, as summarised above in the methods. In our approach the gap anisotropy is put in by hand but naturally arises from orbital-dependent effects in microscopic
models [68]. In setting up the two-band model, integrals over the Fermi surface are partitioned into discrete sums over a pair of bands. Our model is phenomenological in nature and is therefore not specific about how the Fermi surface is partitioned between the bands—this would require momentum-resolved probes and microscopic theory. In particular, the two-band approach does not require that there be a one-to-one mapping between Fermi surface sheets and ‘bands’. As a case in point, the MgB2 superconductor, with four Fermi surface sheets of complex topology, is well described by two-band BCS theory [51, 73, 75–77]. In our model the fitting parameters are the DOS imbalance between the two bands (parameterised by \( n_1 = N_1/N_{\text{total}} \)); the coupling constants \( \lambda_{11}, \lambda_{22} \) and \( \lambda_{12} = \lambda_{21} \), which represent intraband and interband pairing, respectively; the gap anisotropy parameters, \( \alpha_1 \) and \( \alpha_2 \); and a factor \( \gamma \) that controls the relative superfluid weights. Early on in this process we observed that the large-gap band seemed to require little anisotropy, so \( \alpha_1 \) was set to zero in the remaining work. Equally good fits to \( \rho_s(T) \) are obtained in the range \( 0.25 < n_1 < 0.5 \), with the fit for \( n_2 = 0.35 \) shown in figure 3(a). A typical value of the anisotropy parameter for band 2 is \( \alpha_2 = 0.42 \). The schematic forms of the energy gaps are shown as functions of angle and temperature in figure 3 for \( 0.25 < n_1 < 0.5 \). Note that while there is some variation of \( \Delta_1 \) and \( \Delta_2 \) with the choice of \( n_1 \), all fits agree well on the minimum energy gap, \( \Delta_{\text{min}}/\kappa_0 \approx 0.25 T_C = 2.3 \) K, which is a factor of 8 smaller than the large gap in band 1. Interestingly, very similar conclusions have independently been reached by a field-dependent thermal conductivity study carried out on similar crystals [35], and from penetration-depth experiments [33], where fits to a one-band extended s-wave model estimate \( \Delta_{\text{min}}/\kappa_0 \approx 0.3 T_c \). Within our multiband interpretation, the large conductivity peaks seen in figure 1(b) are the result of long-lived quasiparticles that are thermally excited in the vicinity of the deep gap minima in band 2. This has two important consequences for the relaxation dynamics: there will be a strong reduction in the phase space for recoil when low energy quasiparticles undergo elastic impurity scattering [64, 65, 69]; and Umklapp events [62, 63], which are necessary if inelastic processes are to relax the electrical current, will require the low energy quasiparticles to partner with quasiparticles on the large-gap band, and these excitations will be strongly gapped.

We emphasise that while the two-band model is used mainly for illustrative purposes, the importance of multiband effects and the presence of non-zero gap minima—also visible in \( R_s(T) \) and \( \sigma_1(T) \)—are robust conclusions. In fact, each of the gap values can be directly tied to qualitative features in the temperature dependence of the superfluid density: the gap minimum, \( \Delta_{\text{min}} \), is fixed by the small range of temperatures over which thermally activated behaviour is observed; \( \Delta_2 \), the average value of the gap in the subdominant band, is linked to the upwards curvature in \( \rho_s(T) \), which occurs in a range near \( T_C/3 \); and the dominant gap, \( \Delta_1 \), is set by \( T_C \) itself. It is worth mentioning that the factor of 3 ratio between \( \Delta_1 \) and \( \Delta_2 \) is difficult to obtain in purely repulsive models of pairing, as the dominant interaction in that case is the interband pairing [51], and an unrealistically large imbalance of the DOS (\( n_1 \approx 0.05 \)) must then be assumed to obtain \( \Delta_1 \approx 3 \Delta_2 \).

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**Figure 3.** Superfluid density in a two-band extended s-wave model. (a) Superfluid density is calculated using a two-band extended s-wave model and fit to the experimentally determined London superfluid density, \( 1/\lambda^2(T) \). Inset: a polar plot showing the schematic form for the two-band extended s-wave gap at zero temperature, for various values of the DOS parameter, \( 0.25 < n_1 < 0.5 \). (b) Temperature dependence of the rms gap amplitudes on bands one and two, and the overall gap minimum, for the same range of \( n_1 \).
5. Conclusion

The observation of activated exponential temperature dependence in our low temperature data and the concomitant identification of non-zero gap minima are at odds with reports of line nodes in FeSe, in particular the measurements of superfluid density and thermal conductivity in [31]. As pointed out in the introduction, several tunnelling spectroscopy experiments present V-shaped spectra that at first sight appear to be indicative of gap nodes [31, 36], although the finite-temperature effects (in particular, nonzero conductance at zero bias) that would be expected in the nodal case are not reported. Instead, our observations are in close keeping with the conclusions of a recent thermal conductivity study carried out on the same samples as ours [35], and with heat capacity [29], lower critical field [34] and penetration depth studies [33]. While it is tempting to attribute the lifting of accidental gap nodes to impurity scattering [33, 35], which should homogenise the energy gap in an anisotropic $c$-wave superconductor, we find this hard to reconcile with the exceptionally long quasiparticle mean free path reported here. Nevertheless, sample-to-sample variations of physical properties usually have a microstructural origin. In the case of FeSe, a possible alternative to point-like disorder is the presence of twin boundaries, which form in the nematic phase below 90 K [11, 12, 33, 78]. These have been shown to have a significant effect on the spatial variation of the energy gap in a recent scanning tunneling spectroscopy experiment [37]. Further insights into this may come from local-probe experiments carried out at lower temperatures, if the tunnel junction can be sufficiently cooled, and from measurements on single-domain, macroscopically detwinned samples, when these become available.

Acknowledgments

We thank A V Chubukov, J S Dodge, N Doiron-Leyraud, A V Frolov, P J Hirschfeld, M P Kennett and L Taillefer for discussions and correspondence. We gratefully acknowledge financial support from the Natural Science and Engineering Research Council of Canada, the Canadian Institute for Advanced Research, and the Canadian Foundation for Innovation.

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