Two-gap superconductivity seen in penetration-depth measurements of Lu$_2$Fe$_3$Si$_5$ single crystals

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A single crystal of superconducting Lu$_2$Fe$_3$Si$_5$ was studied using the tunnel-diode resonator technique in the Meissner and mixed states. The temperature dependence of the superfluid density provides strong evidence for two-gap superconductivity and indicates that there are nearly equal contributions from each gap having magnitudes of $\Delta_1/k_B T_c = 1.86$ and $\Delta_2/k_B T_c = 0.54$. In the vortex state, the pinning strength shows unusually strong temperature dependence and is nonmonotonic with the magnetic field (peak effect). The irreversibility line is sharply defined and quite distant from the $H_{c2}(T)$ line, which hints at enhanced vortex fluctuations in this two-gap system. Altogether, our findings from electromagnetic measurements provide strong support for the existence of two-gap superconductivity in Lu$_2$Fe$_3$Si$_5$, as previously suggested from specific-heat measurements.

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I. INTRODUCTION

Initially, interest in the rare-earth iron containing silicides M$_2$Fe$_3$Si$_5$ (M=Y, Sc, and Lu) was due to unusually high superconducting critical temperatures (2.4, 4.5, and 6.0 K, respectively) for compounds containing crystallographically ordered iron sublattices. 1, 5,7 Fe Mössbauer effect measurements indicate that the iron in these materials possess no magnetic moments. 2, 3 Further detailed studies have revealed that other superconducting properties are quite unconventional. The upper critical field $H_{c2}(0)$ for Lu$_2$Fe$_3$Si$_5$ has been found to be unusually large when compared to the other iron-containing silicide superconductors 4, 5 and its temperature dependence differs from convention. Anisotropy and a pronounced peak effect have also been reported in magnetic measurements. 6 The presence of a large residual electronic term in the specific heat below $T_c$, as well as a reduced specific heat jump at $T_c$, has been observed and confirmed indicating departure from the standard Bardeen-Cooper-Schrieffer (BCS)-like behavior. 7, 6 Nonmagnetic impurities have been shown to suppress $T_c$ at a significant rate, which is incompatible with the isotropic $s$-wave BCS picture. 8, 9 On the other hand, ac Josephson effect measurements have indicated an $s$-wave pairing mechanism. 10 Vining, Shelton, Braun, and Pelizzzone have proposed a two-band model in order to explain their specific-heat data. 7 Their model assumes a two-band Fermi surface with one band being superconducting and gapped, and another being normal. This represents an extreme case of multiband superconductivity as we know it today, for example in MgB$_2$, where different bands have gaps of different magnitudes. 11, 12 Later detailed measurements of Lu$_2$Fe$_3$Si$_5$ crystals and analysis have shown that specific-heat data is explained quantitatively well within a two-band model of superconductivity where both bands are gapped but with different gap amplitudes. 13 Recently, a class of superconductors, the iron-containing oxypnictides, was discovered. 14 It has been suggested that these materials could also be multigap superconductors. 15

In this contribution, precision measurements of the London and Campbell penetration depths are presented, the superfluid density is analyzed, and unusual vortex properties are reported. We conclude that Lu$_2$Fe$_3$Si$_5$ is, indeed, a two-gap superconductor. It seems that multiband superconductivity is more widespread and develops when there are different dimensionalities of the Fermi surface on different bands, which leads to reduced interband scattering. In MgB$_2$ there are two- and three-dimensional bands 11, 12 whereas Lu$_2$Fe$_3$Si$_5$ has one- and three-dimensional Fermi surfaces. 13

II. EXPERIMENT

A. Tunnel-diode resonator technique

Measurements of the Lu$_2$Fe$_3$Si$_5$ single crystal were performed using a tunnel diode resonator (TDR). 16-18 An extended review of the use of a TDR to study superconductors is given in Ref. 16. The main components of the TDR are an LC tank circuit and a tunnel diode. The tunnel diode has a region of negative differential resistance in its $I$-$V$ curve. If a dc bias voltage is applied across the diode in this region, then it acts as an ac power source for the LC tank circuit. This results in a self-oscillating circuit, which resonates continuously at a constant frequency for given values of L and C. The resonance frequency of the circuit used in our measurements was near 14 MHz. All throughout the measurements the circuit is kept at a constant temperature, 4.8 K ± 1 mK, allowing for a stability of 0.05 Hz in the resonance frequency over several hours. The sample to be studied is mounted on a sapphire rod with a small amount of Apiezon N grease. The sapphire is inserted inside of the inductor coil of the tank circuit. It is important that the sample and its mount do not make physical contact with the coil so that the temperature of the sample may be changed while keeping the circuit at a constant temperature to maintain the stability. As the magnetic susceptibility of the sample changes with temperature, so does the inductance of the tank coil. This results in a change in the TDR resonance frequency. By measuring the shift in the resonance frequency, we are able to sense changes in the penetration depth on the order of 0.5 Angstroms. Specifically, the frequency shift, $\Delta f = f(T) - f_0$, with...
resistance, evaluated independently from the temperature-dependent relaxation of superfluid density, \(\rho(T)\), obtained from measurements along and perpendicular to the \(c\) axis. The \(c\)-axis penetration depth is measured directly. However, this configuration is more difficult to deal with due to a large demagnetization factor. When the excitation field is applied parallel to the \(c\) axis, currents circulate in both the \(ab\) plane and along the \(c\)-axis direction. However, since our sample is thin along the \(c\)-axis direction, the relative contribution of the currents along this direction, \(\sim t/\omega_{\perp}/\omega_{\parallel} \approx 0.16\), can be neglected and hence \(\lambda_{ab}(T)\) is measured for this orientation as well. Here \(w\) and \(t\) are the planar dimension and thickness, respectively. In addition, the anisotropy of this system is small so the error introduced by the above approximation is minimal. As can be seen in Fig. 2, the penetration depth is very nearly the same for the sample measured in both orientations. The value of \(\lambda_{ab}(0)\) was obtained, as described in Ref. 16, from the reversible magnetization \(dM/d\ln H\). This was measured independently on the same sample using a Quantum Design magnetometer. In further analysis, possible uncertainty in this number up to 25% was examined and confirmed not to change our conclusions in any way.

The symbols in Fig. 3 show the temperature-dependent superfluid density, \(\rho_{s}(T)=[\lambda(0)/\lambda(T)]^2\), calculated from the penetration depth shown in Fig. 2. The solid red curve is the total superfluid density calculated from the \(\alpha\) model, which
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assumes two independent contributions to the total superfluid density and has been successfully applied to the well-known two-gap superconductor MgB2.11,12 In this model each superconducting gap, denoted by \( \Delta /H_{9004} \) and \( \Delta /H_{20849} \), has a similar temperature dependence as that given by the weak-coupling BCS self-consistency equation.16 However, there are different ratios of \( \Delta /k_B T_c \) that become two fitting parameters. A third fitting parameter gives the relative contribution of each band to the total superfluid density, \( \rho_{\text{total}}(T) = x \rho_1(T) + (1 - x) \rho_2(T) \). Each superfluid density is calculated by using the full temperature range semiclassical BCS treatment as described in detail elsewhere.16 These partial \( \rho_1(T) \) and \( \rho_2(T) \) are shown by marked solid lines in Fig. 3. The best fit was achieved with \( x = 0.51 \), \( \Delta_1/k_B T_c = 1.86 \), and \( \Delta_2/k_B T_c = 0.54 \). The first gap is quite close to the weak-coupling value of 1.76, whereas the second gap is much smaller and it is surprising that the earlier two-band model assumed it to be fully normal.7 Similar to MgB2, the two gaps contribute equally to the superfluid density. A dashed line, which almost follows the data, is calculated from the parameters obtained from analyzing the specific-heat data. The parameters obtained from this line are \( x = 0.47 \), \( \Delta_1/k_B T_c = 2.2 \), and \( \Delta_2/k_B T_c = 0.55 \).13 This is in quite good agreement with the values obtained for the superfluid density, given the very different nature of the measurements. To further highlight the qualitative differences between single- and two-gap behavior, we have plotted \( d\rho_s/dT \) in Fig. 4. Note the characteristic nonmonotonic behavior in the case of two gaps. It is neither present in the pure \( d \)-wave nor the pure \( s \)-wave case.

B. Campbell penetration depth (\( H_{dc} \neq 0 \))

While the situation is quite clear for the London penetration depth, measurements in an applied magnetic field reveal more puzzling behavior of the studied compound. When an external dc field is applied and a small amplitude ac response is probed, the vortices respond elastically and the overall susceptibility is governed by the Campbell penetration depth, \( \lambda^2 = \lambda_L^2 + \lambda_s^2 \) where \( \lambda_L \) is the usual London penetration depth described above, and \( \lambda_s(B, T, j) \) is the Campbell penetration depth,19 \( \lambda_s^2 = \phi_0 B/4 \pi \alpha(j) \). Here \( \phi_0 \) is the flux quantum and \( \alpha(j) \) is the Labusch parameter that generally depends on the biasing Bean current generated in the sample, for example after applying a field after cooling in zero field. The magnetic susceptibility of the sample (and the frequency shift) in the vortex state is still given by Eq. (1), but with a generalized penetration depth.

In conventional type-II superconductors there is no hysteresis for zero-field-cooled (zfc) and field-cooled (fc) curves of the small amplitude ac response. However, in materials where \( j_c \) is strongly temperature dependent (e.g., high-\( T_c \) cuprates), a large hysteresis is observed.19 As shown in Ref. 19, a cubic correction to a parabolic potential well for vortex pinning leads to \( \alpha(j) = \alpha_0 \sqrt{1 - j/j_c} \), where \( j_c = \sigma_0 \rho_p \phi_0 \) is the critical current and \( \rho_p \) is the radius of the pinning potential. This model explains why the zero-field-cooled curve differs from subsequent cooling and warming, and it was successfully used to explain the data for the Bi2Sr2CaCu2O8+\( \delta \)+ superconductor.

\( 4 \pi \chi(T) \) in the vortex state of Lu2Fe2Si4 is shown in Fig. 5 for three representative fields. In each case the sample was cooled in zero applied field to the base temperature and the indicated magnetic field was applied. Then the measurements were taken while warming up the sample above \( T_c \) (zfc-w). Then, the sample was cooled and warmed twice without changing the field and while taking the data (fc-c and fc-w). For low field values there is no hysteresis observed, while at intermediate fields the hysteresis becomes very pronounced. Clearly, the hysteresis is associated with the static Bean current, \( j_c \), induced by applying field. We also note that this effect is not associated with the vortex density (e.g., less vortices after zfc) because then the initial Campbell length would be smaller than it is at equilibrium, not larger as observed.

By measuring many \( 4 \pi \chi(T) \) curves at different magnetic fields, we extracted field dependence of the initial suscep-
bility obtained after zfc and fc. Figure 6 shows the resulting curves at $T=0.7$ K. Figure 7 shows the difference between the two curves. This difference is directly related to the strength of pinning and magnitude of the apparent Bean current density, $j$, where we have assumed $j\approx j_c$. There is a clear peak effect and its location is quite compatible with direct measurements reported in Ref. 6.

Finally, we construct the $H-T$ phase diagram obtained from our measurements for both directions. While the Meissner response is governed by currents flowing in the $ab$ plane, in an applied magnetic field the response is anisotropic and is determined by the orientation of the vortices with respect to the crystal axes. We observe large anisotropy of the upper critical field, $H_{c2}(T)$, down to 1 K as shown in Fig. 8, which has not been reported in earlier papers. Furthermore, $H_{c2}(T)$, determined from the TDR measurements, is in excellent agreement with the specific-heat data. Note that $H_{c2}(T)$ is linear in temperature down to $0.15T_c$. Figure 8 also shows position of the irreversibility line (see Fig. 5 for definition) for both orientations. Unlike conventional superconductors, where $H_{irr}(T)$ is very difficult to determine due to its gradual merging into $H_{c2}(T)$, in Lu$_2$Fe$_3$Si$_5$ it is sharply defined and is quite distant from the $H_{c2}(T)$. This is another indication of significant reduction of the critical current possibly due to enhanced fluctuations in the two-gap system.

IV. CONCLUSIONS

In conclusion, we have found that Lu$_2$Fe$_3$Si$_5$ shows a Meissner response compatible with two-gap $s$-wave superconductivity. In the vortex state, it shows unusually strong temperature dependence of the critical current, which is also nonmonotonic with magnetic field (peak effect). The upper critical field is anisotropic and linear in temperature. All these observations are reminiscent of unconventional superconductivity and further theoretical insight to connect these properties is needed.
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