Magnetic penetration depth of single-crystalline SmFeAsO $_{1-x}F_{y}$

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We report measurements of the in-plane magnetic penetration depth λ in single crystals of SmFeAsO_{1-x}F_y ($x \approx y \approx 0.2$) with $T_c \approx 44$ K. We find that at low temperature λ has an exponential temperature dependence which suggests that the Fermi surface is fully gapped. The magnitude of the minimum energy gap, $\Delta_1 = 1.1 \pm 0.1k_BT_c$ at T=0, is significantly smaller than the BCS weak-coupling value suggesting that the gap is either strongly anisotropic or varies significantly between the different Fermi-surface sheets. Our data are well fitted by a two-gap model with the larger gap ($\Delta_2 = 1.7 \pm 0.2k_BT_c$) associated with ~80% of the total superfluid density.

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Superconductivity in the Fe oxypnictide compounds, $LnFeAsO_{1-v}F_{v}$ (where Ln=La, Sm, Ce, Nd, or Pr) has generated an enormous amount of interest. The maximum value of $T_c \simeq 55$ K (Ref. 1) found so far in this series is the highest for any noncuprate superconductor. It is significantly higher than that found for the previous noncuprate record holder MgB₂ ($T_c \simeq 40$ K). The electronic structure of these materials has many similarities with the cuprates. Calculations have shown that the Fermi surface is expected to be quasi two dimensional and strong ferromagnetic and antiferromagnetic spin fluctuations are predicted.^{$\overline{2}$,3} Importantly, the calculations suggest that the electron-phonon interactions are much too weak to produce a T_c of ~55 K.³ Hence, in many ways it might be expected that the superconductivity has more in common with the cuprates than the phonon mediated superconductor MgB₂.

The determination of the symmetry of the superconducting order parameter is an important first step toward uncovering the mechanism of superconductivity in any material. In this regard, measurements of the magnetic penetration depth λ play an important role. Although not a true bulk probe, such as specific heat, penetration depth measurements in the Meissner state probe a few thousand Angstroms below the crystal surface and so should be reasonably representative of the bulk. However, experience with cuprates and MgB₂ has shown that the data are most reliable if measurements are performed on high quality single-crystal surfaces.^{2–4}

Here we report measurement of the in-plane magnetic penetration depth of single-crystal SmFeAsO_{1-x}F_y ($x \approx y \approx 0.2$) with $T_c \approx 44$ K. The data below $T \sim 15$ K show that λ follows an exponential temperature dependence, characteristic of a fully gapped superconductor. The value of the minimum energy gap is found to be significantly lower than the BCS weak-coupling value.

Single crystals of nominal composition SmFeAsO_{0.8}F_{0.2} were grown in Zürich using a high-pressure cubic anvil technique and a NaCl/KCl flux.⁵ The small platelike single crystals have typical dimensions ($80 \times 80 \times 20$) μ m³, with the smallest dimension being along the *c* axis. The penetration

depth was measured using a radio frequency ($F \simeq 12$ MHz) tunnel diode oscillator technique.⁶ The sample was attached with vacuum grease to the end of a high purity sapphire rod and placed in a solenoid coil, which forms part of the inductor of the resonant circuit. The magnetic field was directed along the c direction, and hence all the screening currents flow in the *ab* plane. We estimate that the RF field was $\sim 10^{-6}$ T and the Earth's field was screened with a mu-metal can; hence, we do not expect any contributions from mobile vortices. The change in the resonant frequency of the circuit as the temperature is varied is directly proportional to the change in λ_{ab} . The constant of proportionality was determined from the measured *ab*-plane dimensions of the samples, and the total frequency shift obtained when the sample was extracted from the coil at base temperature using the procedure described in Ref. 7. Past experience has shown this to be accurate to $\sim 20\%$.

Results for the low-temperature behavior of three different crystals are shown in Fig. 1(a). Samples 1 and 2 were from the same batch, and sample 3 was from a different batch and has a somewhat sharper superconducting transition [see Fig. 1(c)]. In the figure we denote the change in λ_{ab} from the values at our lowest measurement temperature as $\Delta\lambda(T)$. The results for all three crystals are very similar, showing that below $T \approx 6$ K the penetration depth tends to a constant value. The absolute changes in λ between our base temperature and 15 K are, within the expected error, the same for all samples.

In Fig. 1(b) data are also shown for an unaligned powdered polycrystalline sample of the same material, which has a slightly higher T_c . This sample was prepared by grinding up a sintered polycrystalline pellet to remove intergrain contacts. In Fig. 1(d) we show the data close to T_c before and after the pellet was ground into powder. The pellet shows a sharp transition with T_c (midpoint)=51.5 K. The much broader transition for the powder is due to $\lambda(T)$ becoming comparable in grain size (~0.5-3 μ m) far below T_c and does not indicate inhomogeneity. For highly anisotropic superconductors such as this⁴ the measured oscillator frequency shift (ΔF) is approximately proportional to the changes in the *in-plane* penetration depth,^{8,9} and hence the



FIG. 1. (Color online) (a) Temperature dependence of the inplane penetration depth of three different single crystals of SmFeAsO_{0.8}F_{0.2}. The solid lines are fits to Eq. (1). Data below T= 3.2 K were excluded from the fits. The data sets for samples 1 and 2 are offset for clarity. The inset shows the data for sample 3 (above 3.2 K only) corrected for paramagnetic normal-state contributions, assuming the magnetic moment values listed. The solid lines are fits to a power-law dependence $\Delta \lambda \propto T^n$. (b) Oscillator frequency shift (proportional to changes in the in-plane penetration depth) for a powder sample. The inset expands the low T portion of the plot. (c) Susceptibility close to T_c for the three single-crystal samples and (d) for the powder (solid symbols) and sintered pellet (open symbols).

data can be directly compared the single-crystal data, although uncertainties in the exact grain size distribution prevented us from determining the absolute values of $\Delta\lambda(T)$. It can be seen that the temperature dependence of $\Delta F \propto \Delta\lambda_{ab}$ is very similar to that for the single-crystal samples. A clear transition is evident at T=3.2 K which is likely to be due to the antiferromagnetic ordering of the Sm ion moments as observed previously.^{10,11} The same transition is also evident in the data for single-crystal sample 3 (but not in the other two crystal samples probably because of slightly high noise levels and base temperature).

In the BCS theory for a fully gapped *s*-wave superconductor, the behavior of $\lambda(T)$ asymptotically approaches

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$$\Delta\lambda(T) = \lambda(0) \sqrt{\frac{\pi\Delta_0}{2k_B T}} \exp\left(-\frac{\Delta_0}{k_B T}\right)$$
(1)

at low temperature. Here $\lambda(0)$ and Δ_0 are the values of λ and the superconducting energy gap Δ at T=0. In practice this provides a good approximation to the full theory for $k_BT \leq \Delta_0/5$. If the energy gap is (*weakly*) anisotropic or there are distinct gaps on different Fermi-surface sheets then the same behavior will still be found but with Δ_0 now being approximately equal to the minimum energy gap in the system, and $\lambda(0)$ is replaced by an effective value $\lambda_e(0)$ which depends on the details of the gap anisotropy.^{12,13}

The solid lines in Fig. 1(a) show fits of the data to Eq. (1) between T=3.2 and 15 K. The lower-temperature limit was chosen to avoid the influence of the Sm ion ordering transition. The gap values found were $\Delta_0/k_B=53\pm 2$, 53 ± 5 , and 58 ± 3 K for samples 1–3, respectively. The fitted values of Δ_0 depend slightly on the upper temperature limit of the data included in the fit, and the quoted error bars encompassed the spread in values obtained for an upper temperature limit less than 15 K ($\simeq T_c/3$). $\lambda_e(0)$ was found to be 1400 ± 200 Å.

The behavior of the susceptibility close to T_c for the three crystals is shown in Fig. 1(c). Sample 3 has a significantly sharper transition than the other two, with T_c (midpoint) =45.0 K and width \sim 2 K. Despite these differences in homogeneity, the low-temperature behavior is the same for all the samples. Taking the ratio of gap found from the fits to the low-temperature exponential behavior to T_c we find that $\Delta_0/k_BT_c = 1.1 \pm 0.1$, which is significantly lower than the weak-coupling s-wave BCS value of 1.76, and suggests the possibility of significant gap anisotropy or multiple gaps such as is found in, for example, NbSe₂ (Ref. 13) and MgB_2 ⁸ As remarked above, significant k dependence of the gap means that $\lambda_{e}(0)$ obtained from the fits of our data to Eq. (1) is not simply related to the zero-temperature value of the penetration depth, $\lambda(0)$. The value of Δ_0/k_BT_c found here is significantly higher than that found for the small gap in MgB₂ ($\Delta_0/k_BT_c=0.76$) and is closer to the value found for the minimum gap in NbSe₂.¹⁴ Evidence for multigap behavior in SmFeAsO_{1-x} F_y has also been inferred from strong observed temperature dependence of the anisotropy parameter $\gamma . ^{4,15}$

Given the many similarities with the cuprates, there has naturally been much speculation regarding the possibility that the superconductivity in these pnictide materials is unconventional. For a simple *d*-wave superconductor at low temperature, $\Delta \lambda \approx \ln 2\lambda(0)k_BT/\Delta_0$. Assuming values appropriate for a *d*-wave state in these materials $[\lambda(0)=2000 \text{ Å}$ and $\Delta_0=2.14k_BT_c]$ we estimate that $d\lambda/dT\approx 14 \text{ Å/K}$ which is at least 2 orders of magnitude larger than any linear term present in our data below ~6 K. In a gapless superconductor, impurities produce a finite zero energy density of states and the temperature dependence of λ changes smoothly from *T* to T^2 , below a temperature scale determined by the impurity concentration.¹⁶ A power-law fit $(\Delta\lambda \sim T^n)$ to our data gives $n \geq 3.5$ which seems to rule out a dirty *d*-wave interpretation.

As these materials contain magnetic ions there is an issue as to how these effect the measured $\lambda(T)$. Cooper¹⁷ showed

that if the normal state of a superconductor is paramagnetic the measured $\lambda_m(T)$ is related to the London depth λ_L by $\lambda_m = \sqrt{1 + \chi_N(T)}\lambda_L$, where χ_N is the normal-state susceptibility. This correction can produce a minimum in the measured $\lambda(T)$ which over a short range of temperature which can resemble an exponential BCS-like T dependence (see, for example, the measurements on the cuprate superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-v}$ which are discussed and reanalyzed in Ref. 17). Assuming a Curie-Weiss form for the susceptibility then for $\chi_N \ll 1$, $\Delta \lambda_m \simeq \Delta \lambda_L + \lambda_L(0) \mu_0 \mu^2 / [3V_{\text{cell}} k_B(T + \theta_N)]$, where μ is the effective moment of each magnetic ions (2 per unit cell). In the inset to Fig. 1(a) we show the data for sample 3 (above T_N) with this second term subtracted (with θ_N =3.4 K, μ in the range 0-3 μ_B and $\lambda(0)$ =2000 Å). For free ion Sm³⁺, $\mu = 1.74 \ \mu_B$ and direct measurement¹⁸ of χ_N above T_c in SmFeAsO_{1-x} \tilde{F}_y with T_c similar to ours give μ =1.0 μ_B and θ_N =50 K, so the maximum values we have used are probably an overestimation of the effect by 1 or 2 orders of magnitude. A power-law fit to this corrected data results in minimum exponent of 2.8, which is still far in excess of 2 expected in a dirty d-wave model and closer to that expected for a fully gapped state.

Electronic structure calculations for the sister material LaFeAsO_{1-x} F_x show that the bands crossing the Fermi level originate mostly from the two-dimensional Fe layer and give rise to quasi-two-dimensional cylindrical sheets running along the c axis and centered on the Γ point (hole) and the M point (electron).¹⁰ The general topology of this Fermi-surface is consistent with recent angle resolved photoemission measurements.¹⁹ As discussed by Mazin et al.³ this Fermisurface topology places constraints on the type of pairing symmetries which are likely; for example, it is unfavorable to states which have a strong *c*-axis or inplane angular dependence. In particular, these authors argue that the structure of the magnetic fluctuations favors an unusual type of multiple gap s-wave superconductivity where the order parameter on the electron and hole Fermi-surface sheets has opposite phase. Our experiment contains no phase information, but our results are compatible with a fully gapped s-wave state with moderate anisotropy. Fully gapped states have been conjectured by a number of authors, at least in certain limits of the parameters in their models (for example, see Ref. 20 and references therein). The presence of two different types of Fermi-surface sheets suggests the possibility of two-gap superconductivity although there significant intersheet scattering is expected, which would make the magnitudes of the two gaps similar.³

As remarked above, fitting the low-temperature $\lambda(T)$ data to Eq. (1) will deduce the size of the minimum superconducting gap. To deduce the presence of any other gaps or any other form of gap anisotropy it is necessary to analyze the full temperature dependence of the normalized superfluid density $\rho = \lambda^2(0)/\lambda^2(T)$ up to T_c . In our experiment we are not able to measure the absolute value of λ . So in order to calculate the superfluid density it is necessary to use an estimate of $\lambda(0)$ from other experiments. Weyeneth *et al.*⁴ reported measurement of λ deduced from the reversible magnetic torque measured on samples produced using exactly the same method as those in the present work with a similar T_c . From their data they estimated $\lambda_{ab}(0)=2100\pm 300$ Å, which



FIG. 2. (Color online) Calculated normalized superfluid density $[\lambda^2(0)/\lambda^2(T)]$ versus temperature for sample 3 using different assumed values for the zero-temperature penetration depth $\lambda(0)$. The solid lines are fits to the two-gap model described in the text, and the inset table shows the fit parameters. The dashed line shows the isotropic *s*-wave BCS prediction with $\Delta_0 = 1.76k_BT_c$. Note that for clarity not all data points are plotted. The inset shows the low-temperature data on an expanded scale.

is in reasonable agreement with the value of $\lambda \simeq 1900$ Å measured by μ SR for SmFeAsO_{0.85} (T_c =52 K).¹¹

In Fig. 2 we plot the superfluid density calculated from our data using two realistic value of $\lambda(0)$ [note that because of surface roughness we expect our effective values of $\lambda(0)$ to be higher than the bulk values]. The dashed line in the figure shows the isotropic s-wave BCS prediction with Δ_0 = $1.76k_BT_c$. Although this is an acceptable fit to the highertemperature data it fails to account for the lowesttemperature data because the gap is too large. We have therefore attempted to fit the data with a simple two-gap BCS model (as used for MgB₂, for example^{8,13}). Here it is assumed that the gaps (Δ_1 and Δ_2) on each Fermi-surface sheet (or section of sheet) follow the weak-coupling BCS temperature dependence but have a variable low-temperature absolute value. There is an additional parameter x which is the fraction of the superfluid density on the sheet with the smaller gap. In principle, there are four free parameters, $(x, T_c, \Delta_1, \text{ and } \Delta_2)$, but we fix $\Delta_1 = 1.1 T_c$ as suggested by the low-temperature exponential fits and $T_c=44$ K as suggested by a linear extrapolation of the superfluid density close to T_c . This model fits the data well over the full temperature range, except very close to T_c . The value of the larger gap is determined to be $\Delta_2 = 1.7 \pm 0.2$ (6.4 ± 0.7 meV) which accounts for $80\pm5\%$ of the superfluid density. This value of Δ_2 is close to the BCS weak-coupling gap value. This is consistent with the gap value found by Chien et al.²¹ using point contact Andreev spectroscopy on SmFeAsO_{0.85} $F_{0.15}$ (T_c =40 K), although they saw no evidence for the smaller gap. This difference may be because of the much smaller amount of *c*-axis warping expected for the hole sheets.

In summary, our data for the in-plane penetration depth of the oxypnictide superconductor SmFeAsO_{0.8}F_{0.2} show an exponential temperature dependence at low temperature indicating a fully gapped pairing state. Our results show evidence for a moderate variation in the gap on the different Fermi-surface sheets. Fitting our data with a two-gap model suggests that the second gap is ~60% larger than the first.

Note added. There have been a number of important de-

velopments in this field recently. Penetration depth data for PrFeAsO_{1-y} (T_c =35 K) (Ref. 22) also show evidence for fully gapped behavior similar to that reported here. However, data for Ba(Fe_{1-x}Co_x)₂As₂ (T_c =13–24 K) (Ref. 23) show a robust power-law behavior of $\lambda(T)$ with exponent *n* in the range of 2–2.5. Very recently, data for very clean samples of LaFePO (T_c =5.6 K) (Ref. 24) display a linear *T* dependence of λ which is strongly indicative of nodes. Taken together the

- ¹Z. Ren et al., Chin. Phys. Lett. 25, 2215 (2008).
- ²D. J. Singh and M.-H. Du, Phys. Rev. Lett. 100, 237003 (2008).
- ³I. I. Mazin, M. D. Johannes, and G. A. Sawatzky, Phys. Rev. Lett. **101**, 089703 (2008).
- ⁴S. Weyeneth, U. Mosele, N. D. Zhigadlo, S. Katrych, Z. Bukowski, J. Karpinski, S. Kohout, J. Roos, and H. Keller, J. Supercond. Novel Magn. **22**, 325 (2009).
- ⁵N. D. Zhigadlo, S. Katrych, Z. Bukowski, and J. Karpinski, J. Phys.: Condens. Matter **20**, 342202 (2008).
- ⁶A. Carrington, R. W. Giannetta, J. T. Kim, and J. Giapintzakis, Phys. Rev. B **59**, R14173 (1999).
- ⁷R. Prozorov, R. W. Giannetta, A. Carrington, and F. M. Araujo-Moreira, Phys. Rev. B **62**, 115 (2000).
- ⁸F. Manzano, A. Carrington, N. E. Hussey, S. Lee, A. Yamamoto, and S. Tajima, Phys. Rev. Lett. **88**, 047002 (2002).
- ⁹W. Anukool, S. Barakat, C. Panagopoulos, and J. R. Cooper, (to be published).
- ¹⁰A. J. Drew et al., Phys. Rev. Lett. **101**, 097010 (2008).
- ¹¹R. Khasanov, H. Luetkens, A. Amato, H.-H. Klauss, Z. A. Ren, J. Yang, W. Lu, and Z.-X. Zhao, Phys. Rev. B **78**, 092506 (2008).
- ¹²In the simple case of two gaps with $\Delta_1 \gg \Delta_2$, then at low temperature $(k_B T \ll \Delta_1) \lambda_e \simeq x \lambda(0)$.
- ¹³J. D. Fletcher, A. Carrington, O. J. Taylor, S. M. Kazakov, and J. Karpinski, Phys. Rev. Lett. **95**, 097005 (2005).
- ¹⁴J. D. Fletcher, A. Carrington, P. Diener, P. Rodiere, J. P. Brison,

evidences to date indicate that either the order parameter is nonuniversal in the ferropnictides or that there are intrinsic nodes in all materials which are lifted by disorder.²⁵

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- R. Prozorov, T. Olheiser, and R. W. Giannetta, Phys. Rev. Lett. **98**, 057003 (2007).
- ¹⁵S. Weyeneth, R. Puzniak, N. Zhigadlo, S. Katrych, Z. Bukowski, J. Karpinski, and H. Keller, J. Supercond. Novel Magn. **22**, 347 (2009).
- ¹⁶P. J. Hirschfeld and N. Goldenfeld, Phys. Rev. B 48, 4219 (1993).
- ¹⁷J. R. Cooper, Phys. Rev. B **54**, R3753 (1996).
- ¹⁸M. R. Cimberle, C. Ferdeghini, F. Canepa, M. Ferretti, A. Martinelli, A. Palenzona, A. Siri, and M. Tropeano, arXiv:0807.1688 (unpublished).
- ¹⁹C. Liu et al., arXiv:0806.2147 (unpublished).
- ²⁰K. Seo, B. Bernevig, and J. Hu, Phys. Rev. Lett. **101**, 206404 (2008).
- ²¹T. Y. Chen, Z. Tesanovic, R. H. Liu, X. H. Chen, and C. L. Chien, Nature (London) **453**, 1224 (2008).
- ²²K. Hashimoto et al., Phys. Rev. Lett. 102, 017002 (2009).
- ²³R. T. Gordon, C. Martin, H. Kim, N. Ni, M. A. Tanatar, J. Schmalian, I. I. Mazin, S. L. Bud'ko, P. C. Canfield, and R. Prozorov, Phys. Rev. B **79**, 100506 (2009).
- ²⁴J. D. Fletcher, A. Serafin, L. Malone, J. G. Analytis, J.-H. Chu, A. S. Erickson, I. R. Fisher, and A. Carrington, Phys. Rev. Lett. (to be published).
- ²⁵ V. Mishra, G. Boyd, S. Graser, T. Maier, P. Hirschfeld, and D. Scalapino, Phys. Rev. B **79**, 094512 (2009).