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Normal-State Electrical Resistivity and Superconducting Magnetic Penetration Depth in $Eu_{0.5}K_{0.5}Fe_2As_2$ Polycrystals¶

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We report measurements of the temperature dependence of the electrical resistivity, $\rho(T)$, and magnetic penetration depth, $\lambda(T)$, for polycrystalline samples of $Eu_{0.5}K_{0.5}Fe_2As_2$ with $T_c = 31$ K. $\rho(T)$ follows a linear temperature dependence above T_c and bends over to a weaker temperature dependence around 150 K. The magnetic penetration depth, determined by radio frequency technique displays an unusual minimum around 4 K which is associated with short-range ordering of localized Eu^{3+} moments.

The recent discovery of superconductivity in LaOFeP at $T_c \approx 4$ K by Kamihara et al. [1] has lead to intensive studies on electron and hole doped iron arsenide oxide superconductors RFeAsFO (R = La, Sm) with T_c as high as 55 K in $SmFeAsO_xF_{1-x}$ [2]. Very recently, Rotter et al. [3] found that the oxygen free iron arsenide $BaFe_2As_2$ in which Ba is partially substituted by potassium ions, is a superconductor below $T_c = 38$ K, which was confirmed for (KSr) Fe_2As_2 compounds with $T_c = 37$ K [4]. The FeAs layers common to both series of compounds seem to be responsible for superconductivity. Jeevan et al. recently observed that $EuFe_2As_2$ shows a spin-density wave (SDW) type transition at 190 K, and becomes superconductive below 32 K after partial substitution of Eu by 50% K [5]. Below about 10 K, short-range magnetic order of the Eu moments was suggested by a feature in the magnetic susceptibility. Here we focus at first on the temperature dependence of the normal-state resistivity and then on the superconducting magnetic penetration depth in order to probe the influence of local Eu^{2+} moments on superconductivity.

Polycrystalline samples of $Eu_{0.5}K_{0.5}Fe_2As_2$ were synthesized from stoichiometric amounts of the starting elements Eu (99.99%), K (99.9%), Fe (99.9%), and As (99.999%) by solid-state reaction method under Argon atmosphere, as described in [5]. The sample crystallizes in the tetragonal structure with lattice parameters $a = 3.8671$ Å and $c = 13.091$ Å [5]. X-ray analysis reveals that the composition of the samples is close to the expected 0.5 : 0.5 : 2 : 2 stoichiometry. Samples had form of rectangular bars of about $1.7 \times 1.7 \times 1.1$ mm.

A standard four-probe *ac* (9 Hz) technique was used for resistance measurements. A well-defined cubic geometry of the samples provided for the precise $\rho(T)$ and superconducting properties measurements through van der Pauw four probe method. The temperature was measured with platinum (PT-103) and carbon glass (CGR-1-500) sensors. The measurements were performed in a liquid Helium variable temperature cryostat in the temperature range between 1.3 and 300 K. Magnetic measurements of $\rho(T)$ and $\lambda(T, H)$ were carried out using a superconducting coil in applied fields of up to 3 T and at temperatures down to 1.3 K.

We used a radio frequency LC technique [6] to measure $\lambda(T)$ of $Eu_{0.5}K_{0.5}Fe_2As_2$ samples. This technique employs a simple rectangular solenoid coil into which the sample is placed. Changes in the magnetic penetration depth of the sample lead to the change of the coil's inductance L that in turn results in the change of the resonance frequency ω (2–20 MHz) of the LC circuit. The connection between parameters of the circuit and $\lambda(T)$ is described by following simple equation:

$$\lambda(T) - \lambda(0) = \delta\frac{\omega^{-2}(T) - \omega^{-2}(0)}{\omega^{-2}(T_n) - \omega^{-2}(0)}. \quad (1)$$

Here $\delta = 0.5\sqrt{c^2\rho/2\pi\omega}$ is the imaginary part of a skin depth above T_c, which was determined from the $\rho(T)$ measurements [6], $\omega(T)$ is the resonance frequency of the circuit at arbitrary T, $\omega(T_n)$ and $\omega(0)$ are the same one's above T_c and at zero temperature, respectively.

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Figure 1 shows the normal-state resistivity $\rho(T)$ of $Eu_xK_{1-x}Fe_2As_2$ sample at a doping $x = 0.5$. $Eu_{0.5}K_{0.5}Fe_2As_2$ is a bad metal with a specific resistivity around 300 μOhm cm at room temperature. To emphasize the variation of $\rho(T)$ in a superconducting state, we plot these data below 50 K in the inset. $\rho(T)$ decreases smoothly with temperature, while drops abruptly to zero with a midpoint at $T_c = 31$ K, which clearly indicates superconductivity. Above T_c, $\rho(T)$ exhibits a linear temperature dependence up to 120 K and develops a remarkably pronounced downturn from its linear-T behavior at higher temperatures. We first try to analyze the $\rho(T)$ dependence in terms of the Bloch–Grüneisen (BG) equation for the electron-phonon (e–p) scattering:

$$\rho(T) - \rho(0) = 4\rho_1 t^5 \int_0^{1/t} \frac{x^5 e^x dx}{(e^x - 1)^2}. \qquad (2)$$

Here, $\rho(0)$ is the residual resistivity, $\rho_1 = d\rho(T)/dt$ is the slope of $\rho(T)$ at high $T > T_R$, $t = T/T_R$ and T_R is the resistive Debye temperature. It is clear from Fig. 1 that the BG model describes the $\rho(T)$ dependence below 120 K with rather low $T_R = 180$ K, suggesting an importance of the e–p interaction. However, we could not fit $\rho(T)$ in the entire temperature range with Eq. (2) because the resistance bending over 120 K.

Such an unusual $\rho(T)$ dependence in Fe_2As_2 compounds is far from being clear and disputed in the scientific community. The abrupt changes in the $\rho(T)$ dependence at 150 K may be considered as a signature of a phase transition, where the crystal structure changes from tetragonal to orthorhombic, as was observed by Rotter et al. [5] at 140 K for different compositions of $Ba_{1-x}K_xFe_2As_2$. The reduction of the lattice symmetry was visible by (110)-reflections XRD peak splitting up to $x = 0.2$, however is absent for superconducting samples at $x = 0.3$. Thus, the tetragonal to orthorhombic phase transition, as well as the magnetic (spin-density-wave) transition are completely suppressed in superconducting $Ba_{0.6}K_{0.4}Fe_2As_2$ [5]. At the same time the resistivity bending over at 120 K is still present [7].

Very recently, Gooch et al. [8] fitted the low-temperature part of $\rho(T)$ at $T < 100$ K of $Ba_{1-x}K_xFe_2As_2$ to a power-law dependence, $\rho(T) - \rho(0) = AT^n$, and found evidence for quantum critical behavior: The exponent n sharply decreases with x from $n = 2$ to $n = 1$ near a critical concentration $x_c = 0.4$, and then increases again to a value close to 2 at $x = 1$ [8]. Furthermore, the thermoelectric power divided by temperature displays a logarithmic dependence $S(T)/T \propto \log T$ near critical doping. Both results would be compatible with a quantum critical point at x_c which is hidden by superconductivity, similar as found in various heavy-fermion systems [9]. Whereas in the heavy-fermion case the characteristic magnetic energy scale is

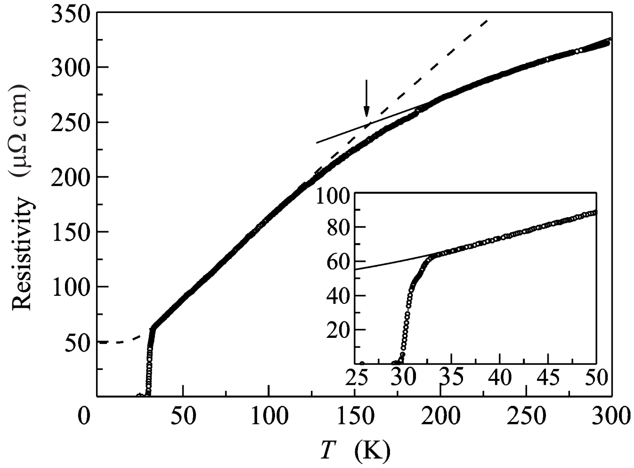

Fig. 1. (Color online). Temperature variation of the resistivity of $Eu_{0.5}K_{0.5}Fe_2As_2$ sample. The inset shows the superconducting transition on an enlarged scale. Dashed line is a fit with BG Eq. (2) below 150 K and solid line is extrapolation from $\rho(T)$ above 150 K.

of the order of 10 K and quantum criticality is typically cut-off above this temperature, in Fe_2As_2 systems, the SDW transition takes place at about 200 K and thus, quantum criticality is expected to extend up to much higher temperatures. In this scenario, the observed crossover in $\rho(T)$ of $Eu_{0.5}K_{0.5}Fe_2As_2$ at 150 K would then mark the upper limit of the universal quantum critical regime in the system. Certainly, the existence of quantum critical fluctuations in Fe_2As_2 systems needs to be investigated by inelastic neutron diffraction or other magnetic probes. We also note, that the $\rho(T)$ dependence in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystals in the normal state remains almost linear up to room temperature [10].

We now turn to the magnetic penetration depth in the superconducting state of $Eu_{0.5}K_{0.5}Fe_2As_2$. Given that the $\lambda(T)$ dependence has a BCS form close to T_c:

$$\lambda(T) = \frac{\lambda(0)}{\sqrt{2\left(1 - \frac{T}{T_c}\right)}}, \qquad (3)$$

we plot $(\omega^{-2}(T) - \omega^{-2}(0))/(\omega^{-2}(T_n) - \omega^{-2}(0))$ data versus BCS reduced temperature: $1/\sqrt{2(1 - T/T_c)}$ in the region close to T_c. We use the slope of $\lambda(0)/\delta$ vs. $1/\sqrt{2(1 - T/T_c)}$ and Eq. (3) to obtain an unusually large value of $\lambda(0) = 4.02 \times 10^{-4}$ cm from $\delta = 1.088 \times 10^{-2}$ cm.

For a BCS-type superconductor with the conventional s-wave pairing form, the $\lambda(T)$ has an exponen-

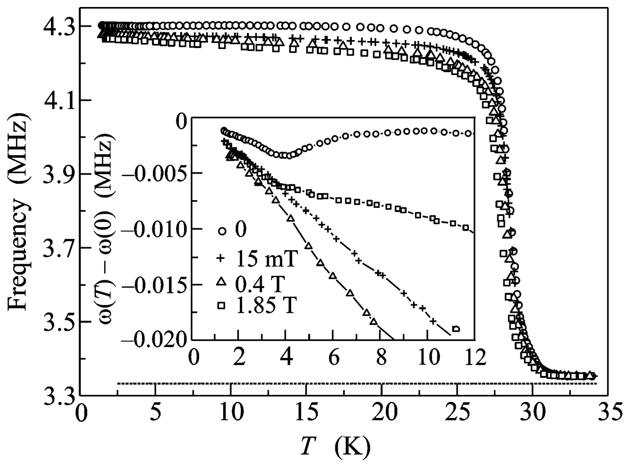

Fig. 2. (Color online). Temperature variations of resonance frequency of LC circuit $\omega(T)$ for $Eu_{0.5}K_{0.5}Fe_2As_2$ sample. The inset shows the temperature dependence of $\omega(T) - \omega(0)$ in extended scale. The dashed curve is for empty coil.

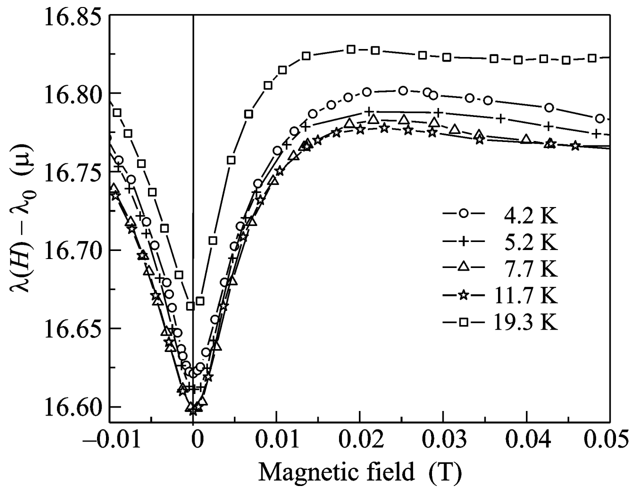

Fig. 3. (Color online). Typical magnetic field variation of $\lambda(H) - \lambda(0)$ of a $Eu_{0.5}K_{0.5}Fe_2As_2$ sample at different temperatures: 4.2; 5.2; 7.7; 11.7 and 19.3 K. The solid lines are the guides for the eye.

tially vanishing temperature dependence below $T_c/2$ (where $\Delta(T)$ is almost constant) [6]:

$$\lambda(T) = \lambda(0)\sqrt{\frac{1}{\tanh(\Delta(0)/2k_BT)}} \quad (4)$$

for dirty limit: $l < \xi$ [6]. Here $\Delta(0)$ is the energy gap.

In Fig. 2 we compare the temperature dependencies of $\omega(T)$ behavior at rather small magnetic fields. As we can see from the inset, the low T part of this dependence has unconventional minimum around 4.2 K, which become a break like in small magnetic field 15 mT, and completely disappear at larger field 0.4 T. Also, the magnetic field dependence of $\omega(T)$ is quite strong. On the other hand, the $\omega(T)$ curves clearly display a smooth variation below 3 K which simplifies the extrapolation of the resonance frequency $\omega(T)$ of our LC circuit down to zero temperature in order to calculate $\lambda(T)$ from Eq. (1). At the same time the existence of this minima makes impossible the exploration of the exponentially vanishing BCS temperature dependence according to Eq. (4) below $T_c/2$ for the determination of $\Delta(0)$.

We plot in Fig. 3 the deviation $\lambda(H) - \lambda(0)$ as a function of the magnetic field at very small H. In contrast to measurements of the magnetic induction on $PrFeAsO_{1-y}$ [11], the $\lambda(H) - \lambda(0)$ dependence displays a sharp signature in the magnetic field dependence with clear tendency towards saturation at 15 mT independently from temperature, while we expect a linear dependence with a break point at low fields caused by the Meissner effect [6]. The observed smooth minimum in $\lambda(H)$ at 4.2 K has the same origin as $\omega(T)$ shown in Fig. 2. This result indicates that there is no edge point in $\lambda(H)$ close to the true field of flux penetration in striking contrast with magnetization data in $PrFeAsO_{1-y}$ used to deduce H_{c1} [11]. Thus we could not determine the value of H_{c1} in contrast to, e.g., the case of ZrB_{12} [6], apparently due to possibly melting of the vortex solid and the presence of strong vortex pinning [12].

In the absence of vortices we probe the London penetration depth λ. Important problems for $\lambda(T)$ measurements are: (i) the determination of the basic superconducting parameter $\lambda(0)$ and (ii) its temperature dependence, to see whether *s*-wave or *d*-wave pairing form exist. Both these problems can be addressed from the low-T $\lambda(T)$ dependence. However, one can easily notice from Fig. 4 an unconventional

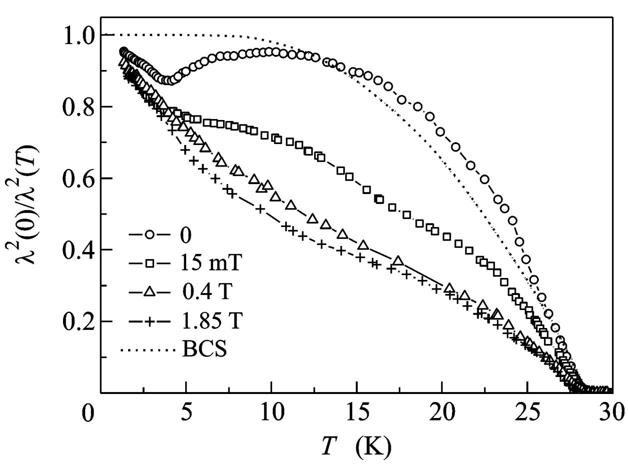

Fig. 4. (Color online). Superfluid density, $[\lambda(0)/\lambda(T)]^2$, of the $Eu_{0.5}K_{0.5}Fe_2As_2$ sample in different magnetic fields for the $\lambda(0) = 4.02 \times 10^3$ nm. The predicted behavior of $[\lambda(0)/\lambda(T)]^2$ within the BCS model is shown by dotted line.

behavior of the superfluid density $[\lambda(0)/\lambda(T)]^2$ of $Eu_{0.5}K_{0.5}Fe_2As_2$ at low temperatures. In contrast to BCS-type behavior, we observe a small but well defined anomaly with a pronounced minimum at 4 K. Small magnetic fields wash out this feature and strongly influence the superfluid density.

Apparently, the strong magnetic field dependence of $\lambda(T)$ is due to magnetic flux lines partially penetrating the sample in the vortex state of the superconductor. Very strong flux pinning was also observed by Eskildsen et al. [12] in $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystals with a disordered vortex arrangement. In our system the magnetic field will also affect the Eu ions. The observed anomaly in $\lambda(T)$ is very likely related to short-range ordering of the Eu^{2+} moments coexisting with the superconducting state below 10 K, as seen in the magnetic susceptibility [5] and ^{151}Eu Mössbauer spectroscopy [13].

The magnetic susceptibility anomaly at low T was absent in $(KSr)Fe_2As_2$ compounds [3, 4] as well as in the $\lambda(T)$ dependence for $Ba(Fe_{0.93}Co_{0.07})_2As_2$ single crystals [14]. While the specific heat vs. T signature associated with the superconducting transition provides clear evidence of the bulk nature of superconductivity in $Eu_{0.5}K_{0.5}Fe_2As_2$ [5], the rather large $\lambda(0)$ indicates an unusually large penetration of the electromagnetic field in this compound with composition close to the quantum critical point. We would like to stress that $\lambda(0)$ was determined from the temperature dependence of $\lambda(T)$ close to T_c by assuming a BCS-like form, but not from low T data, which are masked by magnetism of Eu ions. The influence of the short-range Eu-ordering on the lower-critical field and on the pinning behavior in $Eu_{1-x}K_xFe_2As_2$ should be studied in more detail.

In summary, we have performed a systematic study of the temperature and magnetic field dependence of the resistivity, $\rho(T)$, and the magnetic penetration depth, $\lambda(T)$, on polycrystalline samples of $Eu_{0.5}K_{0.5}Fe_2As_2$. The $\rho(T)$ dependence may be described by the Bloch–Grüneisen formula only in a limited temperature regime below 120 K and bends over at higher temperatures. Alternatively, the observed $\Delta\rho \propto T$ dependence, may be interpreted in terms of quantum critical behavior which is cut-off above 120 K. The superfluid density does not exhibit a BCS-type dependence and has an unconventional minimum close to 4 K, very likely due to a short-range ordering of Eu ions. Small magnetic fields destroys this signature. Altogether, our results indicate unusual normal and superconducting properties in $Eu_{0.5}K_{0.5}Fe_2As_2$.

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