

# Normal-State Electrical Resistivity and Superconducting Magnetic Penetration Depth in $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ Polycrystals<sup>¶</sup>

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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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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  $\text{Eu}^{3+}$  moments.

The recent discovery of superconductivity in  $\text{LaOFeP}$  at  $T_c \approx 4$  K by Kamihara et al. [1] has led to intensive studies on electron and hole doped iron arsenide oxide superconductors  $\text{RFeAsFO}$  ( $\text{R} = \text{La}, \text{Sm}$ ) with  $T_c$  as high as 55 K in  $\text{SmFeAsO}_x\text{F}_{1-x}$  [2]. Very recently, Rotter et al. [3] found that the oxygen free iron arsenide  $\text{BaFe}_2\text{As}_2$  in which Ba is partially substituted by potassium ions, is a superconductor below  $T_c = 38$  K, which was confirmed for (KSr)  $\text{Fe}_2\text{As}_2$  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  $\text{EuFe}_2\text{As}_2$  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  $\text{Eu}^{2+}$  moments on superconductivity.

Polycrystalline samples of  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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  $\omega$  (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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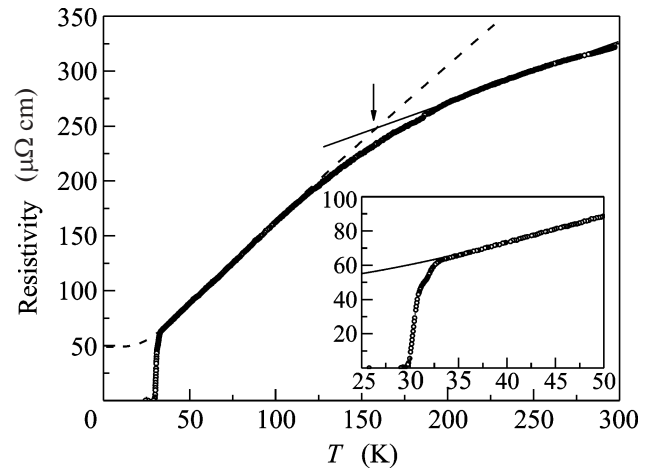
Figure 1 shows the normal-state resistivity  $\rho(T)$  of  $\text{Eu}_x\text{K}_{1-x}\text{Fe}_2\text{As}_2$  sample at a doping  $x = 0.5$ .  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  is a bad metal with a specific resistivity around  $300 \mu\text{Ohm cm}$  at room temperature. To emphasize the variation of  $\rho(T)$  in a superconducting state, we plot these data below  $50 \text{ K}$  in the inset.  $\rho(T)$  decreases smoothly with temperature, while drops abruptly to zero with a midpoint at  $T_c = 31 \text{ K}$ , which clearly indicates superconductivity. Above  $T_c$ ,  $\rho(T)$  exhibits a linear temperature dependence up to  $120 \text{ K}$  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:

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

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  $120 \text{ K}$  with rather low  $T_R = 180 \text{ K}$ , 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  $120 \text{ K}$ .

Such an unusual  $\rho(T)$  dependence in  $\text{Fe}_2\text{As}_2$  compounds is far from being clear and disputed in the scientific community. The abrupt changes in the  $\rho(T)$  dependence at  $150 \text{ K}$  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  $140 \text{ K}$  for different compositions of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ . 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  $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$  [5]. At the same time the resistivity bending over at  $120 \text{ K}$  is still present [7].

Very recently, Gooch et al. [8] fitted the low-temperature part of  $\rho(T)$  at  $T < 100 \text{ K}$  of  $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  sample. The inset shows the superconducting transition on an enlarged scale. Dashed line is a fit with BG Eq. (2) below  $150 \text{ K}$  and solid line is extrapolation from  $\rho(T)$  above  $150 \text{ K}$ .

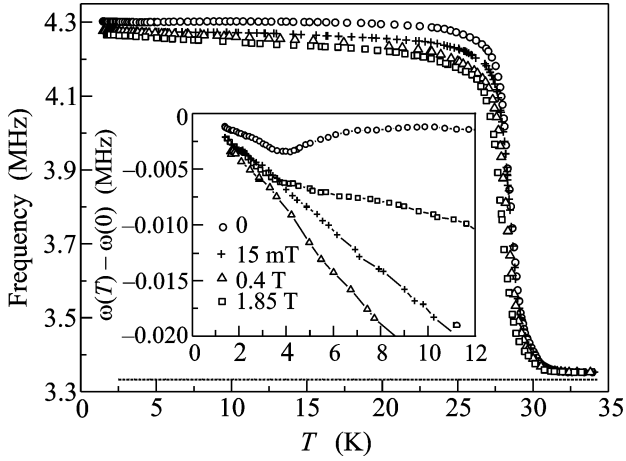
of the order of  $10 \text{ K}$  and quantum criticality is typically cut-off above this temperature, in  $\text{Fe}_2\text{As}_2$  systems, the SDW transition takes place at about  $200 \text{ K}$  and thus, quantum criticality is expected to extend up to much higher temperatures. In this scenario, the observed crossover in  $\rho(T)$  of  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  at  $150 \text{ K}$  would then mark the upper limit of the universal quantum critical regime in the system. Certainly, the existence of quantum critical fluctuations in  $\text{Fe}_2\text{As}_2$  systems needs to be investigated by inelastic neutron diffraction or other magnetic probes. We also note, that the  $\rho(T)$  dependence in  $\text{Ba}(\text{Fe}_{0.93}\text{Co}_{0.07})_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ . Given that the  $\lambda(T)$  dependence has a BCS form close to  $T_c$ :

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

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  $\lambda(0) = 4.02 \times 10^{-4} \text{ cm}$  from  $\delta = 1.088 \times 10^{-2} \text{ cm}$ .

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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  sample. The inset shows the temperature dependence of  $\omega(T) - \omega(0)$  in extended scale. The dashed curve is for empty coil.

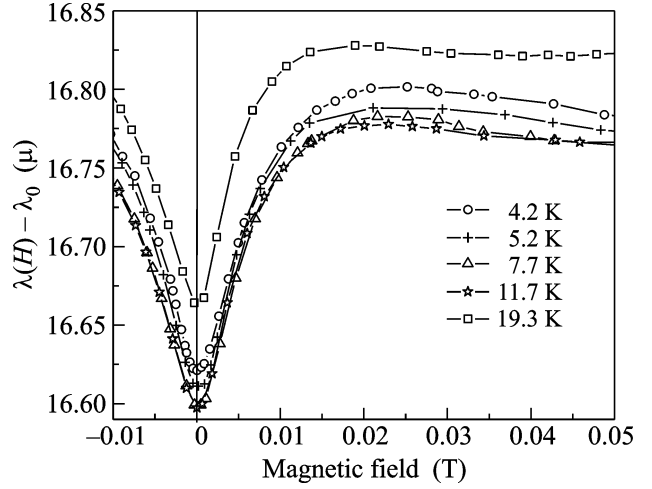
tially vanishing temperature dependence below  $T_c/2$  (where  $\Delta(T)$  is almost constant) [6]:

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

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  $\Delta(0)$ .

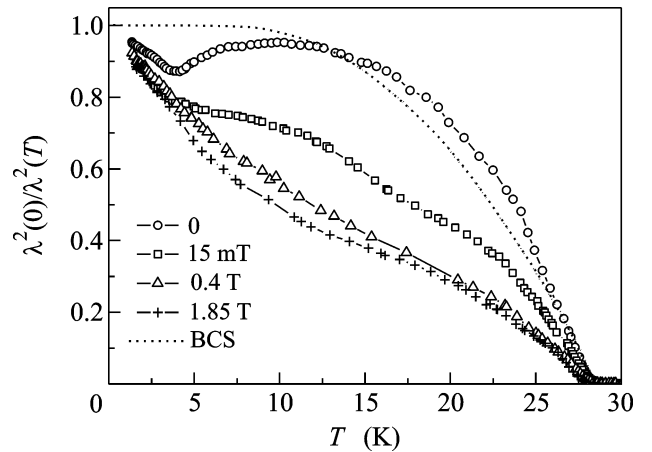
We plot in Fig. 3 the deviation  $\lambda(H) - \lambda(0)$  as a function of the magnetic field at very small  $H$ . In contrast to measurements of the magnetic induction on  $\text{PrFeAsO}_{1-y}$  [11], the  $\lambda(H) - \lambda(0)$  dependence displays a sharp signature in the magnetic field dependence 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



**Fig. 3.** (Color online). Typical magnetic field variation of  $\lambda(H) - \lambda(0)$  of a  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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.

data in  $\text{PrFeAsO}_{1-y}$  used to deduce  $H_{c1}$  [11]. Thus we could not determine the value of  $H_{c1}$  in contrast to, e.g., the case of  $\text{ZrB}_{12}$  [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  $\lambda$ . 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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  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  $\text{Ba}(\text{Fe}_{0.93}\text{Co}_{0.07})_2\text{As}_2$  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  $\text{Eu}^{2+}$  moments coexisting with the superconducting state below 10 K, as seen in the magnetic susceptibility [5] and  $^{151}\text{Eu}$  Mössbauer spectroscopy [13].

The magnetic susceptibility anomaly at low  $T$  was absent in  $(\text{KSr})\text{Fe}_2\text{As}_2$  compounds [3, 4] as well as in the  $\lambda(T)$  dependence for  $\text{Ba}(\text{Fe}_{0.93}\text{Co}_{0.07})_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$  [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  $\text{Eu}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$  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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ . 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  $\text{Eu}_{0.5}\text{K}_{0.5}\text{Fe}_2\text{As}_2$ .

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