Normal-state electrical resistivity and superconducting magnetic penetration depth in Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ 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$_2$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 120 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$^{2+}$ moments.

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The recent discovery of superconductivity in LaOFeP at $T_c\approx4$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$_x$F$_{1-x}$ [2]. Very recently, Rotter et al. [3] found that the oxygen free iron arsenide BaFe$_2$As$_2$ in which Ba is partially substituted by potassium ions, is a superconductor below $T_c=38$ K, which was confirmed for (K/Sr)Fe$_2$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 EuFe$_2$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$^{2+}$ 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$_2$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 $a=3.8671\,\text{Å}$ and $c=13.091\,\text{Å}$ [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 $1.7\times1.7\times1.1\,\text{mm}^3$.

A standard four-probe $ac$ (9Hz) technique was used for resistance measurements. A well-defined cubic geometry of the samples provided for the precise $\rho(T)$ measurements through van der Pauw four probe method and the superconducting magnetic penetration depth experiments. 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 K 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$_2$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 [6]:

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

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.

Fig.1 shows the normal-state resistivity $\rho(T)$ of Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ sample at a doping $x = 0.5$. Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ is a bad metal with a specific resistivity around 300 $\mu\Omega$ 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 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:

$$\rho(t) - \rho(0) = 4 \rho_1 t^{1/4} \int_0^{1/x} \frac{x^5 e^x dx}{(e^x - 1)^2}$$
Here, $\rho(0)$ is the residual resistivity, $\rho(T) = \frac{d\rho(T)}{dt}$ is the slope of $\rho(T)$ at high $T > T_R$, $t = \frac{T}{T_R}$ 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 unusual $\rho(T)$ dependence in Fe$_2$As$_2$ 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. [7] at 140 K for different compositions of Ba$_{1-x}$K$_x$Fe$_2$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 $x \geq 0.3$. 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$_2$As$_2$ [7]. 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$_x$Fe$_2$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 of the order of 10 K and quantum criticality is typically cut-off above this temperature, in Fe$_2$As$_2$ 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$_2$As$_2$ 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$_2$As$_2$ 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$_{0.07}$)$_2$As$_2$ single crystals in the normal state remains almost linear up to room temperature [14].

We now turn to the magnetic penetration depth in the superconducting state of Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$. Given that the $\lambda(T)$ dependence has a BCS form close to $T_c$:

$$\lambda(T) = \frac{\lambda(0)}{\sqrt{2\cdot(1-\frac{T}{T_c})}}$$

we plot $[\omega^2(T) - \omega^2(0)]/[\omega^2(T_c) - \omega^2(0)]$ 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 (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 exponentially vanishing temperature dependence below $T_c/2$ (where $\Delta(T)$ is almost constant) [6]:
FIG. 2. (color online) Temperature variations of resonance frequency of LC circuit $\omega(T)$ for Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ sample. The inset shows the temperature dependence of $\omega(T)$-$\omega(0)$ in extended scale. The dashed curve is for empty coil.

\[
\lambda(T) = \lambda(0) \cdot \sqrt{\frac{1}{\tanh(\frac{\Delta(0)}{2k_BT})}} \tag{4}
\]

for dirty limit, i.e. with short mean free path $l<\xi$ [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 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 $\lambda(H) - \lambda(T=0)$ as a function of the magnetic field at very small $H$. In contrast to measurements of the magnetic induction on PrFeAsO$_{1-x}$ [10], 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$_{1-x}$ used to deduce $H_{c1}$ [10]. Thus we could not determine the value of $H_{c1}$ in contrast to e.g. the
case of ZrB$_2$ [6], apparently due to possibly melting of the vortex solid and the presence of strong vortex pinning [14].

![Graph](image-url)

**FIG.3.** (color online) Typical magnetic field variation of $\lambda(H)$- $\lambda(0)$ of a Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ sample at different temperatures: 4.2 K, 5.2 K, 7.7 K, 11.7 K and 19.3 K. The solid lines are the guides for the eye.

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 behavior of the superfluid density $[\lambda(H)/\lambda(0)]^2$ of Eu$_{0.5}$K$_{0.5}$Fe$_2$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. [13] in Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$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 Eu$^{2+}$ moments coexisting with the superconducting state below 10 K, as seen in the magnetic susceptibility [5] and $^{151}$Eu Mössbauer spectroscopy [11].

The magnetic susceptibility anomaly at low-T was absent in (K/Sr)Fe$_2$As$_2$ compounds [3,4] as well as in the $\lambda(T)$ dependence for Ba(Fe$_{0.93}$Co$_{0.07}$)$_2$As$_2$ single crystals [12]. 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$_2$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$^{2+}$ ions. The influence of the short-range Eu-ordering on the lower-critical field and on the pinning behavior in Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ should be studied in more detail.

FIG. 4. (color online) Superfluid density, $[\lambda(0)/\lambda(T)]^2$, of the Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$ sample in different magnetic fields for the $\lambda(0)=4.02\times10^3$nm. The predicted behavior of $[\lambda(0)/\lambda(T)]^2$ within the BCS model is shown by dotted line.

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$_2$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 BCS type dependence and has an unconventional minimum close to 4 K, very likely due to a short-range ordering of Eu$^{2+}$ ions. Small magnetic fields destroy this signature. Altogether, our results indicate unusual normal and superconducting properties in Eu$_{0.5}$K$_{0.5}$Fe$_2$As$_2$.

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