Unusual Field-Dependence of the Intrgrain Superconductive Transition in RuSr$_2$EuCu$_2$O$_8$

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I. INTRODUCTION

Interest has been raised since the reported co-existence of superconductivity (SC), weak ferromagnetism (FM), and antiferromagnetism (AFM) in Ru$_2$Sr$_2$EuCu$_2$O$_8$ (Ru1212Gd) and Ru$_2$Sr$_2$EuCu$_2$O$_8$ (Ru1212Eu). The superconducting transition in these compounds show the typical behavior of a granular superconductor. Two steps of the transition, ascribed to intergrain and intragrain transitions, are well separated in magnetic susceptibility and electrical resistivity experiments. However, recent neutron scattering experiments have shown a more complex nature of the magnetic ordering including a change of the principal axis of the magnetic moments with an applied external field. Therefore, the homogeneity of the intragrain magnetic and superconducting states is questioned. Extrinsic structural defects or the possible phase separation of FM and AFM regions are expected to have a major effect on the intragrain superconductivity. In such a scenario a grain of a ceramic Ru1212 sample could actually consist of nanoscale SC domains coupled through Josephson junctions below a thermodynamic transition temperature, $T_c$. Therefore, the behavior of the intragrain SC under magnetic field, $H$, is of particular interest. The magneto-transport properties of Ru1212 should strongly depend on the intragrain Josephson coupling strength. In the strong coupling limit, the intragrain Josephson-junction penetration depth ($\lambda_2$) may roughly be equal to the London penetration depth ($\lambda_L$) below an intragrain phase-lock transition of $T_2$, and the intragrain SC properties are similar to those of other "ordinary" cuprates. In the weak coupling limit, however, individual grains may rather be similar to disordered Josephson-junction arrays (JJA) with an unusually large $\lambda_2$. In this case a severe suppression of $T_2$ by a comparatively small magnetic field is expected.

Another interesting observation is the recently reported field dependence of a specific heat anomaly near 46 K in Ru1212Gd. It was suggested that the $T_c$ increases with magnetic field $H$ up to 4 to 5 T due to a possible p-wave superconductivity. Such an abnormal $H$-dependence of $T_c$ should show up also in the magneto-transport properties. In the strong (intragrain) Josephson-coupling limit $T_2$ is expected to show a similar field dependence as $T_c$ whereas in the weak-coupling limit $T_2$ should decrease with $H$ as is qualitatively known for Josephson junction arrays. Previous magneto-resistance (MR) measurements below the ferromagnetic transition temperature, $T_F$, show a positive MR at low fields passing through a maximum at about 2 Tesla but no effect on the superconducting $T_c$ was reported.

Despite the intensive investigations of the last decade the SC properties of a disordered 2D JJA under magnetic field is still a matter of debate. Depending on the nature of the disorder and the model used different ground states have been proposed, e.g. various glass states, paramagnetic Meissner state and chirally ordered normal phase. Various phase transitions may appear (e.g. glass transitions). The intragrain magnetotransport of Ru1212, therefore, offers the opportunity to explore 2D JJA if the coupling strength is moderate to weak, i.e. if the intragrain $T_2$ is clearly smaller than $T_c$ and the penetration depth, $\lambda_2$, is unusually large.

This motivated our investigation of the magnetotransport of Ru1212 ceramic samples. In order to avoid any additional magnetic contribution from the Gd-moment the measurements were mainly done on Ru1212Eu which has been shown to be very similar to Ru1212Gd with respect to the FM and SC transitions. AC susceptibility $\chi$ under a dc bias field $H$ combined with bulk resistivity was used to extract the intragrain SC transition. Various powder samples were also used to identify the characteristic length scales involved.
II. EXPERIMENTAL SETUP

Ceramic samples with a nominal composition RuSr$_2$EuCu$_2$O$_8$ were prepared by solid-state reaction techniques as described earlier. The X-ray diffraction pattern shows that the sample is nearly single phase with a small amount of SrRuO$_4$. Elemental analysis reveals a slight Ru deficiency and some extra copper with the actual cation composition of Ru: Sr: Eu: Cu = 0.91(5): 2.106(2): 2.29(2). From structural, magnetic and transport properties it was concluded that RuSr$_2$EuCu$_2$O$_8$ is very similar to its extensively investigated sister compound, RuSr$_2$GdCu$_2$O$_8$. In particular, the superconducting transition proceeds in two steps indicating intergrain and intragrain transitions of a granular superconductor.

DC and AC susceptibility measurements were performed using the Quantum Design SQUID magnetometer. The AC measurements were done with a field amplitude of 3 Oe in a DC bias field up to 5 Tesla. Magnetoresistance data were measured in the same SQUID by an AC resistance bridge from Linear Research.

III. RESULTS AND DISCUSSION

The zero-field cooled (ZFC) and the field cooled (FC) magnetizations of a bulk piece of Ru1212Eu at 5 Oe are shown in Fig. 1. The magnetic transition temperature $T_F = 115$ K is slightly lower than the 133 K observed in Ru1212Gd probably due to the possible Cu/Ru site mixing as suggested by the cation nonstoichiometry. The intergrain coupling seems to be rather weak so that the nonlinearity of the ZFC $M - H$ branch begins below 1 Oe and $M$ saturates at 4 Oe (Inset, Fig. 1). Two drops of $M_{ZFC}$ appear at 20 K and 28 K, respectively. The susceptibility $(M_{ZFC}/H)$ decreases quickly below 20 K and reaches -0.04 emu/cm$^3$ (about 50% shielded-volume fraction) at 5 K. The magnetization of a powder sample with an average particle size of about 3 $\mu$m is also shown in Fig. 1 (solid lines). Based on the comparison of the data we conclude that the 20 K transition indicates the intergrain phase-lock temperature and the 28 K diamagnetic transition is the intragrain transition at $T_2$. This interpretation is further supported by the AC susceptibility data shown in Fig. 2. The peaks in the imaginary part, $\chi''$, and the drops in the real part, $\chi'$, appear around 20 K and 30 K, respectively. Note that these temperatures are about 10 K lower than those of Ru1212Gd ($T_1 \approx 30$ K and $T_2 \approx 40$ K) probably due to the chemical pressure caused by the replacement of Gd by the smaller Eu. The diamagnetic drop between $T_1$ and $T_2$, $\Delta M_{ZFC} \approx 0.002$ emu/cm$^3$, in Ru1212Eu is significantly larger than that in Ru1212Gd suggesting a shorter $\lambda_2$ in our Ru1212Eu sample. The larger intragrain Meissner effect, $\Delta M_{FC}$, above $T_1$ observed here seems to be closely related to the shorter $\lambda_2$ but will not be discussed further since it is not essential to the topic presented here.

The zero field resistivity, $\rho(0)$, and thermoelectric power, $S$, of the bulk piece of Ru1212Eu have been discussed previously. $\rho$ reaches a maximum around $T_m = 36K$ and drops to zero slightly above 20 K. The thermoelectric power changes slop close to $T_m$ and decreases to zero slightly above the zero resistance temperature. The details of the resistive slope are shown in the derivative, $d\rho/dT$ (Fig. 3). In zero magnetic field (Fig. 3a) $d\rho/dT$ appears as a close superposition of two peaks corresponding to the inter- and intragrain superconducting transitions. The two peaks are split off by small magnetic field and can easily decomposed by assuming Gaussian peak shapes (dotted lines in Fig. 3b). This procedure is used to separate the contributions from both transitions to the overall drop of resistivity and to estimate the transition temperatures, $T_1$ and $T_2$, respectively. In particular, the deconvolution of the two peaks allows us to determine the temperature $T_2$ where the intragrain resistivity is close to zero. This temperature should roughly correspond to the $T_2$ identified in the magnetization measurements. In the following evaluation we define $T_2$ from resistivity by a 95 % drop of the intragrain resistance as indicated by the double sided arrow in Fig. 3b. The chosen criterion may appear somewhat arbitrary but it does not affect the main conclusions. The maximum of resistivity appears at higher temperature, $T_m$, defined by the zero crossing of $d\rho/dT$.

The intergrain transition temperature, $T_1$, decreases quickly in an external magnetic field. This is typical for a granular superconductor with weak intergrain coupling. The field dependence of the intragrain temperature, $T_2$, is of special interest and is estimated from both, magneto-resistivity data as described above, and AC susceptibility experiments in a DC bias field. A typical AC susceptibility curve is shown in Fig. 4 for an external DC field of 1000 Oe. The intragrain transition is clearly detected by the drop of susceptibility. $T_2$ is determined from the crossing of the two extrapolated linear parts of $\chi$ above and below the transition (Fig. 4). Surprisingly, $T_2$ is rapidly suppressed by small magnetic fields with an initial slope of 100 K/T (Fig. 5). The $T_2$’s estimated from resistivity and susceptibility are shown in Fig. 5 by open triangles and closed circles, respectively. The agreement of both data sets is very good, the deviation at $H \approx 0$ is obviously due to the larger uncertainty in the deconvolution of the two peaks in $d\rho/dT$ at very small field (Fig. 3a). The unusually strong decrease of the intragrain $T_2$ of $dT_2/dH \approx 100$ K/T at low field cannot be explained by bulk and homogeneous superconductivity inside the grains. However, a steep field dependence of $T_2$ is expected if the intragrain transition is considered to be a phase-lock transition of another (intragrain) Josephson junction array. The underlying subgrain structures could be structural or magnetic domains of typical nanometer size. The broad intragrain resistivity transition, in particular the broadening of the $d\rho/dT$ peak with field $H$,
might be understood as a percolative transition between domains coupled by junctions of different strength and disorders induced by \( H \). The AC susceptibility, however, shows a narrow phase-lock transition (width \( \approx 0.7 \) K) between the two linear sections (Fig. 4) indicating a true phase-transition. The width of this transition does not change with magnetic field. The linear decrease of \( \chi \) below \( T_2 \) can be explained by the decrease of the penetration depth, \( \lambda_2 \). From an analysis of the intragrain superconducting transitions of powders with different particle size it is concluded that the penetration depth of the Ru1212Eu sample used here is \( \lambda_2(0K) \approx 1 \mu m \). Details of this analysis will be published elsewhere.

Besides \( T_1 \) and \( T_2 \), the maximum in resistivity \( (T_m) \) and the resistance in the ferromagnetic phase, \( \rho(H) \), are relevant quantities for investigating the interplay of superconductivity and magnetic structures. At low field, up to about 0.4 T, \( \rho(H) \) and \( T_m(H) \) are not changed by the magnetic field. For \( H > 0.4 \) T, however, the resistivity increases and its maximum shifts to higher temperature, saturating at about 4 to 5 T. As shown in Fig. 6, \( \rho(H) \) above 50 K and \( T_m(H) \) seem to be strongly correlated. A positive magneto-resistance similar to the present data for Ru1212Eu has also been reported for Ru1212Gd below the ferromagnetic transition temperature. The anomalous increase of \( T_m(H) \) and \( \rho(H) \) above 0.4 T may be due to a change in the magnetic structure of Ru1212Eu as previously proposed from neutron scattering experiments on Ru1212Gd. According to the neutron scattering data the Ru moments are ordered antiferromagnetically with a small residual ferromagnetic moment. The direction of the moments was suggested to be parallel to the tetragonal c-axis. At 0.4 T the Ru moments are assumed to rotate into another antiferromagnetic structure and a sizable ferromagnetic magnetization can be induced at higher fields. This change in the magnetic structure is obviously reflected in the positive magneto-resistivity and the increase of \( T_m \) as discussed above. As a consequence \( \rho(H) \) and \( T_m(H) \) should be correlated. In fact, the inset in Fig. 6 indicates a linear relation between \( \rho \) and \( T_m \), but a change of slope possibly takes place at a field of about 1.5 T. The origin of this slope change is not clear and needs further exploration. It should be noted that a similar field induced shift to higher temperature was qualitatively observed in the peak of specific heat in Ru1212Gd and was interpreted as a possible signature of p-wave superconductivity. However, since no heat capacity data between 0 and 4 T have been reported a more detailed comparison with the present resistivity data is not possible and it is not clear if both phenomena are of the same physical origin.

The key issue to understand the unusual field dependence of \( \rho(H) \) is the magnetic structure involving antiferromagnetic ordering with a weak ferromagnetic component. The strong field dependence of the intragrain \( T_2 \) observed in the present data indicates inhomogeneities in the intragrain structure resulting in a weak link JJA in the superconducting state. Therefore, phase separation into nanoscale AFM and FM domains may be considered as a possible scenario. The field \( H \) is expected to change the intra-domain magnetic order as well as the domain structure. The growth of FM domains with increasing field \( H \) will result in enhanced carrier scattering and a positive magneto-resistance as long as the induced FM moment does not dominate the AFM order. Only at high field (>5 T) the FM domains determine the transport properties and, due to reduced carrier scattering, the magneto-resistance should drop again. Percolative effects could play an essential role.

The physical origin of the observed increase of the resistivity maximum temperature, \( T_m \), with the applied field is still an open question. If \( T_m \) is considered as the onset of superconductivity the phenomenon must be related to the magnetic microstructure and its correlation to the superconducting state. It is interesting that the field effect on \( T_m \) is only observed above 0.4 T where it has been suggested that the principal axis of alignment of the AFM moments changes from c-axis to in-plane. This observation calls for a more detailed investigation.

**IV. CONCLUSIONS**

We have shown that the intragrain superconducting transition temperature of RuSr\(_2\)EuCu\(_4\)O\(_8\) decreases steeply as a function of an external magnetic field. This effect is explained by assuming that the intragrain superconductivity is due to a phase-lock transition of a nanoscale Josephson junction array. The resistivity in the ferromagnetically ordered phase and the temperature of the resistivity maximum both increase as function of magnetic field above 0.4 T. The positive magnetoresistance and the Josephson junction nature of the intragrain superconductivity could be explained by a phase separation into antiferromagnetic and ferromagnetic domains below the magnetic transition temperature.

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FIG. 1. DC susceptibility of bulk and powder sample of RuSr$_2$EuCu$_2$O$_8$

circles: ZFC mode; triangles: FC mode
upper and lower lines: powder sample in FC and ZFC mode
inset: M vs. H at 5 K

FIG. 2. AC susceptibility of RuSr$_2$EuCu$_2$O$_8$ in a DC bias field of 1000 Oe.
The intragrain transition temperature, $T_2$, is determined from the crossing of the two straight lines.

FIG. 3. Derivative of resistivity at the superconducting transition
(a) zero magnetic field; (b) $H=500$ Oe
Part (b) shows the decomposition separating intergrain and intragrain superconducting transitions.
The temperature $T_2$ where the intragrain resistivity drops to 5% is indicated

FIG. 4. AC susceptibility of RuSr$_2$EuCu$_2$O$_8$ in a DC bias field of 1000 Oe.
The intragrain transition temperature, $T_2$, is determined from the crossing of the two straight lines.

FIG. 5. Field dependence of $T_2$ from resistivity (open triangles) and AC susceptibility data (closed circles)
The inset shows the Ginzburg-Landau dependence for a bulk superconductor.

FIG. 6. Magnetoresponse, $\rho(50K)$, (open circles) and temperature of resistivity maximum, $T_m$, (closed squares) as function of the magnetic field. The inset shows the correlation between $\rho$ and $T_m$. 

FIG. 1. DC susceptibility of bulk and powder sample of RuSr$_2$EuCu$_2$O$_8$
circles: ZFC mode; triangles: FC mode
upper and lower lines: powder sample in FC and ZFC mode
inset: M vs. H at 5 K

FIG. 2. AC susceptibility of RuSr$_2$EuCu$_2$O$_8$
open triangles and squares: real and imaginary part of bulk sample
solid line: real part of powder sample
The graph shows the relationship between temperature (T, K) and magnetic susceptibility (M/H, emu/cm$^3$) and magnetization (M, emu/cm$^3$) for different values of magnetic field (H, Oe) at 5 K.
(a) $H=0$

(b) $H=500$ Oe

$\frac{d\rho}{dT} (\Omega \text{cm}/K)$

$T (K)$

$T - 5\% R_{\text{intra}}$
\[ H_{dc} = 1000 \text{ Oe} \]
Ginzburg-Landau:
$$T_c(B) = T_c(0) \left[1 - \frac{B}{B_c(0)}\right]^{1/2}$$
