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Magnetism, upper critical field and thermoelectric power of the magnetosuperconductor $RuSr_2Eu_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$

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Abstract
The magnetic susceptibility, M–H plot, magnetoresistance and thermoelectric power of the $RuSr_2Eu_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ superconductor are measured. Values of the magnetic transition temperature T_{mag}, superconductivity transition temperature T_c, upper critical field H_{c2}, chemical potential μ and energy width for electric conduction W_σ are obtained from these measurements. It has been found that $T_{mag} = 140$ K, $T_c = 25$ K (33 K) from susceptibility (magnetoresistance) measurements, $H_{c2}(0) > 32$ T, $\mu = 8$ meV and $W_\sigma = 58.5$ meV. These values are compared with other ruthenate superconductors, and the resulting physical information is discussed.

1. Introduction

The coexistence of superconductivity and magnetism was reported in ruthenium copper oxide materials $RuSr_2(Gd, Sm, Eu)_{1.6}Ce_{0.4}Cu_2O_{10-\delta}$ (Ru-1222) in 1997 [1, 2] and later in $RuSr_2GdCu_2O_8$ (Ru-1212) in 1999 [3–5]. Both of these oxides were synthesized in 1995 and studied for their transport properties [6]. The Ru-1212 phase is structurally related to the $CuBa_2YCu_2O_{7-\delta}$ (Cu-1212) phase such that the Cu–O chain of Cu-1212 is replaced by the RuO_2 sheet. In the Ru-1222 structure furthermore, a three-layer fluorite-type block instead of a single oxygen-free R (=rare earth element) layer, is inserted between the two CuO_2 planes of the Cu-1212 structure [7].

Substantial work has been carried out on the Ru-1212 phase. The magnetic structure was studied through neutron diffraction experiments [8]. Electron microscopy revealed a

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superstructure along the a–b plane due to tilting of the RuO_6 octahedra [9], which was further confirmed by neutron diffraction studies [10]. The appearance of bulk superconductivity at low temperatures in Ru-1212 was initially criticized by Chu *et al* [11]. However, later works by Bernhard *et al* [12] and Tokunaga *et al* [13] showed that superconductivity exists in this compound within a magnetically ordered state. A few substantial review articles/book chapters are also available on these ruthenocuprate magnetosuperconductors [14–17].

We notice from the recent work on ruthenocuprates [18–21] that the magnetism of the RuO_2 layers in the Ru-1222 system is quite different from that of Ru-1212. The superstructures due to the tilting of the RuO_6 octahedra in Ru-1222 are also qualitatively different from those for the Ru-1212 system. Thus there is a need to investigate the magnetism in these two ruthenocuprates [22]. A related problem is the nature of superconducting and transport behaviour due to the presence of the magnetic effect in the ruthenocuprates. Hence we also study this problem. For specificity, we consider the ruthenocuprate $RuSr_2Eu_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ ($Eu_{1.5}$-1222), and measure its susceptibility, magnetization, magnetoresistance, thermoelectric power and lattice expansion. From the susceptibility measurements we extract the temperature values where different types of magnetic order (antiferromagnetism, weak ferromagnetism and diamagnetism) take place. The temperature dependence of the upper critical field $H_{c2}(T)$ has been extracted from the magnetoresistance data. The thermoelectric power S has been analysed in terms of a normal band model, and the relevant values of the parameter are extracted.

Many other workers have also studied the physical properties of different variants of the Ru-1222 system by taking either different rare earths (Gd, Sm etc) or different contents ($Gd_{1.5}$, $Gd_{1.4}$ etc) of these ions [18, 23, 24]. Cardoso *et al* [18] have made (dc and ac) magnetic measurements on the $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ ($Gd_{1.5}$-1222) system. They report, in particular a spin glass transition over a significant temperature range. Escote *et al* [23] have studied three samples of $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ ($Gd_{1.4}$-1222) with varying oxygen content. They found that for high oxygen content $Gd_{1.4}$-1222 is a superconductor with metallic resistivity in the normal state. When the oxygen content is reduced the metallic behaviour of resistivity ρ shrinks to a limited temperature range. In particular they studied the occurrence of an antiferromagnetic state and an upper critical field. Shi *et al* [24] studied the electrical, transport and magnetic properties of $RuSr_2Sm_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ ($Sm_{1.4}$-1222). These authors also found that the metallic portion of the resistivity shrinks with reduced oxygen content. We shall also compare our thermoelectric power data with those for the $RuSr_2Gd_{1-x}La_xCu_2O_8$ sample of Liu *et al* [25].

It is well known that the physical properties of the ruthenocuprates depend on the preparation conditions [26]. This limits the scope for comparison of our data with the data of other groups [18, 23, 24] on different Ru-1222 systems. In fact, a commendable attempt was made as early as 2002 to summarize the x-ray absorption near-edge spectroscopy, electrical resistance and thermopower measurements for $RuSr_2Gd_{2-x}Ce_xCu_2O_{10+\delta}$ compounds [27]. In particular, any comparison in terms of the specific features of the constituent atoms (like the magnetic nature of the Gd ions and the non-magnetic nature of the Eu ions) will lose its meaning. However, a comparison in terms of the relative values of the physical parameters (ρ, T_c, H_{c2} etc) is still expected to be meaningful. So, below we shall limit ourselves to this type of comparison only. We found that this superconductor falls into the clean limit with a mean free path of 56 Å and a coherence length of 24.5 Å. We also estimated the zero-temperature upper critical field of $Eu_{1.5}$-Ru-1222 to be $H_{c2}(0) = 55$ T. A narrow-band approach is found to explain the observed thermoelectric power of $Eu_{1.5}$-Ru-1222 above 100 K in terms of two parameters, μ and W_σ. The values of μ and W_σ obtained from the fit of the theory with experimental data are $\mu = 8.0$ meV and $W_\sigma = 58.5$ meV.

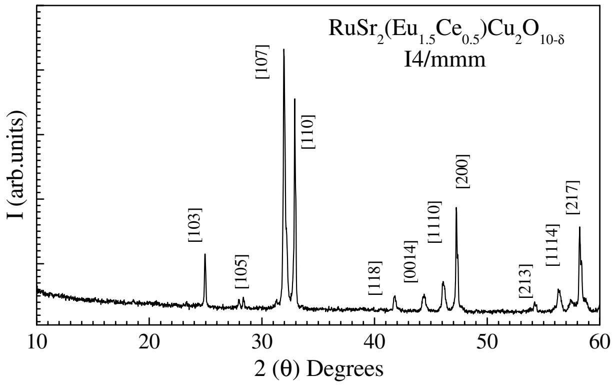

Figure 1. Room-temperature XRD patterns for $Eu_{1.5}$-1222 system.

2. Experimental details

The $RuSr_2Eu_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ sample was synthesized via a solid-state reaction route from RuO_2, SrO_2, Eu_2O_3, CeO_2 and CuO. Calcinations were carried out on the mixed powder at 1020, 1040 and 1060 °C each for 24 h with intermediate grindings. The pressed bar-shaped pellets were annealed in a flow of oxygen at 1075 °C for 40 h and subsequently cooled slowly over a span of another 20 h down to room temperature. X-ray diffraction (XRD) patterns were obtained using Cu Kα radiation. Magnetization measurements were performed on a SQUID magnetometer (Cryogenic Ltd, model S600). Resistivity measurements were made in the temperature range of 5–300 K under applied magnetic fields of 0–8 T using a four probe technique.

3. Results and discussion

3.1. X-ray diffraction

$Eu_{1.5}$-1222 copper oxide crystallizes in a tetragonal structure of space group $I4/mmm$ with the lattice parameters $a = b = 3.8378(2)$ Å, $c = 28.4849(2)$Å. An x-ray diffraction (XRD) pattern for the oxide is shown in figure 1. It is to be noted that a few unidentified lines are also seen in the XRD pattern, namely at 2θ of nearly 28° and 58°. Though we could not identify these, they are not from Ru-1212, $SrRuO_3$ or another possible culprit $RuSr_2EuO_6$. Most probably they arise from the superstructures of the tilted RuO_6 octahedra of the system [9, 16, 22]. The lattice parameters and quality of the XRD pattern is similar to earlier reported data for various Ru-1222 compounds [1, 2, 14–17]. Ru-1222 compounds are structurally related to the $CuA_2QCu_2O_{7-\delta}$ [Cu-$1^{(A)}2^{(Q)}12$ or Cu-1212, e.g. $CuBa_2YCu_2O_{7-\delta}$] phase with Cu in the charge reservoir replaced by Ru such that the Cu–O chain is replaced by a RuO_2 sheet. Furthermore, a three-layer fluorite-type block instead of a single oxygen-free R (=rare earth element) layer is inserted between the two CuO_2 planes of the Cu-1212 structure [15–17]. The oxygen content of the present sample is not determined, but it must be in line with our previous works (see [28]) in which we showed an oxygen content close to 9.60 for Ru-1222 samples having their T_c close to 30 K and no T_c with an oxygen content lower than say 9.40. With this reasoning, an oxygen content of nearly 9.60 is assumed for the current sample as its T_c is close to 30 K, as discussed in the coming sections.

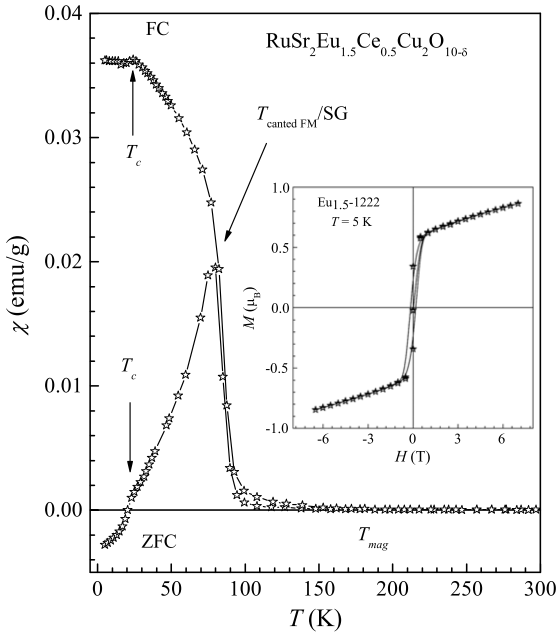

Figure 2. Behaviour of the magnetic susceptibility χ versus temperature T in the temperature range 5–300 K for $Eu_{1.5}$-1222. The inset shows the M–H curve at 5, 50 and 100 K for the same sample.

3.2. Magnetic behaviour

Figure 2 shows the behaviour of the magnetic susceptibility χ with temperature T in the temperature range 5–300 K for the $Eu_{1.5}$-1222 sample under an applied field of 5 Oe. Both types of measurement, the zero-field-cooled (ZFC) and the field-cooled (FC), are shown in this figure. The ZFC and FC curves start branching at 140 K from the higher-temperature side with a sharp upward turn at around 100 K. The branching of the FC and ZFC curves signifies the onset of the antiferromagnetic (AFM) effect. This means that there is a magnetic transition of the Eu–Ru-1222 system at $T_{mag} = 140$ K. On moving further towards the low-temperature side, it is seen that the ZFC branch shows a cusp at $T_{cusp} = 75$ K, a superconducting transition temperature at $T_{c,\chi} = 25$ K and finally a diamagnetic transition at $T_d = 20$ K. The down-turn cusp at 75 K in low fields is indicative of the onset of weak ferromagnetism (FM) or the spin glass nature of the Ru spins [18]. This weak FM effect appears due to canted antiferromagnetic spins of the Ru ions. The existence of weak FM below T_{cusp} is also seen in the FC branch. In fact, the FC curve increases fast at T_{cusp}, and then saturates at lower temperatures. This shows the presence of weak FM in the system.

Compared with $T_{mag} = 140$ K for the $Eu_{1.5}$-1222 system, the systems $Gd_{1.5}$-1222 [18], $Gd_{1.4}$-1222 [23] and $Sm_{1.4}$-1222 [24] have values of T_{mag} equal to 160, 175 and 150 K respectively. The corresponding values of $T_{c,\chi}$ for these systems are 45, 30 and 28 K, compared with $T_{c,\chi} = 25$ K for the $Eu_{1.5}$-1222 system. From these relative values of T_{mag} and $T_{c,\chi}$ we are unable to find whether there is any correlation between these quantities. The only thing we see is that the values of both T_{mag} and $T_{c,\chi}$ are lowest for the present $Eu_{1.5}$-1222 system.

To further elucidate the magnetic properties of the $Eu_{1.5}$-1222 superconductor we show isothermal magnetization (M) for various values of the applied field (H) at 5 K in the inset to figure 2. The isothermal magnetization as a function of magnetic field may be viewed as the

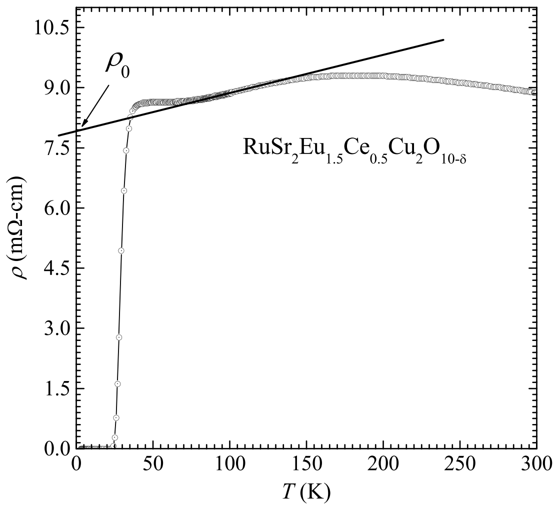

Figure 3. Behaviour of resistivity ρ versus temperature T for $Eu_{1.5}$-1222 up to $T = 300$ K.

sum of a linear part and a nonlinear part. That is to say, $M(H) = \chi H + \sigma_s(H)$. Here the linear contribution χH arises from the combined effects of the antiferromagnetic (spin glass) Ru spins and the paramagnetic Eu spins. $\sigma_s(H)$ represents the ferromagnetic component of the Ru moments. The appearance of $\sigma_s(H)$ at low temperatures within antiferromagnetic/spin–glass Ru spins could be due to a slight canting of the spins, as seen from neutron diffraction for another similar magnetosuperconductor Ru-1212 [8–10]. The contribution from the weak FM is clearly seen in the inset of figure 2 at 5 K. At $T > 100$ K, the occurrence of weak FM is not so sharp (plot not shown). In combination, these features are consistent with $T_{cusp} = 75$ K.

3.3. Magnetoresistivity and the upper critical field

In figure 3 we show the resistivity (ρ) of the $Eu_{1.5}$-1222 system up to $T = 300$ K. It is clear that between 75 and 140 K ρ shows a metallic behaviour. As mentioned above, the limited temperature range of metallic behaviour in ruthenocuprate occurs due to the decreasing oxygen content [18, 23, 24, 26]. In this sense we expect a linear metallic behaviour at all temperatures (above T_c) by the present $Eu_{1.5}$-1222 system if the oxygen content is increased in it. Since the temperature range for the metallic behaviour in the present case, 75–140 K, is a significant range, we do not expect much effect of either the low-T value of ρ or the remaining behaviour of high T on the middle linear portion of ρ. When this is so, we may fit the metallic portion of ρ by a straight line (cf. figure 3). Extrapolation of this straight line (marked in figure 3) to $T = 0$ K leads to the zero-temperature resistivity $\rho_0 = 7.9$ mΩ cm. Assuming that the plasma frequency and Fermi velocity of th $eEu_{1.5}$-1222 sample have the same values as for the $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ system [23], and using equation (2) of [23] ($l = 4.95 \times 10^{-4} \upsilon_F/(\hbar\omega_p)^2\rho$, where υ_F is the Fermi velocity, $\hbar$ is the Planck constant and ω_p is the plasma frequency), we estimate the mean free path of the Eu–Ru-1222 system to be $l = 56$ Å at $T = 0$ K. This is in between the 95 atm and 95 atm $- 2x$ samples of the $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ system of [23]. In fact, the 95 atm (95atm $- 2x$) sample of [23] corresponds to $l \sim 4$ Å (200 Å). From figure 3 we observe that below 75 K the resistivity increases with decreasing temperature. Since above 75 K (but up to 140 K) ρ shows a metallic behaviour, we may argue that the upturn of ρ below 75 K is due to weak localization.

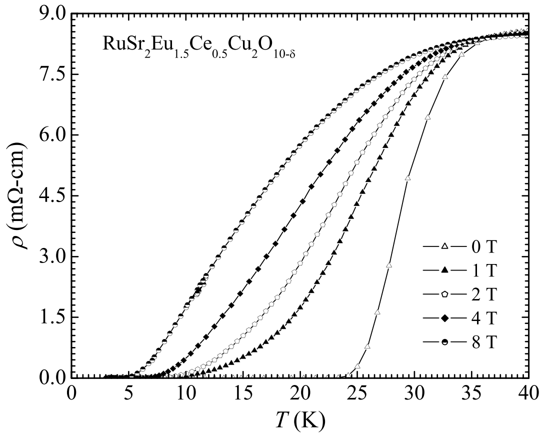

Figure 4. Magnetoresistance of a $Eu_{1.5}$-1222 sample for the magnetic field values of 0, 1, 2, 4 and 8 T in the temperature range of 5–40 K.

In figure 4 we show the magnetoresistance of our sample for magnetic field values of 0, 1, 2, 4 and 8 T in the temperature range of 0–40 K. From these values we estimate $T_c(H)$ for all the values of the magnetic field H from the intersection of the top of the transition line and the straight line passing through the linear portion of the ρ–T curve around the point of inflection near T_c. This method is quite familiar for estimating the temperature $T_c(H)$ for a given magnetic field in various cuprate superconductors [29]. On the basis of this method we find that $T_c(H = 0) = 33$ K. This is significantly higher than $T_{c\chi} = 25$ K. However, at the same time $T_c(H = 0)$ is significantly lower than the onset temperature of 43 K (for $H = 0$) obtained from the ρ versus T plot (figure 3). In the following description we shall treat $T_c(H)$ as the superconducting transition temperatures of the considered ruthenocuprate for different values of H.

The H_{c2} versus T curve, obtained in the above way, is shown in figure 5. First of all we see from figure 5 that H_{c2} (T) has a positive curvature in the observed range of temperatures. This agrees qualitatively with the H_{c2} (T) versus T behaviour found for the $RuSr_2Gd_{1.4}Ce_{0.6}Cu_2O_{10-\delta}$ superconductor by Escote *et al* [23]. It may be noted that positive curvature of $H_{c2}(T)$ is an essential feature of the cuprates, and is observed in these systems even at $T < 1$ K [29, 30]. In this sense it becomes imperative to consider such a T dependence of $H_{c2}(T)$ which leads to a positive curvature in the entire temperature range ($T = 0$ to T_c). In particular the form $H_{c2}(T) = H_{c2}(0)[1 - (T/T_c)^2]^\alpha$, considered by Escote *et al* [23], does not lead to positive curvature near $T = 0$ K, and so we feel that the value of $H_{c2}(0)$ obtained by Escote *et al* [23] is not reliable. The point is that even at zero curvature, the value of $H_{c2}(0)$ from the estimates of T_c and the slope $s(T_c) = -\mathrm{d}H_{c2}/\mathrm{d}T|_{T=Tc}$ turns out to be 44 T. Thus for a positive curvature $H_{c2}(0)$ will essentially be larger than 44 T.

In the present case we have very few points of the $H_{c2}(T)$ versus T relationship. So it is not possible to obtain a reliable value of $H_{c2}(0)$. However, since the $H_{c2}(T)$ versus T curve should have a positive curvature, the straight line extrapolation up to $T = 0$ of the slope $s(T_c)$ will give a lower limit of $H_{c2}(0)$ equal to 32 T. This is considerably lower than the corresponding value of 44 T for the $Gd_{1.4}$-1222 system [23]. Using the relation $H_{c2}(0) = \Phi_0/2\pi\xi(0)^2$, where Φ_0 is a flux quantum and $\xi(0)$ is the zero-temperature Ginzburg–Landau coherence length, we find that $\xi(0)$ will be less than 32 Å. This value of $\xi(0)$ is much smaller than the mean free path $l = 56$ Å estimated above. This means that the present sample of $RuSr_2Eu_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$

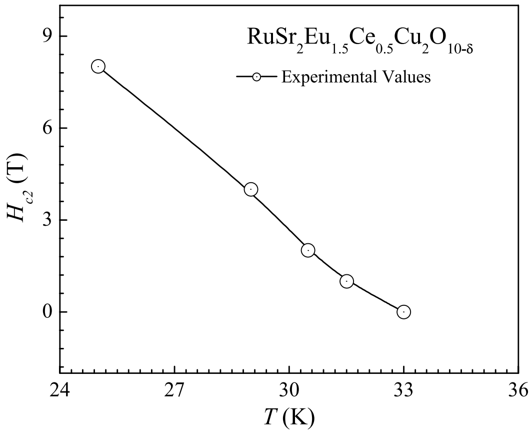

Figure 5. H_{c2} versus T plot for $Eu_{1.5}$-1222 compound.

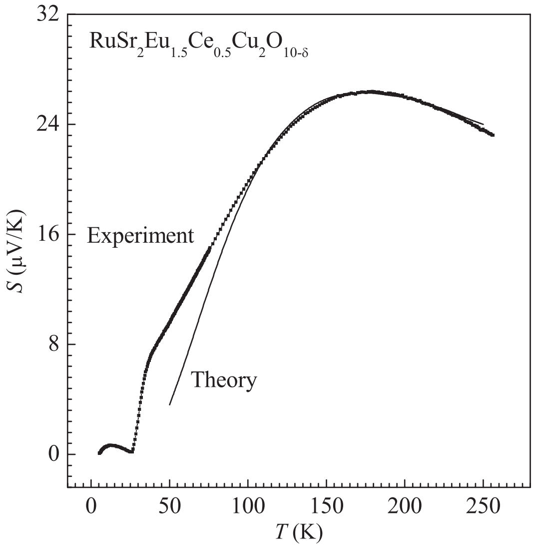

Figure 6. Plot of thermoelectric power S versus temperature T for a $Eu_{1.5}$-1222 sample. The solid line shows the fitted curve to equation (1).

is in the clean limit. In fact the positive curvature of H_{c2} (T) near T_c also indicates that the superconductor is in the clean limit.

3.4. Thermoelectric power

We show the experimentally observed thermoelectric power S in figure 6 by solid squares. We present an analysis of this thermoelectric power on the basis of the equation by Gasumyants *et al* [31]. This approach for calculating the transport properties of cuprate systems is based on the narrow-band picture. Gasumyants *et al* have obtained an expression for S which is expressed in terms of three parameters—band filling F, total effective band width W_D and the effective width of the energy interval W_σ for electron conduction. However, when we limit our study to the temperature range given by $2k_BT \ll W_D$, the approach of Gasumyants *et al* may be expressed in terms of just two parameters—the chemical potential μ and the effective width W_σ. Since typically $W_D > 100$ meV [31] and in our measurements $2k_BT < 45$ meV, we can use a two-parameter (μ and W_σ) version of the approach of Gasumyants *et al*. In this sense, we may rewrite equation (26) of Gasumyants *et al* as,

$$S = \left(\frac{k_B}{e \sinh W_\sigma^*}\right)\left[W_\sigma^* \sinh \mu^* + \mu^*\left[\cosh \mu^* + \exp(W_\sigma^*)\right] + \left[\cosh \mu^* + \cosh W_\sigma^*\right] \times \ln\left(\frac{1+\exp(W_\sigma^* - \mu^*)}{1+\exp(W_\sigma^* + \mu^*)}\right)\right]. \quad (1)$$

Here, $\mu^* = \mu/k_BT$ and $W_\sigma^* = W_\sigma/k_BT$. Although mathematically equation (1) is equivalent to equation (26) of Gasumyants *et al*, it (equation (1)) clarifies that S is an odd function of μ i.e. $S(-\mu) = -S(\mu)$. This property of S is not obvious from equation (26) of Gasumyants *et al*.

We find that equation (1) fits the experimental data very well (figure 6) for $\mu = 8.0$ meV and $W_\sigma = 58.5$ meV, except below 100 K. The deviation of equation (1) from the observed values below 100 K may be attributed to fluctuation effects. For comparison we have also fitted the thermoelectric power data of Liu *et al* [25] for the $RuSr_2GdCu_2O_8$ superconductor on the basis of equation (1). It is found that $\mu = 28.0$ meV and $W_\sigma = 77.0$ meV give an excellent fit. This shows that the ratio W_σ/μ is larger in the present case ($W_\sigma/\mu = 7.31$) than in the case of Liu *et al* ($W_\sigma/\mu = 2.75$). This, according to [31], means that the resistivity of the sample of Liu *et al* must be significantly larger than that of our sample. This is indeed the case, as the (weakly) metallic properties of the $RuSr_2GdCu_2O_8$ sample of Liu *et al* correspond to $\rho_0 = 44$ mΩ cm (or to l = 10 Å), compared with $\rho_0 = 7.9$ mΩ cm for the present case of the $Eu_{1.5}$-1222 system. On this basis we may argue that the values of μ and W_σ estimated here provide a consistent understanding of the behaviour of the thermoelectric power.

4. Conclusions

We have synthesized the $Eu_{1.5}$-1222 system, and have measured its various properties. The susceptibility measurements show magnetic order in this system below 140 K and a superconducting transition at 25 K. The resistivity of the $Eu_{1.5}$-1222 sample shows a metallic behaviour between 75 and 140 K. From this we estimate a mean free path of 56 Å at zero temperature. The upper critical field of the $Eu_{1.5}$-1222 system shows a positive curvature, and corresponds to a minimum value of the upper critical field equal to 32 T at $T = 0$ K. This value is found to be lower than the corresponding value for the $Gd_{1.4}$-1222 system of [23]. This is in accordance with expectations since the T_c of $Eu_{1.5}$-1222 system is lower than that of the $Gd_{1.4}$-1222 system. A narrow-band approach is found to explain the observed thermoelectric power of $Eu_{1.5}$-1222 above 100 K in terms of two parameters, μ and W_σ. The values of μ and W_σ obtained from the fit of the theory with experimental data provides $\mu = 8.0$ meV and $W_\sigma = 58.5$ meV.

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