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Complex magnetic order in $EuRh_2Si_2$

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Abstract

We present resistivity, magnetoresistance, magnetisation and heat capacity results on $EuRh_2Si_2$. Magnetic susceptibility and specific heat data confirm the reported magnetic transition at 25 K and reveal an additional magnetic transition at 23.5 K. Isothermal magnetization reveal a metamagnetic transition at a rather low field, $B_m \approx 0.1$ T. The resistivity data show a large decrease of ρ at the lowest transition at 23.5 K, and the metamagnetic transition lead to a large negative magnetoresistance at 2 K. The temperature dependence of the specific heat below the transition is unusual, being proportional to T over a significant temperature range.

Keywords: Magnetically ordered materials; X-ray diffraction; Electronic transport; Magnetisation; Heat capacity

1. Introduction

Because of the instability of the Eu-4*f*-shell, Eu based intermetallic compounds present different types of ground states, ranging from trivalent Eu showing Van Vleck susceptibility at low temperature to mixed valent and finally divalent Eu, showing magnetic order at low temperature as in the homologous Gd compounds. Within the EuT_2Si_2 (T=transition element) series of compounds, the observed ground state evolves systematically with the position of the T-element in the periodic table, from trivalent to divalent as one moves from left to right or top to bottom. $EuRh_2Si_2$ is an exception to this systematic [1,2], since it is divalent despite being positioned between trivalent $EuRu_2Si_2$ and mixed valent $EuPd_2Si_2$. Since unusually high magnetic ordering temperature (T_N~37 K) in $CeRh_2Si_2$ [3] and a non Fermi liquid (NFL) behavior were found in $YbRh_2Si_2$ [4], one can expect to find interesting magnetic behavior in $EuRh_2Si_2$ as well. From Mössbauer spectroscopy and susceptibility data, $EuRh_2Si_2$ was claimed to be primarily a divalent compound with about 25% mixed valent component which orders magnetically at 25 K [1]. A later Mössbauer investigation showed that $EuRh_2Si_2$ is purely a divalent compound since the mixed valent line was absent in the sample [2]. To gain further knowledge on the magnetic behavior of $EuRh_2Si_2$, we have prepared polycrystalline sample and investigated the magnetic susceptibility, magnetization, resistivity, specific heat and magnetoresistance of this compound.

2. Experimental

Polycrystalline samples were prepared by heating constituent elements in a sealed Ta-crucible and subsequent annealing at 800°C for 3 days. Resistivity and magnetoresistance measurements were carried out using four probe ac technique. Magnetisation measurements were performed using a commercial SQUID magnetometer. The heat capacity was measured using the relaxation method in a Physical Property Measurement System (PPMS-Quantum Design).

3. Results and discussion

Powder X-ray diffraction reveals that $EuRh_2Si_2$ crystallizes in $ThCr_2Si_2$ type structure with lattice parameters $a=4.082$ Å and $c=10.202$ Å, which are in agreement with those in Ref. [2]. The lattice parameters of the isostructural $GdRh_2Si_2$ are $a=4.043$ Å and $c=9.988$ Å. Thus the unit cell volume of $EuRh_2Si_2$ is about 4% larger and c/a is ~1% larger compared to $GdRh_2Si_2$. On the other hand the

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unit cell volume of $EuRh_2Si_2$ is 2.4% larger and c/a is 8% larger compared to $GdRu_2Si_2$.

At high temperature the magnetic susceptibility follows a Curie–Weiss behavior with effective magnetic moment $\mu_{eff}=7.25\ \mu_B$ and Curie–Weiss temperature $\theta_p=24$ K. The magnetic susceptibility data at low temperature for different field values are shown in Fig. 1. At 100 G two prominent anomalies at 25 and 23.5 K and a broad anomaly at ~8 K are seen. The magnetic susceptibility is almost temperature independent between 25 and 23 K followed by a large drop below 23 K. At $B=0.1$ T the anomalies at 25 and 23 K merged together giving rise to a single antiferromagnetic-like peak in the susceptibility at $T=23$ K whereas the small anomaly at ~8 K is much more pronounced and shifted to a higher temperature. At $B=1$ T the susceptibility data appears more like that of a ferromagnet suggesting the presence of metamagnetic transition in this compound. It is clear from the data that the transition at ~8 K is very much field dependent and is not observable in the susceptibility data measured at 1 T. It is to be noted that though θ_p is positive the magnetic ordering at low field seems to be of antiferromagnetic nature with $T_N \approx \theta_p$. This is possibly due to the fact that within the rare earth plane the Eu ions undergo ferromagnetic ordering and the planes are antiferromagntically coupled as has been observed in many magnetic compounds with $ThCr_2Si_2$-type structure [5]

In the magnetically ordered state the isothermal magnetization at 2 K reveals a field induced metamagnetic transition at low field (see Fig. 2). The critical field at which the metamagnetic transition takes place is estimated from the dM/dB vs. B curve to be $B_m \approx 0.1$ T. The saturation magnetisation at 5.5 T is 5.6 μ_B (Fig. 2) which is ~80% of the saturation value expected for divalent Eu (7 μ_B). There is a slight nonlinearity in the magnetisation curve even at 30 K (not shown in figure), which suggests presence of short range order above the magnetic ordering temperature.

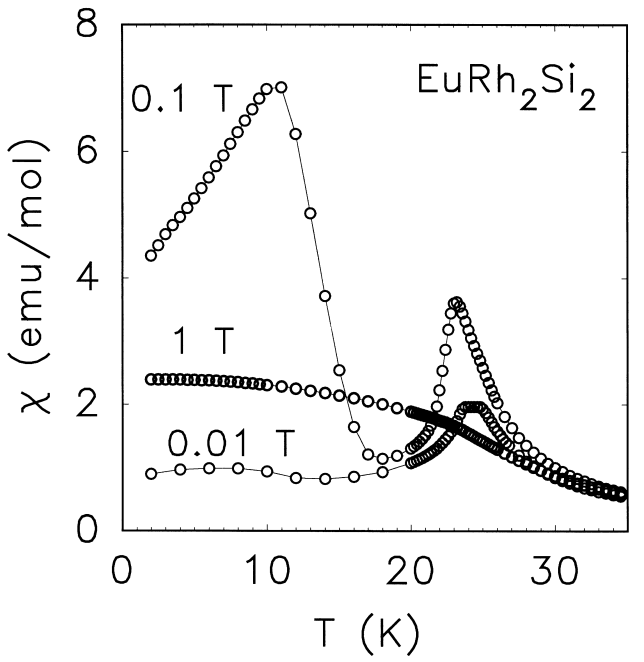

Fig. 1. DC magnetic susceptibility measured at different applied fields for $EuRh_2Si_2$.

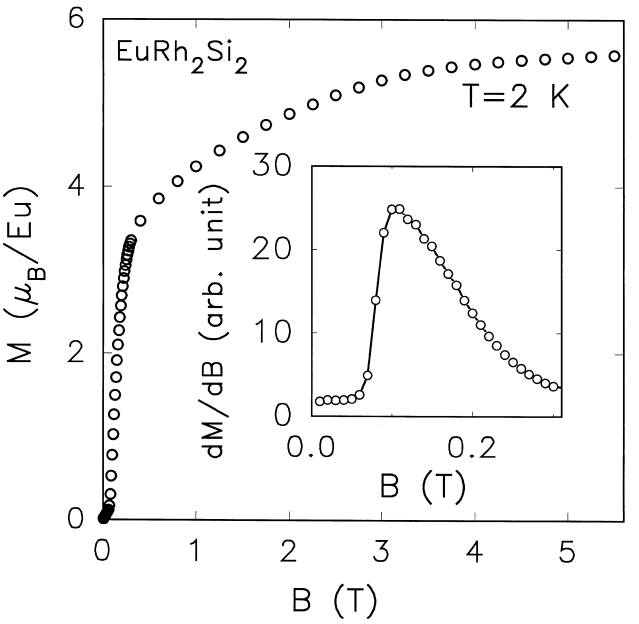

Fig. 2. Magnetisation as a function of externally applied field at 2 K. Inset shows dM/dB vs. B curve.

It is to be noted that the isothermal magnetisation of $EuRh_2Si_2$ is more like that of $GdRu_2Si_2$ which also shows metamagnetic transition [6]. $GdRh_2Si_2$ does not show metamagnetic behavior [7]. Further the magnetic ordering temperature of $EuRh_2Si_2$ ($T_N=25$ K) is much lower compared to that of $GdRh_2Si_2$ ($T_N=106$ K) [8]. Since Gd is in trivalent state whereas Eu is in divalent state in RRh_2Si_2 series these results show that the band filling drastically affect the density of states at the fermi level and the exchange-interaction. From the point of view of the band filling the properties of $EuRh_2Si_2$ is expected to be between $GdRh_2Si_2$ (which has one electron more because Gd gives one more electron than Eu to the conduction band) and $GdRu_2Si_2$ (which has one electron less since Gd gives one electron more but each Ru donates one electron less to the conduction band). We suggest two possible reasons for the low T_N of $EuRh_2Si_2$ compared to that of $GdRh_2Si_2$: (i) small 4*f*-conduction electron coupling in $EuRh_2Si_2$ than in $GdRh_2Si_2$. This was realized in La–Eu and La–Gd system [9]; (ii) a large enhancement of the density of states at the fermi level for $GdRh_2Si_2$ compared to $EuRh_2Si_2$. This assumption is justified by the finding that in GdT_2Si_2 (T=3d, 4d or 5d elements) compounds maxima of T_N are found for the compounds with T=Co, Rh, Ir (see Fig. 3) which are in the same column in the

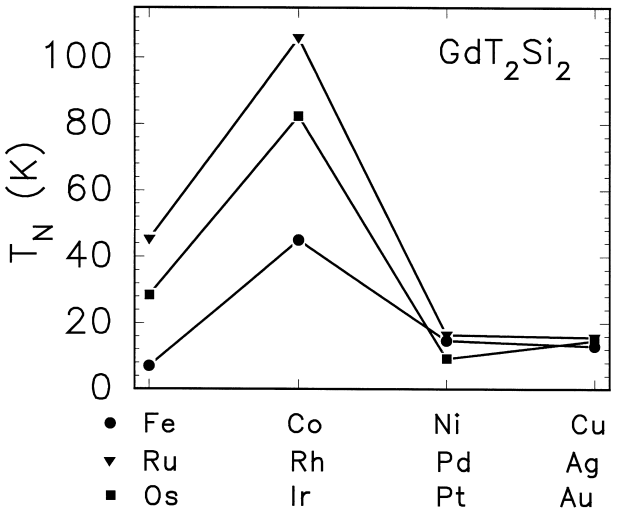

Fig. 3. Neel temperature, T_N, of GdT_2Si_2 (T=3d, 4d or 5d elements) taken from Ref. [8].

periodic table whereas much lower T_N were found for compounds with other transition elements [8].

The temperature dependence of the resistivity is shown in Fig. 4. The residual resistivity ratio $\rho(300)/\rho(1.8)=20$ attests good quality of our sample. Resistivity shows large decrease below the second AF transition at 24 K due to reduction of the spin disorder scattering. $EuRh_2Si_2$ shows an almost linear decrease of ρ down to T_N, whereas the nonmagnetic homologue $LuRh_2Si_2$ shows a pronounced positive curvature in this temperature range. Linear temperature dependence of resistivity over a wide range of temperature have also been observed in the normal state of high T_c-oxides which is believed to be due to presence of antiferromagnetic spin fluctuation [10]. It is not clear at present the reason for the linear temperature dependence of resistivity in $EuRh_2Si_2$. Linear temperature dependence of resistivity have also been found in $EuRh_2P_2$ (T_N=50 K) [11] and in $EuNi_2Ge_2$ [12].

Heat capacity shows large anomalies at ~23 and ~25 K (Fig. 5) confirming that both transitions are intrinsic to the material. We, however, do not see λ-type anomaly in the heat capacity data at ~7 K. We may recall that in $TbNi_2B_2C$ which orders antiferromagnetically at ~15 K a spin reorientation takes place at ~8 K. In this case, though large anomaly was seen in the susceptibility measurement at low field, no clear anomaly was seen in the heat capacity data [13,14]. This type of situation may exist in $EuRh_2Si_2$ as well. An interesting observation of the heat capacity data is the presence of a large T-linear term in the magnetic part of the heat capacity data which manifests itself as a plateau in the C/T vs. T data. Such a plateau was also found in EuB_6 which is a ferromagnet and in $Gd_2Fe_3Si_5$ which undergo antiferromagnetic ordering. In the case of EuB_6 the unusual behavior of the heat capacity was interpreted to arise from a combination of contributions of the ground state multiplet of the Eu ion and magnetic excitations in the magnetically ordered state [15]. T-linear in heat capacity of $Gd_2Fe_3Si_5$ was interpreted to be due to spin-wave type of magnetic ordering [16]. A linear term in the heat capacity could be expected either from two dimensional ferromagnetic spin waves or from one dimensional antiferromagnetic spin waves. Since the magnetic ordering in $EuRh_2Si_2$ seems to be of antiferromagnetic type but the exact magnetic structure is not known we can only speculate about the spin-wave contributions to the heat capacity of $EuRh_2Si_2$. The magnetic entropy at T_N is $\sim 0.9R \ln 8$ which is close to $R \ln 8$, the value expected for Eu^{2+} ions.

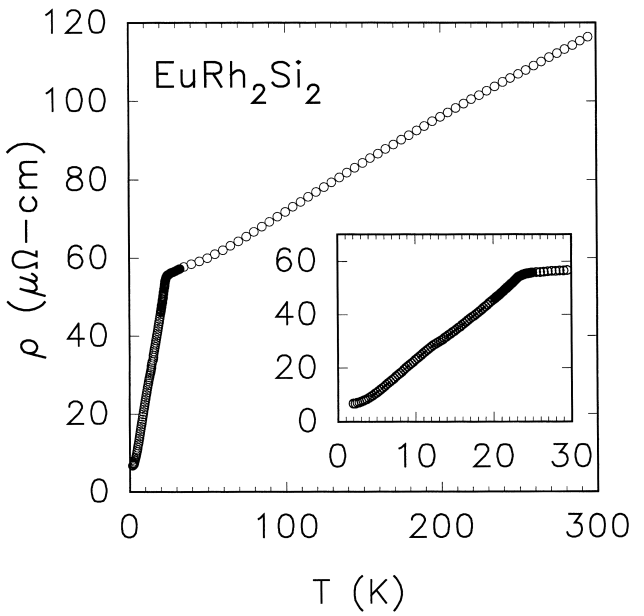

Fig. 4. Electrical resistivity vs. temperature for $EuRh_2Si_2$. Inset shows the expanded view at low temperature region.

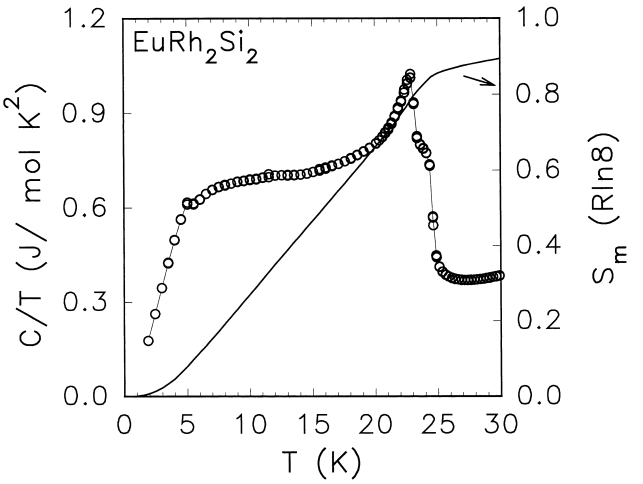

Fig. 5. Heat capacity plotted as C/T vs. T and magnetic entropy for $EuRh_2Si_2$. Note two anomalies due to magnetic transitions at 23 and 25 K and a T-linear part in the heat capacity data which is reflected as a plateau in the C/T vs. T curve.

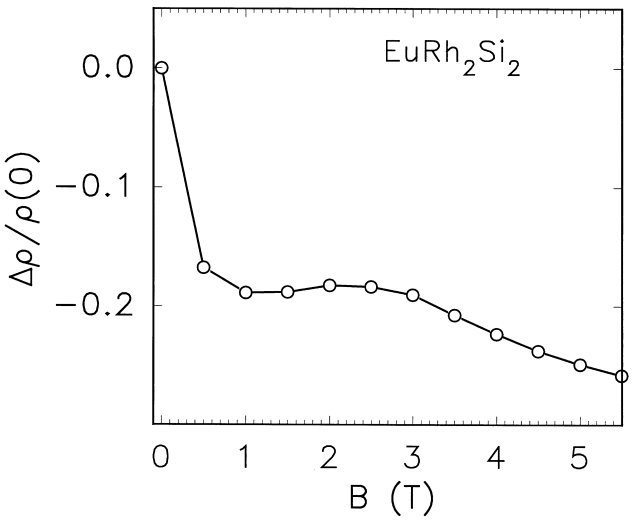

Fig. 6. Normalized resistivity at 2 K as a function of magnetic field. A large negative magnetoresistance due to metamagnetic transition is seen.

The normalized resistivity at 2 K as a function of field is shown in Fig. 6. At low field the resistivity shows a sharp drop as the field is increased. The normalised resistivity shows a hump at ~2.5 T. The magnetoresistance at 2 K is negative due to the presence of metamagnetic transition (Fig. 2). The negative magnetoresistance at 2 K and 5.5 T is ~26% which is rather large. The reason for the hump at ~2.5 T is not clear at present.

4. Conclusion

To summarize, we have shown that $EuRh_2Si_2$ undergoes multiple magnetic transition and shows metamagnetic transition. It has large negative magnetoresistance related to this metamagnetic transition. The presence of multiple magnetic transitions and unusual heat capacity suggest complex nature of the magnetic order which deserves further investigation.

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