Enhanced conduction band density of states in intermetallic EuTSi$_3$ (T = Rh, Ir)

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Abstract

We report on the physical properties of single crystalline EuRhSi$_3$ and polycrystalline EuIrSi$_3$, inferred from magnetization, electrical transport, heat capacity and $^{151}$Eu Mössbauer spectroscopy. These previously known compounds crystallise in the tetragonal BaNiSn$_3$-type structure. The single crystal magnetization in EuRhSi$_3$ has a strongly anisotropic behaviour at 2 K with a spin-flop field of 13 T, and we present a model of these magnetic properties which allows the exchange constants to be determined. In both compounds, specific heat shows the presence of a cascade of two close transitions near 50 K, and the $^{151}$Eu Mössbauer spectra demonstrate that the intermediate phase has an incommensurate amplitude modulated structure. We find anomalously large values, with respect to other members of the series, for the RKKY Néel temperature, for the spin-flop field (13 T), for the spin-wave gap ($\approx$20–25 K) inferred from both resistivity and specific heat data, for the spin-disorder resistivity in EuIrSi$_3$ (240 $\mu$Ω cm) and for the saturated hyperfine field (52 T). The enhanced values of the quantities that depend on the electronic density of states at the Fermi level, imply that the latter must be strongly enhanced in these two materials. EuIrSi$_3$ exhibits a giant magnetoresistance ratio, with values exceeding 600% at 2 K in a field of 14 T.

Keywords: antiferromagnetic materials, EuRhSi$_3$, EuIrSi$_3$, enhanced conduction band density of states

(Some figures may appear in colour only in the online journal)

1. Introduction

Divalent-Eu intermetallic compounds order magnetically due to the indirect RKKY exchange interaction [1] between the Eu 4$f$-spins. Several Eu-based compounds with composition EuTX$_3$, where T is a $d$-transition element and X = Si or Ge, crystallizing in the non-centrosymmetric BaNiSn$_3$-type structure are known. Of these, the magnetic properties of EuPtSi$_3$ [2], EuTGe$_3$ (T = Co, Ni, Rh, Pd, Ir and Pt) [3–7] have recently been reported in the literature. In these materials, an anisotropic behaviour of the 2 K magnetization seems to be the prerequisite for the existence of a cascade of closely spaced magnetic transitions around 15 K; a transition from the paramagnetic to an incommensurate, moment modulated antiferromagnetic (AF) state occurs first, followed by another one to a single moment regular AF state, a few K below. This is the case for EuPtSi$_3$ and EuNiGe$_3$, whereas EuPtGe$_3$ shows a single transition and a nearly isotropic behaviour of the magnetization.

Here, we report on the magnetic properties of iso-structural EuTSi$_3$ (T = Rh and Ir) compounds, with a single crystal sample for EuRhSi$_3$ only. An early Mössbauer spectroscopy study of these two compounds was performed in [8]. We show that these two materials belong to the ‘transition cascade’ type, with an anisotropic behaviour of the magnetization documented for EuRhSi$_3$. We present in addition specific heat, transport and $^{151}$Eu Mössbauer spectroscopy data. We find that the magnetic and transport properties in these two materials are notably enhanced with respect to those in other members of the series, and we show that this enhancement...
can be attributed to an unusually large density of conduction electron states at the Fermi level $n(E_F)$. As a remarkable result, the values of the spin-wave gap derived from such different techniques as resistivity, specific heat and single crystal magnetization measurements are in good agreement.

2. Experimental

Polycrystalline samples of EuIrSi$_3$ and EuRhSi$_3$ were prepared by melting Eu (99.9% purity), Ir/Rh (99.99%) and Si (99.999%) in an arc furnace under an inert argon atmosphere. The alloy buttons were flipped over three times and re-melted to ensure proper homogenization. An excess of about 10% over the stoichiometric amount was taken for Eu, after weight loss due to sublimation of Eu was found in a trial run. Polycrystalline samples of non-magnetic reference compounds LaRhSi$_3$ and LaIrSi$_3$ were also prepared by arc melting. Single crystal growth of the two Eu compounds was tried using Sn and In as flux and following the same protocol as reported in [2, 3]. Powder-diffraction spectra were recorded on a Phillips Pan-analytical set up using Cu-$K_x$ radiation. The magnetization as a function of field (up to 16 T) and temperature (1.8–300 K) was measured using Quantum Design MPMS and VSM magnetometers. The electrical resistivity between 1.8 and 300 K in zero and applied fields, and the heat capacity were measured in a Quantum Design PPMS set-up. $^{151}$Eu Mössbauer spectra were recorded at various temperatures using a commercial $^{151}$SmF$_3$ source mounted on a constant acceleration spectrometer.

3. Results and discussion

3.1. Structure

Our attempt to grow single crystals succeeded only for EuRhSi$_3$, with In as flux. The size of the crystals was small and we could not cut a specimen with well defined geometry, precluding us from a quantitative measurement of the electrical resistivity. Powder diffraction spectra of EuIrSi$_3$ and EuRhSi$_3$ could be indexed on the basis of the BaNiSn$_3$ type structure (space group $I4mm$). The powder x-ray diffraction spectra of EuRhSi$_3$ and EuIrSi$_3$ along with the Rietveld refinement are shown in figure 1. The presence of a parasitic impurity phase in EuIrSi$_3$ is indicated by the peak marked with an asterisk. The lattice parameters obtained by the Rietveld analysis of the powder diffraction spectra are in good agreement with the previously reported values [8].

3.2. Susceptibility and isothermal magnetization

The susceptibility of EuRhSi$_3$ measured in a field of 0.1 T applied along [001] and [100] is shown below 100 K in figure 2. A clear peak near 49 K shows the onset of the AF phase. The close-up in the inset of figure 2 shows that this peak is split (arrows). In agreement with the heat capacity and $^{151}$Eu Mössbauer data (vide infra), these two peaks correspond to closely spaced magnetic transitions near 48 and 46 K. The susceptibility at high temperature (not shown) is nearly isotropic and a fit of the $1/T$ versus $T$ data to a Curie-Weiss law furnishes effective moments $\mu_{\text{eff}} = 7.4$ and 7.52 $\mu_B$ and paramagnetic Curie temperature $\theta_p = -11$ and $-14$ K for H along [001] and [100], respectively. These effective moments are lower than the free ion value of $7.94 \mu_B$ expected for Eu$^{2+}$ ($g = 2, S = 7/2$), which is due either to the presence of residual In-flux or to a slight Eu off-stoichiometry, corresponding to about 10 at.% Eu deficit. As to polycrystalline EuIrSi$_3$, its susceptibility is shown between 30 and 60 K in the inset of figure 2: two anomalies (arrows) are also present, near 52 and 43 K, witnessing the same phenomenon as in EuRhSi$_3$. The $\theta_p$ value is $-17$ K, while $\mu_{\text{eff}} = 7.7 \mu_B$ is closer to the Eu$^{2+}$ free ion value.

The Néel temperature $T_N$ in rare earth intermetallic compounds is the result of the indirect RKKY exchange between 4$f$ spins $S$ mediated by the 4$f$-conduction electron coupling with constant $J_d$:

$$T_{N(d)} = -J_d s.s,$$

where $s$ is the conduction electron spin density, and it has the form: $T_{N(d)} \propto J_d^2 n(E_F) S(S + 1)$ [1]. Its value in these two compounds, near 50 K, is much larger than in other members of the series (≈15 K), pointing to an enhanced value of $n(E_F)$.
The isothermal magnetization versus field scan at 2 K in EuRhSi$_3$, shown in figure 3(a), is a textbook example of an antiferromagnet with the tetragonal [001] c-axis as the easy axis of magnetization and the (001) ab-plane as the hard plane, in line with the susceptibility data of figure 2. For H $\parallel$ [001], an unusually large spin-flop field of 13 T is observed. A linear extension of the magnetization curves for both field directions up to a saturation moment value $m_0 = 7 \mu_B$ yields large spin-flip fields $H_{sf}^c \approx 28$ T and $H_{sf}^a \approx 41$ T. The latter is larger since the field aligns the moments in the hard magnetic plane. In the standard molecular field theory [9], one has: $H_{sf}^c = 2(H_e - H_a)$ and $H_{sf}^a = 2(H_e + H_a)$, where $H_e$ and $H_a$ are, respectively, the exchange and the ‘anisotropy’ field, the latter being defined as $H_a = K/m_0$ where $K$ is the anisotropy energy density. Then we obtain $H_a = 17$ T and $H_e = 3.35$ T, and the critical spin-flop field $H_{sf} = 2\sqrt{H_e(H_e - H_a)} = 13.5$ T, in very good agreement with experiment. The spin-flop field in EuRhSi$_3$ is much larger than in EuNiGe$_3$ [6] (2–3 T), a logical consequence of a large $H_e$ linked to the high $T_N$ value.

In the inset of figure 3(a) are plotted the magnetization curves along the easy axis at selected temperatures. The magnetization at 10 K nearly overlaps with the 2 K data. At higher temperatures, the spin-flop field decreases and the jump at the spin-flop broadens.

The isothermal field scans for EuIrSi$_3$ at various temperatures are shown in figure 4. The magnetizations at 2 and 8 K virtually coincide. At 14 T the magnetization at 2 K is smaller than the corresponding value in EuRhSi$_3$ for H along the hard direction [100], suggesting a larger spin-flop field in the Ir-compound.

### 3.3. Model for computing magnetization

However, such a simple model as described above cannot account for the quite different values of $T_N$ and $\mu_B$ and the oscillatory nature of the RKKY exchange compels one to introduce at least two different exchange integrals. Therefore, we use a numerical self-consistent calculation which has been described thoroughly in [6]: (i) the infinite range dipolar interaction is added, (ii) two exchange integrals $J_1$ (intra-plane...
first neighbor) and $J_z$ (interplane first neighbor) for the centered tetragonal structure are considered, as well as exchange anisotropy, and (iii) the single-ion crystalline anisotropy is described by a term $DS^2$, where $O_2 = c$. We also assume a magnetic structure made of ferromagnetic (ab) planes ($J_1 > 0$) coupled antiferromagnetically along c ($J_2 < 0$), i.e. a propagation vector $k = [001]$. To obtain a first estimation of the exchange constants, the molecular field equations linking $T_N$ and $\theta_0$ with $J_1$ and $J_2$ are used [5, 6], yielding: $J_1 = 1.1$ K and $J_2 = -0.88$ K. We have taken $T_N = 60$ K (see the section about Mössbauer spectroscopy) and a mean value $\theta_0 = -13$ K. Figure 3(b) shows the curves which reproduce best the experimental data, with a small exchange anisotropy: $J_1^g = 0.8$ K, $J_2^g = 1.1$ K, $J_z^g = -0.7$ K, $J_z^s = -0.9$ K and a crystalline anisotropy parameter $D = -0.85$ K. The model yields $T_N = 57$ K and $\theta_0 = -6$ and $-17$ K for $H \parallel [001]$ and $H \parallel [100]$, respectively (experimental values $-11$ and $-14$ K resp.). At 30 K and above, the model cannot exactly reproduce the smoothing of the spin-flop transition (see inset of figure 3(b)).

### 3.4. Electrical resistivity

The electrical resistivity of EuIrSi$_3$ and EuRhSi$_3$ in zero and 14 T (and of non-magnetic LaIrSi$_3$) is shown in figures 5(a) and (b). The resistivity of EuRhSi$_3$ is plotted as $R(T)/R(300$ K) due to a large uncertainty in the geometrical factor. Above $T_N$ the resistivity varies almost linearly up to 300 K in both compounds due to phonon scattering. It shows a small upturn at the magnetic transitions, likely caused by antiferromagnetic fluctuations just above $T_N$ and decreases rapidly on cooling due to a strong depletion in the spin-disorder scattering in the magnetically ordered state. The residual resistivity $\rho_0 = (1.8$ K) is $3.9 \mu\Omega$ cm and the residual resistivity ratio, RRR, defined as $R(300$ K)/$R(1.8$ K), is 94 in EuIrSi$_3$ indicating it is chemically a well ordered compound. In contrast, the RRR in EuRhSi$_3$ is close to 20 and 10 for $J \parallel [100]$ and [001], respectively. Our resistivity data in EuRhSi$_3$ are in good agreement with those reported in [10], which reports a pressure induced valence transition in this compound.

The total spin disorder resistivity $\rho_{sd}$ in EuIrSi$_3$, estimated by extrapolating the high temperature (above $T_N$) linear portion of resistivity plot to $T = 0$, amounts to about 240 $\mu\Omega$ cm while it is of the order of a few $\mu\Omega$ cm in the other compounds of the series [2, 3, 6]. It cannot be attributed to disorder or grain boundary effects as the residual resistivity at 1.8 K is just 3.9 $\mu\Omega$ cm. Since $\rho_{sd} \propto J_1^g \langle m(E_F)S(S+1) \rangle$ [1], its enhanced value in EuRhSi$_3$ is in line with a stronger $m(E_F)$. From the heat capacity data (vide infra) a similar conclusion is also drawn for EuRhSi$_3$. An expression for the spin-disorder resistivity $\rho_{sd}$ given in [11] relates $\rho_{sd}$ to the effective electronic mass $m$ by the relation: $m/m_0 = 7.7(\rho_{sd}/\theta_0)$, where $m_0$ represents the free electron mass. A high electronic density of states is equivalent to an enhanced effective mass $m$ and results in a larger $\rho_{sd}$.

In the AF phase below 20 K, magnon scattering is the dominant mechanism for resistivity. With an antiferromagnetic spin wave dispersion relation:

$$E(k) = \sqrt{\Delta^2 + \sigma k^2}$$

where $\Delta$ is the anisotropy gap and $\sigma$ the spin-wave stiffness, the electrical resistivity in zero field for $T < \Delta$ is given by [12]:

$$\rho(T) = \rho_0 + A\Delta^2 \left( \frac{T}{\Delta} \right)^{1/2} e^{-\Delta/T} \left[ 1 + 2/(3 \left( \frac{T}{\Delta} \right) + 2/15 \left( \frac{T}{\Delta} \right)^2 \right],$$

where $A$ is a material dependent constant. Equation (3) provides a good fit to the zero field data in both compounds between 1.8 and 15 K (and also for a 14 T field), as shown by the solid lines in the insets of figure 5. In EuIrSi$_3$, the zero field spin wave gap is 24.9 K and it is 23.7 K in EuRhSi$_3$. The AF spin wave gap is linked to the exchange and anisotropy fields $H_x$ and $H_y$ by the relation [13]:

$$\Delta = 2g\mu_B \sqrt{H_x (H_x + H_y)}.$$

Using the values determined above in EuRhSi$_3$: $H_x = 17$ T, $H_y = 3.35$ T and $g = 2$, one obtains $\Delta = 22.1$ K, in excellent agreement with experiment in this material and close to the value in EuIrSi$_3$.

At a field of 14 T, $\rho_1$ increases to 27.04 $\mu\Omega$ cm in EuIrSi$_3$. Assuming that equation (3) describes the temperature.
dependence of resistivity in applied field, we obtain $\Delta = 20.8$ and 22.2 K in EuIrSi$_3$ and EuRhSi$_3$ in 14 T, respectively.

The significant increase of $\rho$ with field corresponds to a positive magnetoresistance ratio $MR$, defined as $MR = [\rho(H) - \rho(H = 0)]/\rho(H = 0)$, as expected for antiferromagnets [14, 15]. The plots of $MR$ at selected temperatures in EuIrSi$_3$ are shown in figure 6(a). It reaches giant values: at 14 T, it exceeds 100% at 10 K and it reaches 600% at 2 K. At higher temperatures (not shown), $MR$ decreases and becomes negative near 47 K, shows an absolute minimum near 50 K.

3.5. Heat capacity

The main panels of figure 7 show the heat capacity of EuIrSi$_3$ and EuRhSi$_3$ together with that of the non-magnetic La-reference. The two plots show two major peaks at 51.8 and 43.1 K in EuIrSi$_3$ and at 48.3 and 45.8 K in EuRhSi$_3$ in close correspondence with the anomalies seen in the susceptibility data, thus confirming the occurrence of two magnetic transitions in these two compounds. The jump in the heat capacity at the higher transition temperature $T_{N1}$ is approximately 9.5 J mol$^{-1}$ K and 12 J mol$^{-1}$ K in the Ir and Rh-compound, respectively, which is lower than the value $\delta C_{T2} = 20.14$ J mol$^{-1}$ K for an equal moment antiferromagnetic transition for $S = 7/2$ ions in the mean field model, and closer to that predicted for an amplitude modulated structure (2/3 $\delta C_{T2})$ [2, 16]. This suggests that at $T_{N1}$ occurs a transition to a modulated moment structure. The transition from this intermediate structure to an equal moment structure takes place at $T_{N2}$. This behaviour is confirmed by the $^{151}$Eu Mössbauer spectra recorded at few selected temperatures.

Deep in the AF phase, the specific heat should be the sum of 3 terms [17]:

$$C(T) = \gamma T + \beta T^3 + B \Delta^3 \left( \frac{T}{\Delta} \right)^{1/2} e^{-\Delta/T} \times \left[ 1 + 39/20 \left( \frac{T}{\Delta} \right) + 51/32 \left( \frac{T}{\Delta} \right)^2 \right].$$

where the first linear term is the conduction electron heat capacity, the second is the phonon contribution and the third the magnon heat capacity corresponding to a dispersion.
law given by equation (2) with a gap $\Delta$. In the fitting process for $T < 10$ K, the coefficient $\beta$ was kept fixed, equal to that derived from the low temperature heat capacity of corresponding La-analog. Equation (5) provides a good fit of the data between 1.8 and 10 K, shown by the solid line in the insets of figures 7(a) and (b). For EuRhSi$_3$, the best fit estimates of $\gamma$ and $\Delta$ are 30.5 $\text{mJ mol}^{-1}\text{K}^{-2}$ and 22.9 K respectively. The gap value is very close to that inferred from the resistivity data (24.9 K), which lends credibility to our analysis.

In the case of EuRhSi$_3$, the latter value somewhat lower than the resistivity derived value (23.7 K). It may be noted that the Sommerfeld coefficient $\gamma$ is an order of magnitude larger than in $sp$-metals, and even larger than in many $d$-metal alloys. The Sommerfeld coefficient is expressed as [13]:

$$\gamma = \frac{5\pi^2 k_B^2 n(\varepsilon_F)}{\mu_B},$$

and its large value must be related to an enhanced $n(\varepsilon_F)$.

The entropy $S_{Jf}$ associated with the magnetic ordering was estimated by integrating $C_v/T$, where $C_v$ was obtained by subtraction from $C(T)$ of both the normalized heat capacity of LaTSi$_3$, taking into account the slight difference in the atomic masses of Eu and La, and the large conduction electron contribution $\gamma T$. It is seen that $S_{Jf}$ attains the expected value of $R \ln 8$ (for $S = 7/2$ Eu$^{2+}$ ions) close to $T_N$ in both compounds.

3.6. Eu Mössbauer spectra

The Mössbauer spectra on the isotope $^{151}$Eu in EuIrSi$_3$ are shown in figure 8 in the two temperature ranges defined as phase I ($T < 43.1$ K) and phase II ($43.1 < T < 51.8$ K). Spectra in EuRhSi$_3$ are similar and are not shown for brevity.

The spectrum at 4.2 K is a standard hyperfine field pattern characteristic of Eu$^{2+}$ ($S = 7/2$, $L = 0$), with an isomer shift relative to EuF$_3$ of $-7.92(5)$ mm s$^{-1}$ which matches well with the value reported in [8]. This spectrum presents the peculiarity of a very high hyperfine field $H_{hf}(4.2$ K $)= 52.6(3)$ T (51.6 T in EuRhSi$_3$). This high value can be explained by the large $n(\varepsilon_F)$ prevailing in these materials. Indeed, in magnetically ordered intermetallic materials with the $L = 0$ ion Eu$^{2+}$, the hyperfine field is solely due to the spin polarization of the $s$-type electrons at the nucleus site (i.e. there is no orbital contribution). It can be expressed as [18]:

$$H_{hf}(T) = A_{ce} \mu_B \langle \hat{s} \rangle_T,$$

where the first term is the ionic core polarization field proportional to the Eu$^{2+}$ moment $m(T) = -g\mu_B(S)_{\parallel}$ and worth $\approx -34$ T at saturation [19], the minus sign indicating that this field has a direction opposite to that of the magnetic moment.

The second term $H_{ce}$ is due to the conduction electron spin polarization $\langle \hat{s} \rangle_T$ induced by the 4$f$ shell through $J_{\ell\ell}$ exchange and is given approximately by [18]:

$$H_{ce} = A_{ce} \mu_B \langle \hat{s} \rangle_T,$$

where $A_{ce}$ is a hyperfine constant. According to equation (1), the effective field on the conduction electron spin $s$ is, in the magnetically ordered phase:

$$H_{eff} = -\frac{1}{g\mu_B} J_{\ell\ell} s + \frac{1}{(g\mu_B)^2} J_{\ell\ell} m(T).$$

Introducing the Pauli susceptibility: $\chi_p = 2 \mu_B^2 n(\varepsilon_F)$, the conduction electron spin polarization writes:

$$\langle \hat{s} \rangle_T = -\frac{1}{g\mu_B} \chi_p H_{eff} = -\frac{1}{g} \frac{n(\varepsilon_F)}{\mu_B} J_{\ell\ell} m(T),$$

and finally the conduction electron hyperfine field writes:

$$H_{ce} \approx -\frac{1}{g} A_{ce} J_{\ell\ell} n(\varepsilon_F) m(T),$$

i.e. it is proportional to the density of electronic states at the Fermi energy $n(\varepsilon_F)$. In most Eu$^{2+}$ intermetallic materials and in the other members of the series, the saturated hyperfine field amounts to about $-30$ T, indicating that $H_{ce}$, which is
The difference between the (negative) measured hyperfine field and the ionic core polarization field $-34$ T, is positive and has the small value of a few T. By contrast, in EuIrSi$_3$ and EuRhSi$_3$, $H_{\text{ce}}$ is negative and much larger ($\approx -18$ T) due to the enhanced $n(E_F)$ value according to expression (10). The same situation holds in EuFe$_4$P$_{12}$, which presents the largest hyperfine field ever measured with $^{151}\text{Eu}$ ($-67$ T), implying $H_{\text{ce}} \approx -33$ T [20].

Whereas the spectra in phase I (below 43 K) show the presence of a unique hyperfine field, and hence of a unique Eu$^{2+}$ magnetic moment, there occurs a sudden change of the spectral shape above 43 K, i.e. when entering phase II, where the spectra can be fitted to an incommensurate modulation of hyperfine fields [21]. The latter is well described by the first 3 odd harmonics of a Fourier series:

$$H_{\text{hf}}(kx) = h_1 \sin(kx) + h_3 \sin(3kx) + h_5 \sin(5kx)$$

where $k$ is the propagation vector of the modulation and $x$ the distance along $k$. The three coefficients $h_1$, $h_3$, and $h_5$ were fitted to the spectral shape at each temperature to obtain the modulation profiles shown in figure 9. The modulation becomes more ‘squared’ as temperature decreases and approaches the transition to the equal moment phase at $T_{N2}$.

The thermal variation of the hyperfine field is plotted in figure 10. In phase I, the hyperfine field values approximately follow a mean field law for $S = 7/2$, in line with its proportionality to the Eu$^{2+}$ moment derived above. The transition temperature of this mean field law (60 K) does not correspond to the actual Néel temperature because of the presence of the commensurate—-incommensurate transition at 43.1 K and of the first-order character of the transition to the paramagnetic phase at 51.8 K (see spectrum at 52 K in figure 8).

4. Conclusion

The whole set of our thermodynamic and spectroscopic measurements in the two divalent Eu intermetallics EuIrSi$_3$ and EuRhSi$_3$ can be coherently and qualitatively interpreted by assuming a high density of electronic band states at the Fermi energy, which sets them apart from the other members of the EuTX$_3$ family. We attribute the observed enhanced values to a large $n(E_F)$ rather than to an anomalously large 4f-conduction electron coupling $\lambda_{\text{df}}$ since the Sommerfeld coefficient does not involve $\lambda_{\text{df}}$ and there is a priori no reason for the stable Eu$^{2+}$ 4f shell to be prone to strong hybridization with the conduction band. The two compounds present a cascade of magnetic transitions near 50 K, from a paramagnetic to an incommensurate modulated, then to an equal moment antiferromagnetic phase. In the EuRhSi$_3$ single crystal sample, we could evidence an important anisotropy of the magnetization, confirming the link between these two phenomena.

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