Effect of Pressure on the Electronic state in Eu-Divalent EuTIn4 (T: Ni, Pd, Pt, Au) Compounds

To cite this article: M Hedo et al 2015 J. Phys.: Conf. Ser. 592 012047

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Effect of Pressure on the Electronic state in Eu-Divalent EuTIn₄ (T: Ni, Pd, Pt, Au) Compounds

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Abstract. We grew single crystals of Eu-divalent antiferromagnets EuTIn₄ (T: Ni, Pd, Pt, Au) by the In-self flux method, with the constitution of Eu:T:In = 1.1:1:30. Single crystals are characteristic in shape, being long along the orthorhombic [100] direction. We measured the electrical resistivity, specific heat, magnetic susceptibility, and de Haas-van Alphen effect for these compounds, together with the electrical resistivity under pressure for EuTIn₄ (T: Ni, Pd, Pt). Under pressure, the Eu-divalent electronic state is often changed into the Eu-trivalent state, revealing the valence transition. In the present experiments, the valence transition was, however, not observed even at high pressures up to 8 GPa for these compounds, but a sharp resistivity drop was observed just below the Néel temperature under pressure. This is most likely due to a change of the magnetic structure.

1. Introduction

Recently, the magnetic properties of EuTIn₄ (T: Ni, Pd, Pt, Au) compounds with the orthorhombic YNiAl₄-type structure were studied using single crystal samples[1, 2, 3, 4]. General saying, it is not easy to grow single crystals because of high vapor pressure of a Eu metal. The In-self flux method was applied to EuTIn₄ compounds. These compounds revealed the Eu-diavalent electronic states with the antiferromagnetic ordering. The magnetic properties are summarized in Table I.

The magnetic structures are different each other in EuNiIn₄, EuPdIn₄ and EuPtIn₄. The magnetic easy-axes are the [010] direction (b-axis), the (001) plane, and the (010) plane, respectively. Among them, a multi-step magnetization was observed for the magnetic field along the [010] direction in EuNiIn₄. Magnetic properties in EuAuIn₄ were, however, strange in properties, revealing a small effective magnetic moment of 5.88 μB/Eu without the magnetic ordering at temperatures down to 2 K[3]. Note that the magnetic properties of EuAuIn₄ in Table I were obtained in the present study.

To clarify furthermore the electronic states of EuTIn₄, we studied the electronic states by measuring the electrical resistivity, specific heat, magnetic susceptibility and de Haas-van Alphen
Table 1. Structural and magnetic properties of ETIn₄ (T: Ni, Pd, Pt), cited from refs. [1, 2, 3, 4] and EuAuIn₄.

|       | EuNiIn₄ | EuPdIn₄ | EuPtIn₄ | EuAuIn₄ |
|-------|---------|---------|---------|---------|
| a [Å] | 4.476   | 4.547   | 4.5436  | 4.6080  |
| b [Å] | 16.97   | 17.02   | 16.960  | 17.0457 |
| c [Å] | 7.228   | 7.385   | 7.385   | 7.228   |
| Tₕ [K] | 16      | 15      | 13      | 6       |

μ₀[H/V]/θₑ [K]

| H [100] | 7.87 (-22.4) | 7.99 (-14.4) | 7.45 (-12.1) |
| H [010] | 7.96 (-22.3) | 7.84 (-17.2) | 7.71 (-15.1) |
| H [001] | 7.69 (-19.0) | 7.77 (-13.0) | 7.52 (-11.4) |

estimated

| Hₑ [kOe] | 189 | 145 | 139 | 90 |

(dHvA) oscillations, together with the electrical resistivity under pressure.

2. Experimental Procedures

Single crystals of EuTIn₄ (T: Ni, Pd, Pt, Au) were grown by the In-self flux method, following the previous reports [1, 2, 3, 4]. We show in Fig. 1 the single crystal samples. The sample is a long and thin plate. The flat plane corresponds to the (010) plane and is long along the [100] direction. A large sample consists of these thin plate. The direction of the crystal was determined by the usual X-ray Laue method. The electrical resistivity was measured by the ordinary four-probe DC method. The specific heat was carried out by the quasi-adiabatic heat-pulse method. The magnetic susceptibility and magnetization were measured by a commercial.
SQUID magnetometer. The dHvA experiment was carried out by the standard field modulation method with a modulation frequency of 75 Hz and a modulation field of 90 Oe in magnetic fields up 150 kOe. The electrical resistivity under pressures up to 8 GPa was measured using a cubic anvil cell.

3. Experimental Results and Discussions
First we show in Fig. 2 the temperature dependences of the electrical resistivity $\rho$ for the current $J$ along the [100] direction. The inset show the low-temperature resistivity around a Néel temperature $T_N$. The resistivity decreases linearly with decreasing temperature and steeply decreases below $T_N = 16$ K in EuNiIn$_4$, $T_N = 14$ K in EuPdIn$_4$, $T_N = 13$ K in EuPtIn$_4$, and $T_N = 6$ K in EuAuIn$_4$. These $T_N$ values are in good agreement with the previous reports [1, 4]. Remarkably, the residual resistivity $\rho_0$ and residual resistivity ratio (RRR = $\rho_{RT}/\rho_0$, $\rho_{RT}$: resistivity at room temperature) in EuPtIn$_4$ are $\rho_0 = 0.3 \ \Omega\cdot$cm and RRR = 80, respectively, indicating a high-quality sample. It is also noticed that a change of the resistivity is observed at $T'_{N} = 15$ K, just below $T_N = 16$ K in EuNiIn$_4$. This might be due to a change of the magnetic structure.

We show in Fig. 3(a) the temperature dependences of the magnetic susceptibility $\chi$ and inverse susceptibility $1/\chi$ in EuPdIn$_4$ for three principle directions of $H \parallel [100]$, [010] and [001] at a magnetic field $H = 10$ kOe. From the linear slope of the temperature dependence of $1/\chi$ for $H \parallel [100]$, we obtained the effective magnetic moment $\mu_{eff} = 7.91 \ \mu_B$/Eu and the paramagnetic Curie temperature $\theta_p = -15.1$ K, respectively. The present $\mu_{eff}$ is almost the same as the divalent value of Eu$^{2+}$. As shown in Fig. 3(b), the magnetization curve along the [001] direction increases linearly up to 50 kOe. If we assume a canting process of magnetization for $H \parallel [001]$, the critical magnetic field $H_c$, namely a magnetic field reaching a saturated moment of 7 $\mu_B$/Eu, is 145 kOe, following the relation of $H_c = (k_B/3\mu_B)(T_N - \theta_p)$ [5, 6]. In Tabel I, the estimated $H_c$ values are
Figure 3. (color online) (a) Temperature dependence of the magnetic susceptibility $\chi$ and inverse susceptibility $1/\chi$, (b) magnetization curves, and (c) magnetic phase diagram in EuPdIn$_4$.

shown. Here, the metamagnetic anomalies are observed at 17 kOe for $H \parallel [100]$ and 32 kOe for $H \parallel [010]$. This indicates that the magnetic easy-axes are in the (001) plane. Namely, The [001] direction correspond to the hard-axis in magnetization. These results are in good agreement with the recent data [1]. From these data, we constructed an antiferromagnetic phase diagram, as shown in Fig. 3(c).

Next, we carried out the dHvA experiment for a high-quality sample EuPtIn$_4$. Figure 5(a) shows the typical dHvA oscillations for the magnetic field tilted by $\theta = 13.5^\circ$ from [010] to [100], and the corresponding fast Fourier transformation (FFT) spectrum. The dHvA frequency $F (= c h S_F / 2 \pi e)$ is proportional to the extreme (maximum or minimum) cross-sectional area $S_F$ of the Fermi surface, which is shown as a unit of magnetic field. Several dHvA branches are observed, which are named $\alpha, \beta, \gamma, \delta$ and $\varepsilon$, ranging from ($0.2 - 3.8$) $\times 10^7$ Oe. We rotated the sample against the magnetic field and obtained the angular dependence of the dHvA frequency, as shown in Fig. 4(b).

To clarify these dHvA branches, we carried out a full potential linear augmented plane wave (FLAPW) energy band calculation on the basis of a local density approximation (LDA) for SrPtIn$_4$, using the same structure parameters of EuPtIn$_4$. The theoretical angular dependence of the dHvA frequency and the corresponding Fermi surfaces are shown Figs. 4(c) and 4(d), respectively. Detected dHvA branches named $\alpha, \beta, \gamma, \delta$, and $\varepsilon$ are identified as follows. The branches $\alpha$ and $\delta$ correspond to band 78th and band 76th hole Fermi surfaces, respectively, as shown in Fig. 4(d). The branches $\beta, \gamma$ and $\varepsilon$ originate from a band 80th electron Fermi surfaces.

Here, we estimated $H_c = 141$ kOe using the values of $T_N = 13.3$ K and $\theta_p = -15.1$ K in ref. 4. The present dHvA experiments up to 135 kOe were thus carried out in the antiferromagnetic state. The antiferromagnetic Fermi surface is usually reconstructed on the basis of a small magnetic Brillouin zone. These detected dHvA branches are formed by the conduction electrons which break through the antiferromagnetic Brillouin zone boundary and circulate around the original paramagnetic Fermi surface and/or correspond to Fermi surfaces, which are not affected by antiferromagnetic ordering.
Figure 4. (color online) (a) dHvA oscillations and corresponding FFT spectrum, (b) angular dependence of the dHvA frequency, (c) theoretical one, (d) theoretical Fermi surfaces in EuPtIn$_4$.

Figure 5. (color online) (a) Temperature dependence of the specific heat, (b) temperature dependence of the magnetic susceptibility $\chi$ and its inverse susceptibility $1/\chi$, (c) magnetization curves in EuAuIn$_4$. 
In EuAuIn₄, we observed clear double transitions at $T_N = 15.8$ K and $T'_N = 15.0$ K in specific heat, shown by arrows in Fig. 5(a). We estimated the effective magnetic moment $\mu_{\text{eff}} = 7.45 \mu_B$/Eu and the paramagnetic Curie temperature $\theta_p = -12.1$ K, respectively, from the linear slope of the temperature dependence of $1/\chi$ for $H \parallel [100]$ in EuAuIn₄. The present $\mu_{\text{eff}}$ value is almost the same as the divalent value of Eu$^{2+}$. The effective magnetic moment for EuAuIn₄ is strikingly different from the previous report [3]. We also measured the magnetization, as shown in Fig. 5(c). The magnetization for $H \parallel [010]$ increases linearly up to 50 kOe. On the other hand, the metamagnetic anomalies are observed at 22 kOe for $H \parallel [100]$ and 36 kOe for $H \parallel [001]$.

For EuNiIn₄, EuPdIn₄ and EuPtIn₄, we carried out the electrical resistivity measurements under several pressures up to 8 GPa, as shown in Fig 6. The resistivity at high temperatures decreases in magnitude with increasing pressure, but the low-temperature resistivity around $T_N$ increases slightly. These behaviors are approximately the same in all the EuTIn₄ compounds. The former decrease of the resistivity is due to an effect that overlapping of the conduction bands is enhanced with increasing pressure and correspondingly the band width becomes large. The latter increases of the resistivity and the Néel temperature are due to enhancement of the RKKY interaction. The pressure dependence of $T_N$, obtained from $\rho - T$ curves, is shown in Fig. 7. $T_N$ increases linearly with increasing pressure at the rate of $dT_N/dP = 1.5$ K/GPa in EuNiIn₄, 1.4 K/GPa in EuPdIn₄, and 1.3 K/GPa in EuPtIn₄. Here, the $T_N$ value at 8 GPa is 26.9 K in EuNiIn₄, 25.1 K in EuPdIn₄, and 21.4 K in EuPtIn₄. The Eu-divalent electronic state is thus found to be extremely stable under high pressures up to 8 GPa. It is also noticed that another change of the resistivity is observed at pressures larger than 2 GPa just below $T_N$. This might correspond to a change of the magnetic structure at $T'_N$.

**4. Conclusion**

We grew single crystals of antiferromagnets EuTIn₄ (T: Ni, Pd, Pt, Au) and studied the electronic states. These compounds are very similar each other in the magnetic properties.
Divalent electronic states are found to be extremely stable against pressures up to 8 GPa. We observed another magnetic transition just below the Néel temperature under pressure. This might exist in these compounds even at ambient pressure in EuTIn$_4$.

Acknowledgments
This work was supported by a Grant-in-Aid for Scientific Research (C) (Nos. 23540418 and 25400342), from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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