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Prospect for research on spintronics of U_3As_4 ferromagnet and its semiconducting Th derivatives

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Low magnetic field magnetisation along easy magnetic axis [111], Hall resistivity and the effect of magnetic field of different directions on U_3As_4 resistivity along the easy axis as well as along the hard one ([100]) have been examined. The Hall resistivity reaches the highest value ρ_{H} = 1.75 μΩ·m at T = 113 K where it yields the giant anomalous Hall coefficient R_S = 6.25 μΩ·m·T^{-1} and the tangent of the Hall angle of 0.42. Magnetic field exceeding the demagnetisation field H_{demag} < 0.28 T changes the resistivity by up to 36 % at T = 77 K.

Key words: *ferromagnet; electronic transport; uranium compound, magnetic domain effect*

1. Introduction

Studies of magnetic and semiconducting properties in solid-state systems have served as an important test for understanding basic physics and discovering new applications in spintronics [1, 2]. During the last several decades, the anomalous Hall resistivity (AHR) and ferromagnetic anisotropy of resistivity FAR for sd electron systems have been experimentally investigated, to great extent motivated by the technological importance of both effects. On the other hand, these effects originate from anisotropy of the density of states and spin-orbit interaction [3], which can be fairly high in f-electron systems, thus we chose U_3As_4 and its thorium derivatives for AHR and FAR examinations in f-electron systems.

Uranium pnictides with U_3X_4 (X = P, As, Sb, and Bi) crystallise in a cubic structure and show ferromagnetic ordering with strongly anisotropic magnetisation and complex magnetic structures [4]. Two first compounds of the series show noncollinear magnetic structure with effective moment along the [111] axis (easy magnetic axis). The sublattice of U ions is split into 3 further sublattices with magnetic mo-

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ments of each tilted by a small angle α from the easy axis to [100], [010] and [001] axes, respectively. The two remaining pnictides show collinear magnetic ordering with the easy magnetic axis [100] and one third of uranium moments higher than the others. In this group of compounds, U_3As_4 has the highest Curie temperature T_C = 198 K, while its α = 3.1°. Magnetic field of 2 T applied along the hard axis at 77 K does not tilt noticeably the magnetic moments from the easy axis but 20 T at 4.2 K causes a spin-reorientation transition [5, 6]. These features favoured a long-standing interest in high-field magnetic properties of the U_3X_4 series. In this paper, we turn to examination of low magnetic field effect on electron transport properties of U_3As_4 because of its strong spin-orbit interaction and high anisotropy of density of states [7] that can be related to the spintronics. First Hall effect [8] and resistivity [9] examinations of U_3As_4 single crystals came before any study of their magnetic structure and should have been re-examined in reference to a previous short report on low-magnetic-field effect on resistivity [10].

2. Experimental

Crystals of U_3As_4 and Th_3As_4 were grown by the method of chemical vapour transport [11, 12]. Their crystal axes were determined with an optical goniometer. Next they were shaped into plates of dimensions 2.5×1.4×0.45 mm^3 along the <110>, <112> and <111> directions, respectively, by cutting with a wire saw and polishing. Magnetisation (M) was measured along <111>, Hall resistivity along <112>, with electrical current along <110>. Two specimens of a pillar shape of about 1 mm^2 cross-sections and 3 mm length along [111] and [100], respectively, were used for determination of resistivity in magnetic fields of various directions. Measurements of $M(T,H)$ were performed using a Quantum Design Magnetic Properties Measurement System, while resistivity and the Hall resistivity were determined using the conventional 4-point DC technique and an electromagnet.

3. Spontaneous Hall effect and ferromagnetic anisotropy of resistivity

The magnetisation was determined for the magnetic field H applied along [111] easy axis perpendicular to the plate of U_3As_4. Data presented in Fig. 1 allowed us to determine (by the shown linear extrapolation) the demagnetization field $H_{demag.}$ = 0.286 T and magnetic moment of U ion μ_U = 1.85 μ_B at 4 K, that can be compared to μ_U = 1.82 μ_B determined by means of neutron diffraction [4].

The Hall resistivity data $\rho_H = (V_H d)/I$ for the plate with the previously established magnetisation are presented in Fig. 2 for magnetic field H = 0.78 T (V_H is the Hall voltage, *d* is the plate thickness and *I* is the electric current through the plate). Following Ref. 13 we have calculated the anomalous Hall coefficient using formulae (1):

$$\rho_H = R_0B + R_S4\pi M \quad \text{for } T < T_C \quad \text{and} \quad \rho_H/H = R_0 + R_S4\pi\chi(1 + 4\pi\chi N) \quad \text{for} \quad T_C < T \quad (1)$$

The demagnetising field determines the demagnetising factor *N*, i.e., $H_{demag} = 4\pi NM$. R_0 and R_S denote the normal and anomalous Hall coefficients, and χ is the magnetic susceptibility. Previous high temperature studies (up to 450 K) showed that R_0 is by about 4 orders lower than R_S. ρ_H reaches the highest value at 113 K, where it amounts to $R_S = 6.25$ μΩ·m·T^{-1}. The tangent of the Hall angle at this temperature is of about 0.42.

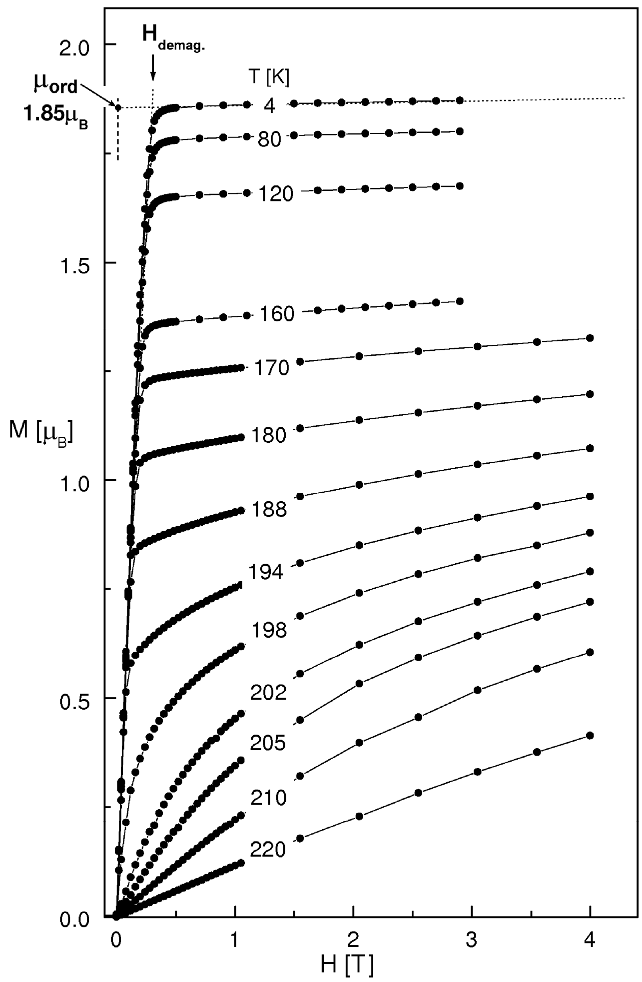

Fig. 1. Magnetisation vs. magnetic field applied along the easy magnetic axis perpendicular to the U_3As_4 plate

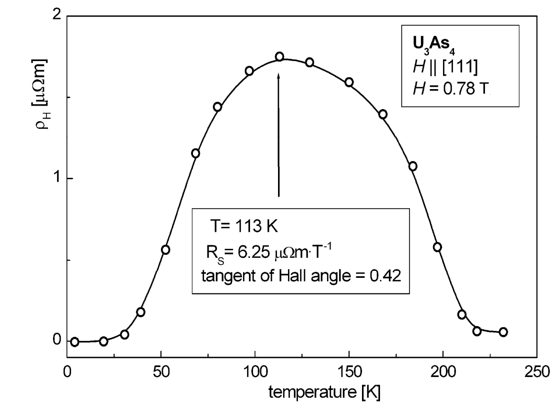

Fig. 2. Temperature dependence of the Hall resistivity for the same plate sample as in Fig. 1

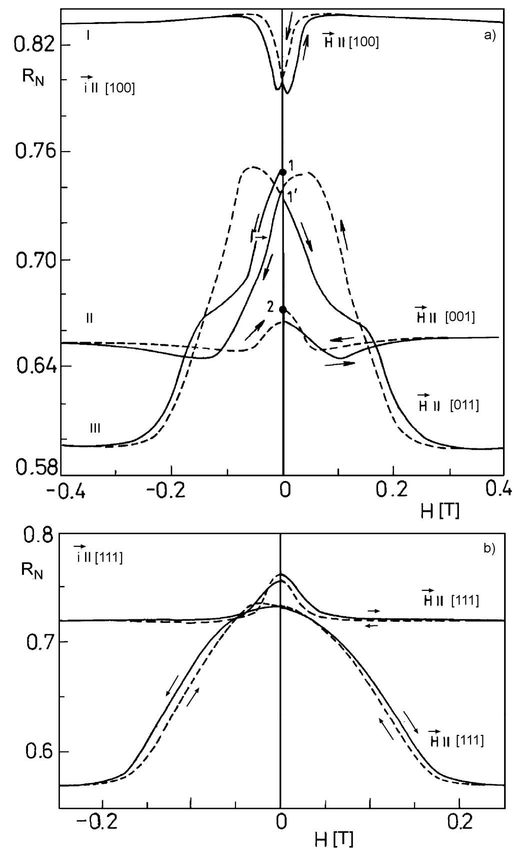

Fig. 3. The dependence of U_3As_4 resistivity measured along: a) [100] axis, b) [111] axis, with the magnetic field along several crystal axes indicated on the plots

Figure 3 shows the dependence of U_3As_4 resistivity along the [100] axis (a) and along [111] axis (b) on the magnetic field in various crystallographic directions at 77 K. The resistivity is normalized to that at T_C i.e. $R_N = \rho(H,T)/\rho(0,T_C)$. In the case of Fig. 3a the direction of magnetic field is in the middle between two ($H \parallel [011]$) or four ($H \parallel [100]$ or $H \parallel [001]$) easy directions. As such a low field cannot tilt the magnetic moment from the easy axis, we assume that in all these cases the saturation of magnetic domain is reached and that three different polydomain states of the sample are attained. We note that the resistivity of the specimen cut along [100] axis saturates in transverse field exceeding the value of $H_{demag.}$ determined, as shown above, from the $M(H)$ dependence (for H along the easy magnetic axis perpendicular to the plate). In the case of Fig. 3b presenting the resistivity of U_3As_4 along the [111] axis, the direction of the field is parallel to the easy axis being either parallel to the pillar specimen axis ($H \parallel [111]$) or at the 70.5° angle with the specimen axis ($H \parallel [11\bar{1}]$). As the magnetisation examination shows, this corresponds to the case of the monodomain state of specimen.

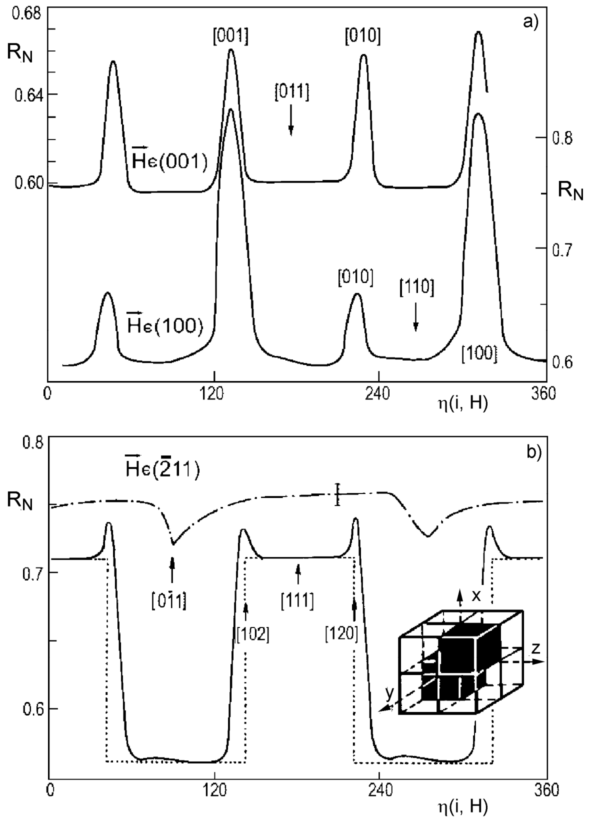

Fig. 4. Resistivity of U_3As_4 measured at T = 77 K along: a) [111] axis, b) [100] axis, magnetic field $H = 0.65$ T, rotating: a) in (100) or (001) plane for upper and lower curve of (a), respectively, b) in $(\bar{2}11)$ plane

The solid line in Fig. 4b represents the resistivity of U_3As_4 along the [111] axis at 77 K in the field of 0.65 T rotating in the $(\bar{2}11)$ plane, thus missing the $H \parallel [11\bar{1}]$ direction. The dotted curve represents the resistivity expressed by the formula:

$$R_N(\alpha_i, \beta_i) = a_0 + a_1 \left(\sum_{i=1,2,3} \alpha_i \beta_i \right)^2 + 2(a_2 - a_1) \sum_{i \neq j} \alpha_i \beta_i \alpha_j \beta_j \tag{2}$$

where α_1, α_2 and α_3 are the directional cosines of the spontaneous magnetisation with respect to the crystal axes [14]. The resistivity is measured in the direction determined by β_1, β_2 and β_3 cosines, while a_0, a_1 and a_2 are electrical anisotropy constants. Their values obtained by fitting Eq. (2) to resistivity measured at 77 K are: $a_0 + a_1/3 = 0.600$ and $a_2 = 0.166$. The step behaviour of the dotted curve is due to passing the magnetic field directions from black to white octant in the cube in Fig. 4b, causing the change of the α_i cosines. The dashed curve presents the resistivity after switching off the magnetic field in the given direction. One can see that there is an additional resistivity over the dotted curve in the cases when the polydomain structure is expected, i.e. at zero field or the peaks at the steps of the solid lines.

From the solutions of Eq. (2) for the resistivity of U_3As_4 along [100] axis at 77 K, we find $R_N = a_0 + a_1/3$ for each magnetic domain and we may expect angle-independent resistivity in the field of 0.65 T, which only switches the spotaneous magnetisation from one easy axis to another. As a matter of fact, we observe peaks of R_N on the field rotating by an angle η. We attribute this effect to polydomain structure and giant Hall resistivity, as proposed ealier [15]. The current passing the magnetic domains is bent to different directions in various domains due to different directions of the spontaneous magnetisation. This extends the current path and hence increases the resistivity

4. Remarks and conclusions

Examination of diluted n-type ($m^*/m_o \sim 0.2$) solid solutions of U_3As_4 [16] in semiconducting Th_3As_4 ($\Delta E = 0.43$ eV) showed a location of the 5*f* states well below the conduction band. The recent report on obtaining p-type Th_3As_4 with an effective mass of the carrier higher by one order of magnitude seems to open a new ferromagnetic semiconductor field of research.

U_3As_4 exhibits a giant anomalous Hall effect. AHR reaches the highest value $\rho_H = 1.75$ μΩm at temperature $T = 113$ K, where it yields giant spontaneous Hall coefficient $R_S = 6.25$ μΩ·m·T^{-1}. The tangent of the Hall angle at this temperature is about 0.42. On the other hand, the magnetic field of 0.3 T at 77 K is sufficient to switch the magnetisation of any domain to an easy magnetic axis closest to the field direction. This changes the resistivity due to FAR by up to 30 %. Additional components of variation of the resistivity of the same order as the FAR contribute to the AHR. This offers various possibilities to modify the resistivity of U_3As_4.

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