

High-Field Magnetisation and Magnetoresistance of U_3P_4 and Its Solid Solution $U_3(P,As)_4$

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Magnetisation and longitudinal magnetoresistance of single-crystal samples of two compositions: stoichiometric compound U_3P_4 and its solid solution $U_3(P,As)_4$ (with As:P ratio close to 1) were measured in pulsed magnetic fields with strength up to 47 T, and in temperatures from 4 K up to few tens of kelvins above the Curie temperatures (135 K and 181 K for both compositions, respectively). Field was always applied along a hard magnetic direction [100]. Magnetisation experiments showed no sign of expected magnetic-moment-reorientation transition and data look quantitatively similar for both compositions. On the other hand, longitudinal magnetoresistance (MR) is remarkably different for each composition. Onset of the above mentioned transition can be held responsible for broad maxima observed in $MR(B)$ curves for $U_3(P,As)_4$. Values of magnetic field strength corresponding to these maxima follow a linear dependence on temperature. We assume that these features of $MR(B)$ curves are due to progressive deformation of magnetic structure of $U_3(P,As)_4$ in high magnetic fields.

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1. Introduction

Aim of this work was to observe in U_3P_4 and $U_3(P,As)_4$ a magnetic-moment-reorientation transition analogous to that found previously for U_3As_4 compound and characterised by a critical field B_c of about 20 T at the temperature of 4.2 K, and continuous decrease in that critical field down to about 15 T above 100 K [1]. Magnetoresistance (MR) has provided a clear signature of this transition as well [2]. Earlier, also U_3P_4 has been investigated in fields up to 50 T, but transition has not been found either at 4.2 K or at 80 K [3]. Although magnetic and electronic structures of U_3X_4 -type pnictides ($X = P, As, Sb, Bi$) were well characterised theoretically [4] and experimentally [5, 6], the understanding of the magnetic-moment-reorientation transitions is not yet complete (e.g. questions of transition's continuity and of the resulting structure are still open) [7].

Ab initio calculations have predicted a magnetocrystalline anisotropy in U_3P_4 stronger by a factor of 5/3 than that in U_3As_4 [4]. On the other hand, a study of pressure effect on the critical field for U_3As_4 has shown that B_c increases linearly with applied pressure (by about 7.6 T/GPa) [8]. Extrapolation of these data (assuming linear compressibility and using a compressibility coefficient for U_3As_4) yields for U_3P_4 a prediction of $B_c \approx 80$ T at 4.2 K, however for a $U_3(P_{0.5}As_{0.5})_4$ solid solution it should be reduced to ≈ 50 T. Increasing temperature

should further decrease B_c values, making the reorientation transition accessible to an experiment at IFW High-Magnetic Field Laboratory.

2. Experimental details

Single crystals of two compositions: stoichiometric compound U_3P_4 and its solid solution $U_3(P,As)_4$ were grown by chemical-vapour-transport method using iodine as transporting agent. The crystals were characterised by X-ray diffraction that showed a proper stoichiometry for U_3P_4 and a composition with As:P ratio very close to 1 for the solid solution (derived from Vegard's law).

Specific-heat measurement for $U_3(P,As)_4$ showed clearly a phase transition at ≈ 181 K. Linear fit of low-temperature $C/T(T^2)$ dependence yielded Sommerfeld coefficient $\gamma = 82$ mJ/(K² mol). Magnetisation of $U_3(P,As)_4$ measured with a SQUID magnetometer confirmed ferromagnetic ordering below 181 K.

Magnetisation was measured on selected single crystals of approximately spherical form. For MR measurements a 4-contact ac method was used on elongated samples ($\approx 0.3 \times 0.3 \times 4$ mm) cut from single crystals along [100] direction.

Magnetisation and longitudinal MR data were collected at IFW-Dresden High-Magnetic Field Laboratory in pulsed magnetic fields with strength up to 47 T, in

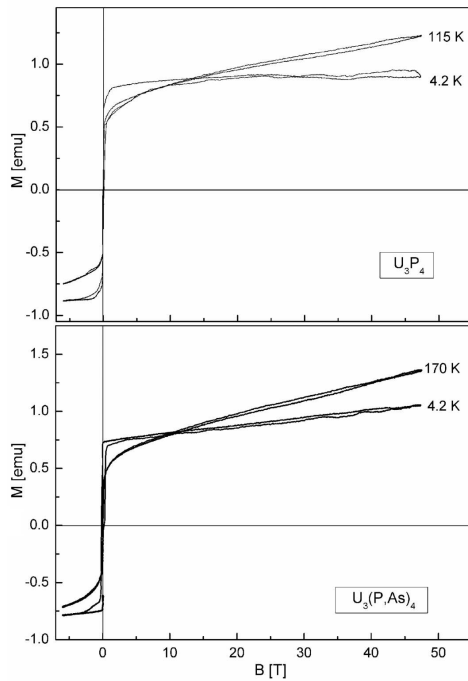


Fig. 1. Magnetisation of U_3P_4 (top) and $U_3(P,As)_4$ (bottom) in magnetic field applied along [100] direction.

wide range of temperatures, from 4 K up to few tens of kelvins above the Curie temperatures (being 135 K and 181 K for both compositions, respectively). In all measurements the field was applied in hard magnetic direction [100] (whereas the easy one is [111] for both compositions).

3. Results and discussion

Our magnetisation experiments showed no sign of the transition either for U_3P_4 or for $U_3(P,As)_4$ and data look qualitatively similar (see Fig. 1). Only visible difference is in a slope of linear dependence of magnetisation vs. field above 5 T: it is zero for stoichiometric compound but has finite, positive value for $U_3(P,As)_4$. On the other hand, longitudinal MR is remarkably different for both compositions (cf. Fig. 2 and Fig. 3). First it has opposite signs, and moreover strongly nonlinear form of $MR(B)$ curves for $U_3(P,As)_4$ is strikingly dissimilar to these for U_3P_4 . Complementary steady-magnetic-field MR measurements (data not shown) revealed for $U_3(P,As)_4$ a hysteretic behaviour with coercive field up to 2.8 T at 4.2 K, whereas corresponding field for U_3P_4 does not exceed 0.2 T. Most likely it is due to a strong domain pinning in $U_3(P,As)_4$ resulting from disorder of P and As atoms. Such domain behaviour may cause strong variations of MR observed in fields under 3 T (see Fig. 3).

Magnetoresistance in the U_3X_4 series can be considered in the frames of a two-current model introduced by Mott [9] and subsequently developed and successfully ap-

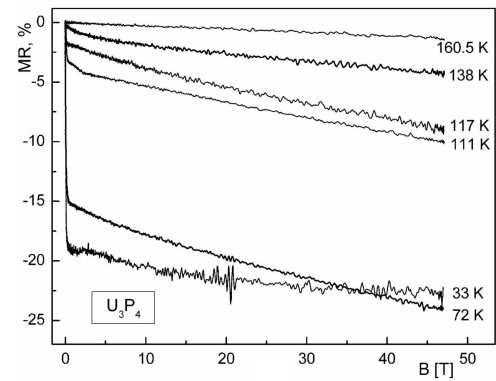


Fig. 2. Longitudinal MR of U_3P_4 in field applied along [100] direction.

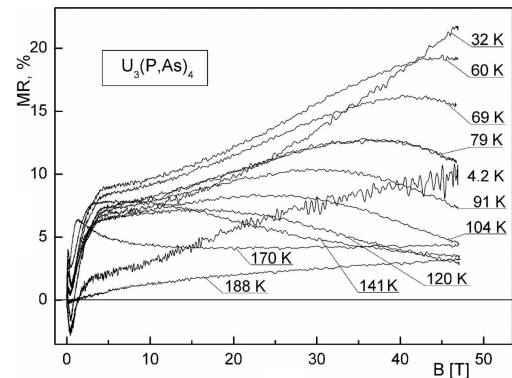


Fig. 3. Longitudinal MR of $U_3(P,As)_4$ in field applied along [100] direction.

plied to describe anisotropic magnetoresistance (AMR) of 3d ferromagnets [10]. In that model currents of spin-up and spin-down electrons are almost independent, and both consist of additive contributions from bands s and d .

When external magnetic field is applied or spontaneous ferromagnetic order appears, d -band is strongly polarised, i.e. density of states (DOS) of spin-up and spin-down d -subbands at the Fermi surface becomes drastically different. Although the conductivity is mainly due to light s -electrons, s - d interband scattering is essential factor in resistivity. When d -band is polarised, the probability of such scattering is reduced or enhanced, for minority or majority band, respectively. This is the main source of MR in 3d ferromagnets. Additional, field-angle dependent scattering between spin-up and spin-down bands is responsible for AMR.

All elements of that model are present in U_3P_4 and U_3As_4 — polarisation of their bands was found in both experiment [2] and theory [4]. However, instead of d -bands, hybridised d - f -bands have to be considered here [4–6, 11]. AMR reaches extremely large values in both compounds (–50% and 50%, respectively), but on the other hand, its temperature dependence is much different for each of them [12]. That indicates how different

are their electron structures near the Fermi level (also directly confirmed by de Haas–van Alphen (dHvA) experiments [6] and by theory [4]), despite very similar crystal and magnetic structures.

Such differences between U_3P_4 and U_3As_4 allow some, albeit only qualitative, analysis of MR behaviour in $U_3(P,As)_4$. Opposite sign of MR in U_3P_4 and $U_3(P,As)_4$ indicates that field dependence of band-polarisation is different for these two compositions. Therefore we can assume that in zero field d - f -bands are already strongly polarised (due to internal field of ferromagnet) and external field diminishes this polarisation in U_3P_4 (thus $MR < 0$), whereas enhances it in $U_3(P,As)_4$ ($MR > 0$).

In the course of a metamagnetic transition, as the magnetic structure is deformed, strength of internal magnetic field in a ferromagnet changes and band polarisation is again modified. Additionally, the direction of internal magnetic field changes, which certainly should influence MR through the anisotropic contribution (AMR) mentioned above. These two factors may cause reducing of MR observed for $U_3(P,As)_4$ at temperatures above 60 K.

Thus an onset of the magnetic-moment-reorientation transition can be held responsible for broad maxima observed in $MR(B)$ curves for $U_3(P,As)_4$. Values of magnetic field strength corresponding to these maxima clearly follow a linear dependence on temperature (Fig. 4). One can assume that such bending of $MR(B)$

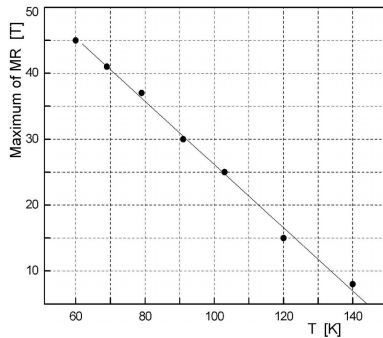


Fig. 4. Temperature dependence of magnetic field strength corresponding to maxima in $MR(B)$ curves shown in Fig. 3.

curves is a result of a deformation of magnetic structure $U_3(P,As)_4$ in high magnetic field. Lack of corresponding features in the magnetisation curve for $U_3(P,As)_4$ is most probably caused by some misorientation of the magnetic field direction and the hard-magnetic direction of the sample.

4. Conclusions

Our results indicate that the giant magnetic anisotropy of U_3P_4 persists in $U_3(P_{0.5}As_{0.5})_4$ composition and most

probably decreases rather abruptly in the composition range somewhere between $U_3(P_{0.5}As_{0.5})_4$ and U_3As_4 , which made possible an observation of the transition for the latter composition. In spite of a lack of a sharp transition in the field/temperature range covered by our investigation, interesting behaviour of both magnetisation and magnetoresistance was observed for both compositions, indicating large difference in their electron structure near the Fermi level. These are the first high-field experiments ever carried out for $U_3(P,As)_4$ composition, and for U_3P_4 they complement earlier low-temperature-only measurements. This work may help further understanding of the nature of magnetic order and metamagnetic transitions in the U_3X_4 series.

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