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Pressure effect on spin reorientation transition in $U_3\text{As}_4$

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Abstract
A linear increase in the critical field $H_c$ ($>200$ kOe) with pressure $p$ (0–0.5 GPa) was observed for the spin reorientation transition in $U_3\text{As}_4$ by measurements of magnetisation along the (100) axis at 5 K; $dH_c/dp = 76$ kOe/GPa. An interpretation of the results and a magnetic phase diagram for the $U_3X_4$ pnictides are proposed.

$U_3X_4$ pnictides are ferromagnetically ordered and yet hybridized systems. The hybridisation-mediated two-ion interaction leads to a highly anisotropic noncollinear ferromagnetic phase proved for $U_3\text{P}_4$ ($T_c = 138$ K) and $U_3\text{As}_4$ ($T_c = 196$ K) by neutron diffraction [1]. Discovery of the field-induced phase transition in $U_3\text{As}_4$ at $H_c = 190$ kOe and $T = 4.2$ K [2] was fruitful in the examination and understanding of magnetism of this compound [3,4]. We present here the effect of pressure on its $H_c$.

Fig. 1 shows that the dependence of magnetisation $\sigma$ on the magnetic field is different for two different $U_3\text{As}_4$ crystals. This has not been noticed before. The specific feature of sample 1 is that the magnetisation along the (100) axis at $H > H_c$ is identical to the spontaneous magnetisation along the (111) axis. For sample 2 the magnetisation above $H_c$ is clearly enhanced. In order to understand this effect we have compared this behaviour with that predicted by the spin Hamiltonian model of Przystawa [4].

The Przystawa model is closely related to the crystal structure of the $U_3X_4$ pnictides (bcc of Th$_3$P$_4$-type), which possess three different types of U sites, all with uniaxial crystal potential. The local symmetry axes fall along three different cubic axes for the three different types of U sites, and $D$ is the crystal field parameter. The two-ion anisotropic exchange-interaction tensor is characterised by components $J$ and $K$. The hitherto most advanced application of this model to the study of the $U_3X_4$ pnictides was for the spin $S = 1$ in Ref. [4], where the model is also described.

The present calculations have been done for the spin $S = 5/2$, which is more closely related to the paramagnetic moment of $U_3\text{As}_4$, $\mu_{\text{eff}} = 2.94\mu_B$. We have obtained better quantitative agreement between the calculated $\sigma(T)$ dependence and experimental results. The zero-field phase diagram for this case is shown in inset a in Fig. 1. It predicts two areas of parameters giving noncollinear ferromagnetic (NCFo) phases. Each phase has three sublattices and resulting magnetic moments along the (111) axis. Magnetic moments of particular U$^{4+}$ ions are tilted by the exchange interactions from the cubic axes toward the body diagonal direction in the case of NCFo-1 structure, or pass on the other side of the diagonal (between the (111) and (110) axes) in the case of the NCFo-2 structure. The NCFo-1 structure has been found for $U_3\text{P}_4$ and $U_3\text{As}_4$ by neutron experiments [1]. The NCFo-1 and NCFo-2 areas are separated by the area of collinear ferrimagnetic (CFi) structure. In this case the magnetic moments flop from two cubic edges onto the third one. The flopped moments are slightly reduced with respect to the remaining one, but all moments are parallel to each other and to the (100) axis. The temperature and magnetic field dependent magnetisation for $U_3\text{Sb}_4$ ($T_c = 140$ K) is consistent with that expected for the CFi phase [4].

The theoretical magnetisations for phases NCFo-1 and NCFo-2, plotted in Fig. 1, are multiplied by the factors 0.802 and 0.883, in order to obtain the same spontaneous magnetisations for the (100) axis as the experimental ones for the samples 1 and 2, respectively. This correction is
applied to show the correspondence between sample 1 and phase NCFo-1, or sample 2 and phase NCFo-2. It is not clear to us why both structures can exist alternately for the same composition.

$H_c = 191$ kOe is the lowest reported for the examined phase transition in $U_3As_4$ at 4.2 K [3]. Our zero-pressure determination of $H_c$ for sample 2 outside and inside the pressure vessel gives values of 205 and 230 kOe, respectively. In terms of the Przystawa model this discrepancy is due to misalignment between the (100) axis and the magnetic field. This effect, shown in inset b in Fig. 1, is consistent with the resulting inaccuracy of sample positioning in the pressure vessel ($< 2°$) and then in magnetic field ($< 2°$). We have also found that the misalignment cannot be responsible for the discovered magnetic moment enhancement above $H_c$.

The Przystawa model allows us to describe the magnetisation for $U_3Sb_4$, $U_3As_4$, and $U_3P_4$ one after the other when increments of $D/2J$ and simultaneous decrements of $K/J$ are proportional to the corresponding decrements of $O = U-U$ distance in these compounds. Hence $O$ scales $H_c$ in the model. Furthermore, it was found that $(dT_c/dp)$ determined for a particular compound follows the slope of $T_c$ versus $O$ dependence for compounds of this series [6,7]. Thus we propose the magnetic phase diagram shown in Fig. 2, following the linear pressure dependence of the $H_c$ shown in the inset of Fig. 2 and using compressibility data from Ref. [6].

It can be concluded that for $H || (100)$ the diagram predicts $H_c$ equal to 790 and 0 kOe for $U-U$ distances corresponding to the compounds $U_3P_4$ and $U_3As_3Sb$, respectively. In the case of $H || (111)$ a small negative value of $dH_c/dp$ is expected for $U_3Sb_4$. Experimental data show that no field-induced phase transition was found in fields up to 500 kOe for $U_3P_4$ [8]. We also know that at $H=0$ the NCFo-1 magnetic structure is observed for $U_3As_4$, while the magnetisation for $U_3Sb_4$ strongly suggests that it already has the CFI structure.

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