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Anisotropic exchange interactions in UNiGe

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UNiGe crystallizing in the orthorhombic TiNiSi-type of structure orders magnetically below $T_N = 50$ K with an additional magnetic phase transition at 42 K. Both structures (below 42 K commensurate antiferromagnetic, between 42 K and T_N incommensurate) are non-collinear with significant a -axis component ($\mu_x = 0.35 \mu_B/U$ at 20 K). The magnetic properties are highly anisotropic both in the ordered and the paramagnetic state. There are two metamagnetic transitions both with the field applied along the b - and the c -axis. While the magnetic structure above the second metamagnetic transition is forced ferromagnetic for both field orientations, for the field applied along the a -axis the magnetization curve at 4.2 K is linear up to 38 T and no change in magnetic structure is observed.

UNiGe provides strong evidence for anisotropic exchange interaction. The fact that the a -axis component cannot be aligned by the highest magnetic field used indicates that the antiferromagnetic interaction is much stronger between the a -axis components than between the others. This behaviour cannot be due to single-ion anisotropy because in that case the free energy does not depend on the sign of the a -axis component.

Keywords: Anisotropy - magnetic, Magnetic interactions, antiferromagnetism, Actinide compounds

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UNiGe is one of the most extensively studied equiatomic uranium ternary compounds. It adopts the orthorhombic TiNiSi-type of structure (space group $Pnma$) [1-3], in which the shortest U-U distance is found within zig-zag chains running along the a -axis (with an amplitude of 0.1 c). Magnetic measurements performed on high-quality single crystals prepared by a modified Czochralski method revealed that UNiGe orders magnetically below $T_N = 50$ K with an additional magnetic phase transition at 42 K [2,3]. The low-temperature magnetic structure, determined both by unpolarised and polarised neutron-diffraction experiments [3,4], is commensurate with a propagation vector $\mathbf{q} = (0, 1/2, 1/2)$ (Fig. 1). The magnetic structure between 42 K and T_N is incommensurate with $\mathbf{q} = (0, \delta, \delta)$ and $\delta = 0.359$ at 46 K. Both structures are non-collinear with significant a -axis components ($\mu_x = 0.35 \mu_B/\text{U}$ at 20 K). The zero-field magnetic structures can be modified by application of a magnetic field along the b and c axes. There are two metamagnetic transitions in UNiGe with the field applied along the c -axis (at 3 and 10 T) and two transitions with the field along the b -axis (at 17 and 25 T) (Fig. 2). While the magnetic structure above the highest critical field is forced ferromagnetic for both field orientations, in the intermediate region an uncompensated antiferromagnetic structure exists [2-4]. For the a -axis orientation, the magnetization curve at 4.2 K is linear up to 38 T and a magnetization value of $0.23 \mu_B/\text{U}$ is found at 35 T qualifying the a -axis as a hard magnetization direction. Strong magnetic anisotropy persists also in the paramagnetic state. At temperatures above 60 K, the magnetic susceptibility of UNiGe follows a Curie-Weiss law with an effective magnetic moment μ_{eff} close to $3.0 \mu_B/\text{U}$, with paramagnetic Curie-temperatures θ_p of -100, 10 and 40 K for the a -, b - and c -axes, respectively [5,6]. These data indicate that the anisotropy energy within the b - c plane, expressed as the difference of paramagnetic Curie-temperatures ($\theta_p^c - \theta_p^b$), is about 30 K, while the anisotropy within the a - b or a - c planes is much stronger.

The observations described above are unusual in two respects. First, it is very unusual to find a component of the ordered moment aligned with the nearest-neighbour U-U links. Secondly, it is surprising that the ordered moments have a component in the hard direction. The first feature contradicts an empirically established rule [7], that the moment direction is always found to be perpendicular to the n.n. U-U links, which itself is only qualitatively understood. The second observation contradicts any expectation based on models involving single-ion anisotropies of various sublattices. The purpose of this paper is to point out that the observation constitutes evidence for anisotropic interactions.

Consider a simple two-sublattice model of an antiferromagnet. The difficulty of generating a magnetic moment with field along the a axis will be reproduced by the model, if a contribution $K [(M^{(1)})^2 + (M^{(2)})^2]$ is included in the free energy, with the anisotropy constant

$K > 0$. Here, the superscripts refer to sublattices 1 and 2, the subscripts to the x component of the magnetization. Clearly, if the interaction between the two sublattice magnetizations is isotropic, i.e., of the form $J \mathbf{M}^{(1)} \cdot \mathbf{M}^{(2)}$, the anisotropy energy will suppress the x component of both sublattice magnetizations. Only an anisotropic interaction of the form $J_{xx} M_x^{(1)} M_x^{(2)} + J_{yy} M_y^{(1)} M_y^{(2)} + J_{zz} M_z^{(1)} M_z^{(2)}$, with $J_{xx} > J_{yy}$, J_{zz} will be found to favour antiferromagnetically aligned moments in the x direction. It is easily seen that in this case the energy of the antiferromagnetic state, where $\mathbf{M}^{(1)} = \mathbf{M}^{(2)} = \mathbf{M}^{(AF)}$, the total energy is $(2K - J_{xx})(M^{(AF)})^2 -$

$J_{yy}(M^{AF}_y)^2 - J_{zz}(M^{AF}_z)^2$, so that if $J_{xx} > 2K$ and if $J_{xx} - 2K$, J_{yy} and J_{zz} are comparable, higher-order anisotropy terms can dictate any direction for the ordered moment. On the other hand, in the forced ferromagnetic state, when $M^{(1)} = M^{(2)} = M^{(F)}$, the energy is $(2K + J_{xx})(M^{(F)}_x)^2 - 2H_x M^{(F)}_x$ if the magnetic field is in the x direction and $J_{yy(zz)}(M^{(F)}_{y(z)})^2 - 2H_{y(z)} M^{(F)}_{y(z)}$, if it is in the y or z direction. Since $2K + J_{xx}$ can be much larger than J_{yy} and J_{zz} , the forced ferromagnetic state may be realized with the field in the y or z direction, while being inaccessible for any available field in the x direction.

There is an interesting implication of the above model, which makes it possible to check its validity experimentally, by neutron diffraction using polarized neutrons. Under the assumptions made, the antiferromagnetically aligned x component of the magnetization should prevail when a ferromagnetic state is forced by a field in the y or z direction. Qualitatively, this can be understood as another manifestation of the inability of the Zeeman energy to overcome the anisotropic exchange. For a quantitative description one needs a model including higher-order anisotropies, which is being worked at presently.

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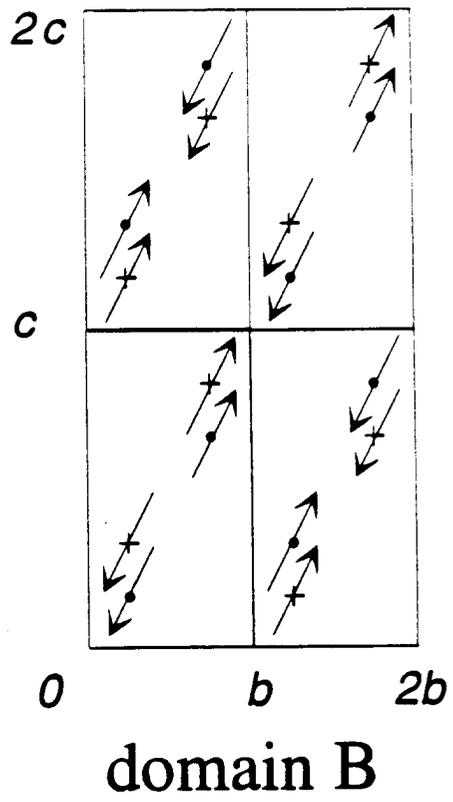
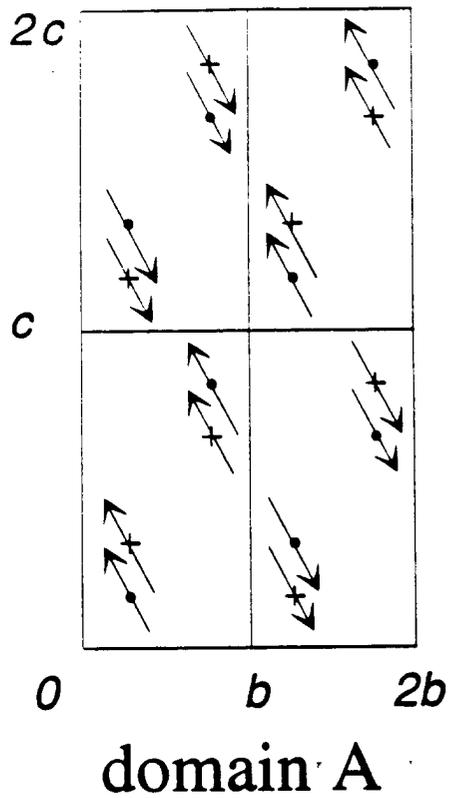
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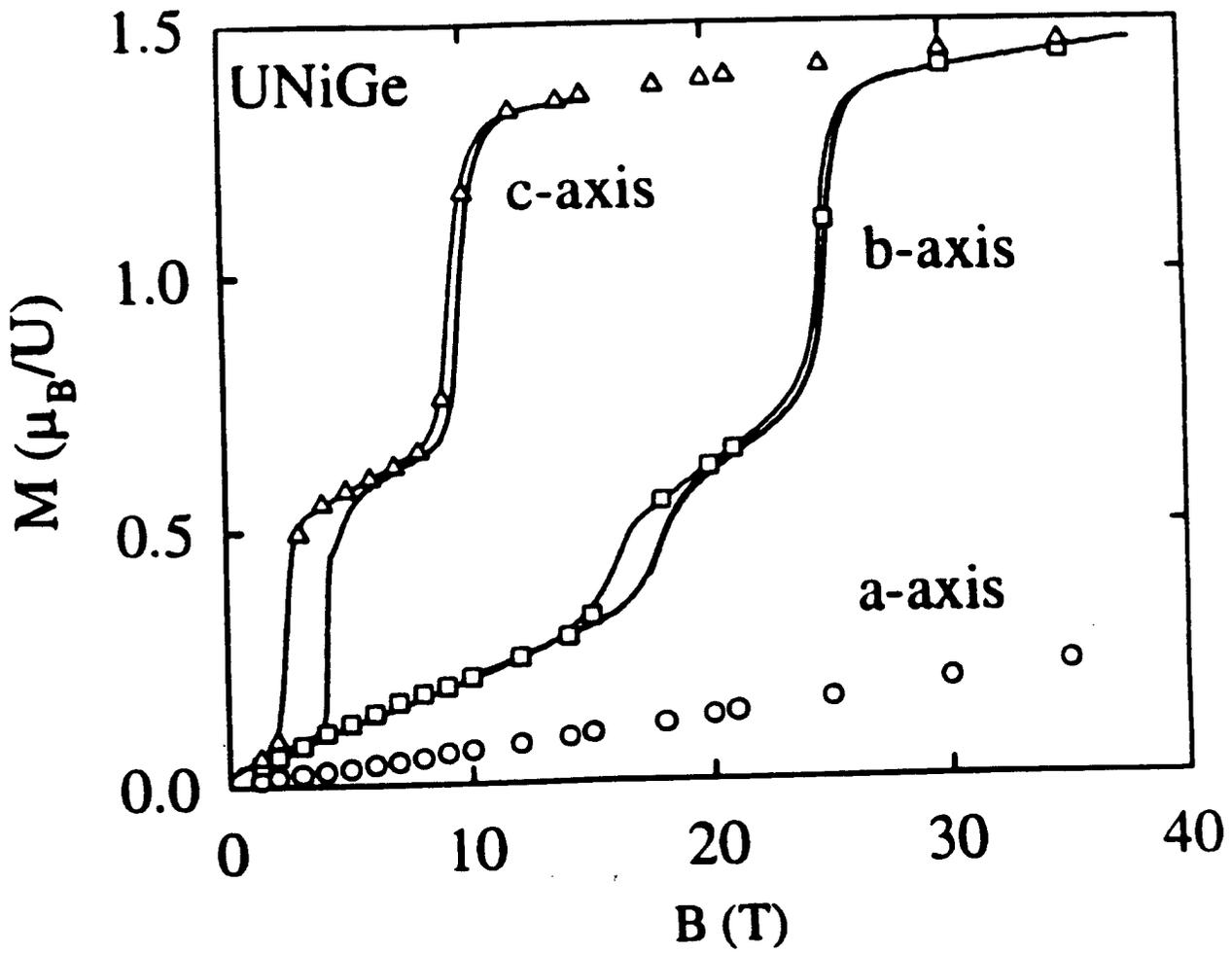
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Figure captions

Figure 1: Schematic representation of the low-temperature magnetic structure of UNiGe consisting from two domains. The arrows represent the moment components in the b - c plane. The dots and crosses represent moment components parallel and antiparallel to the a -axis, respectively. For sake of clarity, only U atoms are shown.

Figure 2: Field dependence of the high-field magnetization of UNiGe measured at 4.2 K along the three principal axes.





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