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Magnetic phase diagram of UNiGa

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Abstract

We present a complex magnetic phase diagram of UNiGa with several antiferromagnetic (AF) phases below $T_N = 39.5$ K. A relatively low magnetic field (~ 1 T) applied along the c -axis induces a transition from the AF phases to an uncompensated AF and/or a ferromagnetic (F) phase. All the magnetic structures are collinear ($\mu_U \parallel c$) and consist of ferromagnetic basal-plane sheets with various coupling along c .

UNiGa belongs to uranium ternaries UTX (T = transition metal, X = p-metal) which crystallize in the hexagonal ZrNiAl-type structure [1]. This structure consists of the U–T and T–X basal-plane layers alternating along the c -axis. The huge uniaxial magnetocrystalline anisotropy in these materials locks the U magnetic moments in the c -axis which leads to an Ising-type magnetism. It apparently originates in the existence of considerable U 5f orbital moments and in the strong bonding of U 5f wave functions in the U–T planes. The anisotropic bonding results in an anisotropic 5f ligand hybridization which mediates anisotropic exchange interactions between the U moments (only a small magnetization density is induced on transition metal sites). The exchange interactions within the U–T planes are strong and yield ferromagnetic basal-plane sheets in the ordered state. The c -axis interaction is much weaker, but it is decisive for the type of ground state, because it provides the coupling between the ferromagnetic sheets. In compounds with Ni, Pd and Pt, it leads to AF ordering. Magnetic phase diagrams of these materials usually contain more phases characterized by different stacking of (+) and (–) oriented ferromagnetic sheets.

Here, we present a detailed magnetic phase diagram (Fig. 1) of UNiGa deduced from results of extensive magnetization [2], magnetoresistance [3], specific heat and

magnetocaloric effect [4], magnetostriction and neutron diffraction (at CENG and BENSCE) studies performed on a single crystal at various temperatures and in magnetic fields applied along the c -axis.

All mentioned experiments were performed on samples originating from the single crystal of UNiGa (~ 2 g) grown in a Czochralski tri-arc equipment of the FOM ALMOS at the University of Amsterdam [5].

Fig. 1. Magnetic phase diagram of UNiGa. (1) Incommensurate AF structure, $q = \pm(0, 0, \delta)$, $\delta \sim 0.36$. (2) AF phase with frustrated 'paramagnetic' moments in each third U–T plane, i.e. $(+0-)$, $q = \pm(0, 0, \frac{1}{3})$. (3) AF phase with the $(++-+-+)$ stacking, $q = \pm(0, 0, \frac{1}{6})$, $\pm(0, 0, \frac{3}{8})$. (4) AF phase with the $(++-+-+)$ stacking, $q = \pm(0, 0, \frac{1}{2})$, $\pm(0, 0, \frac{1}{3})$, $\pm(0, 0, \frac{1}{6})$. (5) Uncompensated AF phase with the stacking $(++-)$, $q = \pm(0, 0, \frac{1}{3})$. (6) Ferromagnetic phase. (7) Paramagnetic phase.

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Fig. 2. Temperature development of reflections in the 11l scans in the range $l = 1.31$ – 1.38 .

The temperature ranges of stability of the AF phases 1–4, which appear consecutively within a very narrow temperature interval, can be deduced from Fig. 2. The onset of magnetic ordering in UNiGa appears by a second order phase transition around 39.5 K, where the incommensurate antiferromagnetic phase (IAFP) with $q = (0, 0, \delta)$, $\delta = 0.37$, is formed. This phase is stable down to 37.3 K (the δ value is slightly decreasing with decreasing temperature, to 0.36 at 37.5 K), where it transforms by a first order transition to the phase 2. The phase 2 has a periodicity $3c$, but zero spontaneous magnetization [2]. This can be understood in terms a stacking sequence $+0-$, where 0 means zero magnetic moment.

With further decreasing temperature, gradual transformation to the ground-state AF phase (4) with the stacking $(++-+-)$ of ferromagnetic planes via the 'intermediate' phase 3 $(++-+-+)$ was observed. While the 2–3 transition is of the first order type, the transformation between phases 3 and 4 is gradual over an interval of several K, where the two phases coexist. This is manifest in a simultaneous occurrence of magnetic reflections $h, k, l \pm \frac{1}{8}$; $h, k, l \pm \frac{3}{8}$, and $h, k, l \pm \frac{1}{2}$; $h, k, l \pm \frac{1}{3}$; $h, k, l \pm \frac{1}{6}$ respectively.

The U magnetic moment, which is $\sim 1.4\mu_B$ at 2 K, does not decrease substantially almost up to 35 K, where it keeps still more than 90% of its low temperature value. This behaviour seems to be more general for the UTX compounds which exhibit the magnetism with lower dimension.

The line 7–5 in fields between ~ 0.2 and 1 T marks first order transitions in contrast to the second order transitions 7–1 in lower fields. Also the transitions 4–5–6 above 14 K are of the second order type. Between phases 5 and 6 seems to appear another phase in about 1 K interval, which is evident also from the specific-heat data taken in 1 T [4]. This phenomenon requires further detailed studies.

The phases 1, 2 and 3 are stable only in very low fields. The propagation vector q of the IAFP gets slightly reduced under application of field (less than by 0.01 in 100 mT).

A special story is connected with the magnetic state below 14 K depending on magnetic history indicated by the bulk measurements [2]. The neutron experiments revealed that these magnetic history effects are reflected also on the microscopic scale including the relaxation phenomena:

(a) for $T = \text{const.}$, for sweeping the applied magnetic field the upper (lower) line in Fig. 1 marks the $4 \rightarrow 6$ ($6 \rightarrow 4$) transition. This fact is well documented by the large hysteresis of the field dependence of the magnetization and magnetoresistance. The hatched region represents the hysteresis, which increases progressively with lowering temperature. The metamagnetic transitions below 14 K are of the first order type. In the hatched region only one phase (either 6 or 4) is present in UNiGa. The same holds for the transitions $4 \rightarrow 6$ and $6 \rightarrow 4$ with varying temperature.

(b) $B = \text{const.} \sim 0.73$ T; by cooling from higher temperatures (phase 5) down to the 'hysteresis region' we reach an admixed state consisting of the phases 4 and 6, which is manifest by observing simultaneously the magnetic intensities characteristic for each phase (4 and 6). The 4 + 6 phase admixture, which we observe also in the magnetoresistance and magnetization, seems to be stable, at least on the time scale of minutes.

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