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Giant magnetoresistance and magnetic phase diagram of UNiGa

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As a result of combined magnetization, specific heat, electrical resistivity, and neutron-scattering studies on single crystals, we present a complex magnetic phase diagram of UNiGa that exhibits several antiferromagnetic (AF) phases below $T_N = 39$ K. A relatively low magnetic field (~ 1 – 1.5 T) applied along the c -axis induces metamagnetic transitions from the zero-field AF structures to an uncompensated AF and/or to a ferromagnetic (F) phase. All the magnetic structures are collinear (uranium magnetic moments parallel to the c -axis of the hexagonal structure). They consist of F basal-plane sheets which are coupled along c in various ways. The strongly anisotropic magnetism in UNiGa causes anisotropy of the electrical resistivity. The magnetic phase transitions are reflected in pronounced resistivity anomalies especially for current along the c -axis. The metamagnetic transitions are accompanied by giant magnetoresistance (GMR) effects which can be correlated with respective changes of the magnetic periodicity. This implies an important feature of the mechanism responsible for GMR phenomena in UNiGa. The strong resistance enhancement in the paramagnetic range when approaching T_N is connected with AF short-range ordering with propagation along the c -axis. © 1996 American Institute of Physics. [S0021-8979(96)56708-7]

UNiGa is one of uranium ternary compounds 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 uranium magnetic moments along the c axis which leads to an Ising-like behavior. The anisotropy is due to considerable uranium $5f$ -orbital moments and in the strong bonding of $5f$ -states in the U–T planes.

The bonding implies the anisotropic $5f$ -ligand hybridization which mediates anisotropic exchange interactions between the U moments. The strong F exchange interaction within the U–T planes yields magnetic structures built up of F basal-plane sheets (with $\mu_U \parallel c$). The interaction along the c -axis is, as a rule, considerably weaker but it plays an important role in determining the type of magnetic ordering, because it provides the coupling between the F sheets.

We present the magnetic phase diagram of UNiGa deduced from results of magnetization,² electrical resistivity and magnetoresistance,³ specific heat,⁴ and neutron-scattering studies performed on single crystals. Results involving application of magnetic fields have been obtained in fields parallel to the c -axis, which is the easy magnetization direction. The huge uniaxial anisotropy prevents perpendicular magnetic fields (at least up to 35 T) from affecting the magnetic state of UNiGa.

All experiments were performed on samples originating from two single crystals grown in a Czochralski tri-arc equipment of the FOM ALMOS at the University of Amsterdam. Both crystals were checked by the electron microprobe and found to be stoichiometric within the experimental accuracy of the method. Other details on sample preparation can

be found in Ref. 5. The low-field magnetization was measured in the field of 1.2 mT in a SQUID magnetometer. The resistivity, specific-heat,⁴ and magnetization² results were taken from our previous work where experimental details can be found. Neutron-scattering studies were carried out at temperatures between 2 and 80 K and in magnetic fields up to 2 T on double-axis spectrometers in CEN Grenoble and HMI Berlin. We also present additional neutron-scattering data obtained in zero magnetic on a triple-axis spectrometer at LLB operating in elastic mode.

In Fig. 1 we present the detailed magnetic phase diagram of UNiGa (B – T diagram for $\mathbf{B} \parallel c$). In zero magnetic field, four antiferromagnetic phases can be observed below T_N within about 5 K. The effects of the magnetic phase transitions on the resistivity, specific heat, magnetization, and peak intensities of characteristic magnetic reflections in the neutron-diffraction experiment are plotted in Fig. 2, from which one can also deduce the temperature ranges of stability of the AF phases.^{1–4}

Magnetic ordering in UNiGa appears through a second-order phase transition around 39 K, where the resistivity and low-field magnetization display only faint anomalies. The appearance of satellite reflections $h, k, l + \delta$ with $\delta = 0.36$ – 0.37 gives a clear evidence of an incommensurate antiferromagnetic (IAF) structure with the wave vector $\mathbf{q} = (0, 0, \delta)$. The transition temperature can be best defined from the maximum critical scattering anomaly measured for $\mathbf{Q} = 1, 1, 0.378$ (Fig. 3). Since previous estimates^{2–4} of T_N from bulk properties had a relatively large scatter due to weakness of the respective anomalies, we consider the present value, $T_N = 39$ K to be the most reliable.

The IAF phase is stable down to 37.3 K (the δ value is

FIG. 1. Magnetic phase diagram of UNiGa for magnetic fields applied along the c axis: (1) incommensurate AF structure, $\mathbf{q}=\pm(0,0,\delta)$, $\delta\sim 0.36$; (2) AF phase with $\mathbf{q}=\pm(0,0,1/3)$, e.g., the structure with frustrated “paramagnetic” moments in each third U–T plane, i.e., $(+0-)$; (3) AF phase with the $(++-+-+)$ stacking, $\mathbf{q}=\pm(0,0,1/8)$, $\pm(0,0,3/8)$; (4) AF phase with the $(++-+-)$ stacking, $\mathbf{q}=\pm(0,0,1/2)$, $\pm(0,0,1/3)$, $\pm(0,0,1/6)$; (5) uncompensated AF phase with the stacking $(++-)$, $\mathbf{q}=\pm(0,0,1/3)$; (6) ferromagnetic phase, $\mathbf{q}=\pm(0,0,0)$; (7) paramagnetic phase.

varying between 0.37 at T_N to 0.36 at 37.5 K), where it transforms to the phase 2 through a first-order transition. This phase has the periodicity $3c$, but zero spontaneous magnetization. This may be conceived with a stacking sequence

FIG. 2. Temperature dependence of the (a) specific heat, (b) electrical resistivity, (c) magnetization in a field 1.2 mT parallel to the c axis, and (d) the peak intensity of neutron-diffraction magnetic reflections (●) 1,1,4/3, (■) 1,1,11/8, and (▼) 1,1,0.36 in zero magnetic field measured on UNiGa in the temperature range 30–40 K.

FIG. 3. Temperature dependence of the peak intensity of the magnetic Bragg reflection $\mathbf{Q}=1,1,0.36$ (●) and intensity of the magnetic scattering at $\mathbf{Q}=1,1,0.378$ (+), normalized to the same monitor measured at zero energy transfer on a triple-axis spectrometer.

$+0-$, where 0 means the basal-plane layer of uranium atoms with the zero-ordered magnetic moment. Such a situation can appear in the case of a balanced AF interaction between nearest-neighbor and next-nearest-neighbor uranium planes. The resistivity drops by several percent at this transition.

With further decreasing temperature, gradual transformation of the magnetic structure to the ground-state AF phase 4 with the stacking $(++-+-)$ via the “intermediate” phase 3 $(++-+-+)$ was observed. While the $2\rightarrow 3$ is associated with a substantial increase of the resistivity, the other ($3\rightarrow 4$) yields a somewhat smaller decrease. As evidenced by the neutron-diffraction results, both transitions are characterized by a coexistence of neighboring phases over a certain critical region which can be more than 1 K wide (the transition $3\leftrightarrow 4$).

The uranium moment, which is $\sim 1.4 \mu_B$ at 2 K, does not decrease substantially with increasing temperature up to almost 35 K, where it retains more than 90% of its low-temperature value. This behavior seems to be more general for the UTX compounds which exhibit high anisotropy yielding an Ising-like behavior.

The line $7\leftrightarrow 5$ in fields between ~ 0.2 and 1 T in Fig. 1 marks the first-order transitions in contrast to the second-order transitions $7\leftrightarrow 1$ in lower fields. Also the transitions $4\leftrightarrow 5\leftrightarrow 6$ above 14 K are second-order transitions. The phases 1, 2, and 3 are stable only in very low fields. The propagation vector $\mathbf{q}$ of the IAF phase gets slightly reduced under application of field (by less than 0.01 in 100 mT).

The hatched area in the magnetic phase diagram below 14 K represents the B – T region in which magnetic history phenomena have been observed by the bulk measurements.² These effects are reflected also on the microscopic scale including the relaxation phenomena, as follows.

(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, magnetoresistance and magnetic reflection displayed in Fig. 4. The hatched region represents the hysteresis, which increases progressively

FIG. 4. Field dependence of the (a) magnetization, (b) reduced electrical resistivity, and (c) the peak intensity of the neutron diffraction reflection 1,1,4/3 measured on UNiGa single crystal at 4.2 K in magnetic field applied along the c axis. The offsets in critical fields are due to different demagnetization fields (different sample shapes) in particular experiments.

with lowering temperature. The metamagnetic transitions below 14 K are first order and only one phase (either 6 or 4) is present dependent on the direction of a field sweep. 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 a mixed state consisting of phases 4 and 6. This is clear from the simultaneous observations of magnetic intensities characteristic for each phase (4 and 6), and from bulk properties,^{2–4} as well. The $4+6$ phase mixture, which we observe also in the magnetoresistance and magnetization, seems to be stable, at least on the time scale of minutes.

As we have mentioned before, the metamagnetic transitions $4 \leftrightarrow 6$ and $4 \leftrightarrow 5 \leftrightarrow 6$ are inducing giant resistance changes. For example, the effect displayed in Fig. 4 represents almost 90% reduction of the resistance [$\Delta\rho/\rho(0T) \sim -0.9$] when we change from the AF structure with $\mathbf{q}=0,0,1/6$ (magnetic periodicity $6c$) to the F phase 6 ($\mathbf{q}=0,0,0$; periodicity $1c$). Above 14 K, the transition from the phase 4 to 6 is realized by two steps via the uncompensated AF phase 5 ($\mathbf{q}=0,0,1/3$, magnetic periodicity $3c$) yielding the spontaneous magnetization of $\frac{1}{3} M_s$ (M_s is the magnetization in the F phase). The magnetoresistance also shows two transitions,³ and the resistivity change between phases 4 and 5 is also approximately $1/3$ of that between phases 4 and 6.

All the magnetoresistance changes connected with the metamagnetic transitions between various phases in UNiGa are qualitatively correlating with the change in the periodicity of corresponding magnetic structures. The increase of the magnetic periodicity is followed by a positive resistivity

change. This observed systematic behavior points to the possible following scenario based on the high-resistivity state in antiferromagnets appearing due to superzone boundary formation (Fermi-level gapping). Thus, the additional magnetic periodicity yields the reduction of the carrier concentration and consequently enhances the resistivity. In the gapping model of Elliot and Wedgwood,⁶ the resistivity in the local-moment AF state can be described by

$$\rho = \frac{\rho_0 + \rho_{e-p} + \rho_{sp}}{1 - gm(T)}, \quad (1)$$

where $m(T)$ is the normalized sublattice (staggered) magnetization, ρ_0 the residual resistivity, ρ_{e-p} the contribution of electron–phonon scattering, and ρ_{sp} is the magnetic contribution (spin-disorder scattering). The truncation parameter g characterizes the effective reduction of the number of conduction electrons due to the Fermi-level gapping. The difference between the resistance in the F and AF state depends on the width in the electron energy spectrum, which scales with the exchange coupling parameter between the local moments and conduction electrons. The exchange coupling parameter can be significantly enhanced in case of U local moments connected with the $5f$ states forming a narrow band at E_F . A more detailed discussion of magnetoresistance in UTX compounds will be published elsewhere.⁷

The last point we want to discuss is the anomalous temperature behavior of the resistivity for the current along the c axis ($i \parallel c$), i.e., sensing the AF coupling between uranium moments. Below 80 K, the resistivity starts to increase with decreasing temperature. The increase becomes gradually more pronounced when approaching T_N . The fact that this increase can be suppressed by applied fields corroborates the idea that the negative $d\rho/dT$ above T_N is due to incipient AF correlations.^{2,5} The fact that a non-negligible neutron scattering (elastic) intensity at $\mathbf{Q}=1,1,0.378$ (see Fig. 3) can be detected far above the Néel temperature indicates that at least part of the enhanced resistance should be due to short-range AF ordering, which probably persists up to $2 T_N$. A more detailed account of the diffuse scattering at T_N will be a subject of a forthcoming article.

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