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Magnetic transitions in UNiGa

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Magnetization, magnetic susceptibility, and electrical resistivity studies of UNiGa in the single-crystalline form revealed a strong uniaxial anisotropy both in the paramagnetic and in magnetically ordered state below $T_c = 38$ K. At $T = 35$ K the system undergoes an antiferromagnetic transition, but by applying a field of about 1 T ferromagnetic alignment can be achieved.

I. INTRODUCTION

The uranium ternary intermetallic compound UNiGa crystallizes in the hexagonal ZrNiAl-type structure. It was reported to be magnetically ordered below 36 K.¹ The value of the ordered moment of $1.2\mu_B/\text{f.u.}$ and the high-temperature Curie-Weiss behavior of the magnetic susceptibility point to a local-moment character of the magnetism in this compound. There remains, however, some uncertainty about the type of the magnetic ordering in the ground state. The behavior of the magnetization in higher fields (above 1 T) points to ferromagnetic ordering, whereas in lower fields the situation is obscured by complicated sample-dependent magnetic history phenomena.¹ Here we describe results of magnetization, magnetic susceptibility, and electrical resistivity measurements performed on high-quality single crystals.

II. EXPERIMENTAL AND DISCUSSION

A large (diameter 6 mm, length 15 mm) single crystal of UNiGa was prepared by the tri-arc Czochralski method from a polycrystalline precursor, obtained by arc melting of stoichiometric amounts of constituents. One part of the single crystal was studied in this "as-obtained" state. The second part was subjected to an additional annealing for 2 weeks at 700 °C and used for comparative measurements of the magnetization. Microprobe analysis proved the homogeneity of the composition within 1%, which is less than the absolute experimental uncertainty.

The temperature dependence of the magnetic susceptibility was measured by means of a pendulum magnetometer in fields up to 1.3 T oriented both $\parallel$ and $\perp$ to the c axis. A Faraday-balance susceptometer, providing higher fields and better accuracy, was also used for measurements with $B\parallel c$, which is the easy-magnetization direction. In the temperature range 80–300 K (Fig. 1) $\chi^\parallel(T)$ follows approximately the Curie-Weiss behavior (with $\Theta_p^\parallel = 48$ K). Slight curvature of $1/\chi^\parallel(T)$ can be connected with a small temperature-independent term ($\chi_0^\parallel = 7 \times 10^{-10}$ m³/mol). Similar values of the effective moment, $\mu_{\text{eff}} = 2.6\mu_B/\text{f.u.}$, were found both for $\chi^\parallel(T)$ and $\chi^\perp(T)$, but the susceptibility in the basal plane is by far lower because of the large negative $\Theta_p^\perp \simeq -300$ K. The virtually parallel $1/\chi^\parallel(T)$ and $1/\chi^\perp(T)$ fit into a conventional local-

moment picture, while $\Delta\Theta = |\Theta_p^\perp - \Theta_p^\parallel|$, which reflects the energy of magnetic anisotropy, reaches the enormous value of $\simeq 350$ K.

Measurements of magnetization at 4.2 K were performed at the high field installation at the University of Amsterdam in fields up to 38 T. If hysteretic processes in low fields are disregarded, the magnetization with field $B\parallel c$, $M^\parallel$, reaches approximately $1.3\mu_B/\text{f.u.}$ and the differential susceptibility $dM^\parallel/dB$ is very small up to the highest fields. On the contrary, $M^\perp$ increases linearly from zero with a slope corresponding to $\chi^\perp$ at 4.2 K. A magnetization of $\simeq 0.15\mu_B/\text{f.u.}$ is achieved in 38 T. The extrapolation provides the intersect of $M^\perp(B)$ and $M^\parallel(B)$ for $B \simeq 330$ T, which is another measure of the anisotropy energy.

Detailed studies of the low temperature magnetic behavior were performed using the Faraday balance system and the extraction magnetometer. As can be deduced from Fig. 2, several magnetically ordered phases can be distinguished. The phase corresponding to the full moment of $1.35\mu_B/\text{f.u.}$, which we call for simplicity ferromagnetic (F), can be induced at low temperatures (below 10 K) by application of a field $B_{\text{crit}}^\parallel$, which decreases from 1.15 T at 2.2 K to 0.84 T at 10 K (values corresponding to the midpoint of the relatively steep transition of the metamagnetic type). Strong relaxation effects were observed for the magnetization near to these thresholds. The width of the hysteresis loop is reduced considerably on increasing temperature but the average $(B_c^\dagger + B_c^\perp)/2 \simeq 0.75$ T remains practically unchanged. The fact, that lattice imperfections play an important role in these phenomena, is demonstrated by the comparison with the annealed sample, for which the hysteresis is reduced considerably and the shoulder on the descending branch of the hysteresis loop is removed.² This shoulder indicates the occurrence of a metastable intermediate phase, possibly of the $++-$ type, denoted as ferrimagnetic (Ferri) with the magnetization equal to $0.43\mu_B/\text{f.u.}$ which is about $1/3\mu_s$. This points to the importance of thermally activated processes for the transition between the low-field antiferromagnetic (AF) and the high-field (F) phase. The situation, where the low-field metamagnetic transition can be accompanied by a considerable hysteresis, can explain the magnetic remanence observed in some cases.¹

When the temperature is increased above 10 K, the

FIG. 1. Temperature dependence of the inverse magnetic susceptibility measured in the field $B = 2$ T oriented parallel to the c axis and in the field $B = 1.2$ T perpendicular to it. The full line represents the modified Curie-Weiss fit with the parameters mentioned in the text. The dashed line represents the interpolation over the temperature range where data deteriorated by a small possible misorientation in the configuration $B_{\perp c}$ are unreliable.

transition AF $\rightarrow$ F and back occurs in two steps. A (possibly other) ferrimagnetic phase with $M = 1/3M_s$ becomes stable in a field interval that increases with increasing temperature. Whereas the higher critical field B_{c2} necessary for the Ferri $\rightarrow$ F transition increases with increasing temperature, the lower critical field B_{c1} (AF $\rightarrow$ Ferri) is gradually reduced and for temperatures just below the magnetic-ordering temperature the narrow ferrimagnetic hysteresis loop intersects the zero-field point. A better insight is here provided by the $M(T)$ scans for different fields (Fig. 3). In the very low-field limit $B = 10$ mT, two very sharp transition were observed, namely the P $\rightarrow$ Ferri one at 38 K (T_{c1}), and the Ferri $\rightarrow$ AF one at 35 K (T_{c2}). Both transitions are virtually free of any temperature hysteresis. The full Ferri moment is not reached in this field, which can be explained by the magnetization processes in the Ferri state. The actual domain structure allows to reach only $\approx 0.1\mu_B/\text{f.u.}$ for the "as obtained" sample, whereas $\approx 0.2\mu_B/\text{f.u.}$ was found for the annealed one in $B = 10$ mT. In a field of 0.1 T we found already the full Ferri moment approaching $0.4\mu_B/\text{f.u.}$ Increase of the applied field leads to a shift of T_{c2} to lower temperatures and the AF phase disappears entirely for $B \geq 0.8$ T. Instead, the F phase is gradually stabilized with the temperature of the Ferri $\rightarrow$ F transition increasing with increasing B .

Both zero-field transitions (i.e., P $\rightarrow$ Ferri at 38 K and Ferri $\rightarrow$ AF at 35 K) were found also in the temperature dependence of the electrical resistivity (Fig. 4). The high-temperature resistivity is practically temperature independent, whereas an upturn is observed below ≈ 80 K. The P $\rightarrow$ Ferri transition coincides with a sharp cusp at 38 K. The Ferri $\rightarrow$ AF transition is then manifest as a steplike increase in $\rho(T)$ at 35 K, which is followed by a decrease down to 4.2 K.

Interesting phenomena are also observed in the suscep-

FIG. 2. Magnetic-field dependence of magnetization measured at various temperatures on the nonannealed sample. Arrows indicate the direction of the field change. Lines are guides for the eye. a: (+) 2.2 K, ($\square$) 4.2 K, ($\triangle$) 6 K, ($\diamond$) 10 K. b: ($\square$) 25 K, ($\triangle$) 34 K, ($\diamond$) 37 K, (+) 40 K.

tibility in the paramagnetic range close to the P $\rightarrow$ Ferri transition. Below 80 K χ starts to deviate from the high-temperature Curie-Weiss law towards lower values. Then, however, instead of a gradual saturation χ turns up abruptly at the P $\rightarrow$ Ferri transition. The maximum of $|d\chi/dT|$ is gradually shifted to higher T and smeared out by applied field.

It is interesting to conceive the Ising-like character of the magnetism in UNiGa in the context of its crystal structure. A relatively close U-U spacing is found within the basal plane, where each U atom is surrounded by 4 U neighbors located at a distance of about 350 pm. This moderate spacing together with the presence of Ni atoms within this plane leads to a stronger hybridization within the plane than along the c axis, where two nearest U neighbours are found at a distance of about 400 pm. Magnetic moments are then perpendicular to the hybridization direction,³ i.e., along the c axis. Furthermore, much stronger magnetic interactions between moments in the basal plane can be expected. In the case of UNiGa a strong in-plane exchange interaction of the ferromagnetic type can be supposed (the positive Θ_p points to dominating ferromagnetic interactions), whereas much weaker interplane exchange

FIG. 3. Temperature dependence of the magnetization of the nonannealed sample in various fields: (+) 0.01 T, ($\square$) 0.1 T, ($\diamond$) 0.2 T, ($\triangle$) 0.4 T, ($\times$) 1 T, (∇) 2 T. Measurements were performed after cooling in the particular field, the thermal hysteresis is negligible. Lines are guides for the eye.

interactions can be responsible for the antiferromagnetic stacking along the c axis in the ground state. As shown in Ref. 4, even the next-nearest-neighbor coupling along the c -direction and the demagnetization field can decide about the stability of different field-induced magnetic phases.

The nature of the observed phase transitions still remains an open question and detailed specific-heat measurements and neutron-diffraction studies should shed some light on it. However, phenomenological similarity can be found to several systems, like CeFe_2 , where the succession $P \rightarrow F \rightarrow AF$ is to be found upon substituting Fe by Co (or some other d or p metals) with similar consequences for $\rho(T)$, in which a sudden increase of ρ accompanies the $F \rightarrow AF$ transition.⁵ The other system behaving reminiscent to UNiGa is the quasi two-dimensional antiferromagnet CsVF_4 , which displays a similar abrupt increase of $\chi(T)$ in the low-field limit. Here this behavior is related to the dimensionality of the system.⁶

One of most important aspects of the magnetism of UNiGa is the degree of localization of magnetic moments, which are expected to be nearly exclusively of the $5f$ origin.¹ The relatively high values of the spontaneous and effective magnetic moments and the Curie-Weiss behavior of $\chi(T)$ in all directions both ascertain us that we can describe UNiGa essentially as a local-moment system. An important argument is also the saturation of the magnetization curve at 4.2 K. This situation is somehow contrasting with, e.g., UNiAl which is a stronger hybridized system.⁷ On the other hand, the deviation of $\chi(T)$ from the CW law and the increase of $\rho(T)$ below 80 K could still reflect some magnetic-moment instability (spin-fluctuation

FIG. 4. Temperature dependence of the electrical resistivity measured with $i \parallel c$. The low-temperature detail is displayed in the insert.

effects). A huge magnetic anisotropy energy, which exceeds the values of similar rare-earth compounds by one order of magnitude,⁸ indicates that it originates not from classical crystal-field effects but from the anisotropy resulting from the $5f$ -hybridization mediated anisotropic coupling.³

III. CONCLUSIONS

UNiGa reveals a high magnetic anisotropy energy (of ≈ 350 K) both in the paramagnetic and magnetically ordered state. The plausible source of the anisotropy, the hybridization-mediated anisotropic coupling of $5f$ magnetic moments explains the c axis being the easy-magnetization direction. The ground-state magnetic ordering is probably antiferromagnetic with alternating orientations of ferromagnetically oriented moments within the basal plane. A weak interplane coupling causes that different types of stacking are nearly degenerate and that the ferromagnetic alignment can be achieved in low magnetic fields of about 1 T.

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