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Magnetic phases and magnetoelastic phenomena in UNiGa under pressure

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Abstract

UNiGa exhibits several antiferromagnetic (AF) phases below $T_N = 39$ K. The related magnetic phase transitions cause pronounced thermoexpansion anomalies of opposite sign for the *c*- and *a*-axes, respectively, leaving volume effects nearly negligible. Magnetic fields applied along the *c*-axis induce transitions from the AF phases to an uncompensated AF and/or to a ferromagnetic (F) phase accompanied by magnetostriction effects. Our results on pressure influence on the thermoexpansion and magnetostriction anomalies allow us to propose a tentative *p*-*T* magnetic phase diagram. A new AF phase induced by pressures above 1.8 GPa has been found. Neutron-diffraction studies under hydrostatic pressure up to 0.9 GPa and in magnetic fields up to 2 T confirmed that the critical temperatures and the critical fields of transitions of UNiGa are strongly pressure dependent (some phases are even suppressed) although relevant magnetic structures themselves remain essentially unchanged. © 1998 Elsevier Science S.A.

Keywords: Thermal expansion; Pressure effects; Phase diagram; Magnetoelastic phenomena

1. Introduction

UNiGa is one of the most thoroughly studied UTX compounds. However, a lot of controversy can be found among the results published earlier. In the course of time, UNiGa was claimed to order ferromagnetically [1,2], antiferromagnetically [3] and, because of pronounced magnetic after-effects, it was also classified as a spin-glass system [4]. Finally, it was recognized that the ground state of UNiGa is antiferromagnetic and that deviations from the exact 1:1:1 stoichiometry may cause a ferromagnetic ground state [5]. A concerted action comprising numerous bulk and neutron scattering experiments on high-quality single crystals allowed to determine a complex magnetic phase diagram (see Fig. 1) of this appealing material [6]. To study the hierarchy of exchange interactions in UNiGa, pressure studies of such a system are of natural interest. Since the magnetic phase transitions seem to be associated with striking resistivity anomalies, the resistivity of UNiGa single crystals exposed to hydrostatic pressures was thoroughly studied and tentative *p*-*T* magnetic phase diagrams have been proposed [7,8]. The phase transitions in UNiGa are clearly marked also by distinct magnetoelastic anomalies [9,10]. The observed pressure effects on these phenomena reflect changes of parameters of these transitions due to pressure-induced variations of magnetic moments (delocalization) and exchange interactions. In this paper we compare results of magnetoelastic measurements with neutron diffraction studies of UNiGa crystals under hydrostatic pressures.

2. Experimental

The single crystal of UNiGa was grown at the FOM-ALMOS centre at the University of Amsterdam from a stoichiometric melt by means of a modified Czochralski technique. The crystal was then closed in a quartz tube under 300 mbar He atmosphere and annealed for 2 weeks

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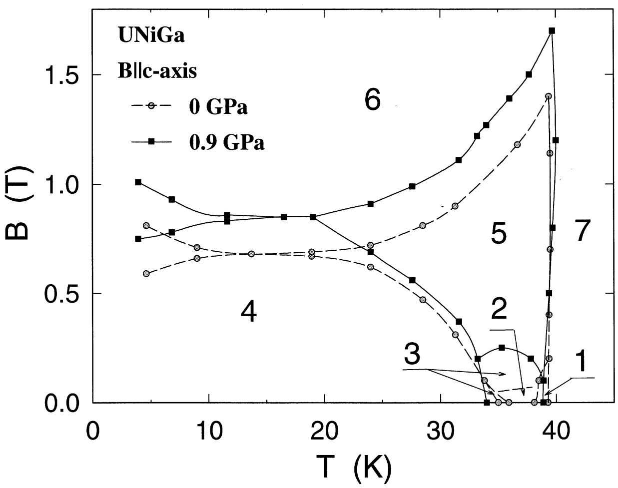

Fig. 1. Magnetic phase diagram of UNiGa at ambient pressure (dashed lines) and at 0.9 GPa (solid lines). (1) Incommensurate structure, $q=\pm(0, 0, \delta)$; $\delta\approx0.36$; (2) commensurate, sine-wave modulated '+−0' structure with $q=\pm(0, 0, 1/3)$; (3) commensurate, square-modulated AF '++−+−−+−' structure with $q=\pm(0, 0, 1/8)$ and $\pm(0, 0, 3/8)$; (4) the ground-state '++−+−−' structure with $q=\pm(0, 0, 1/6)$, $\pm(0, 0, 1/3)$ and $(0, 0, 1/2)$; (5) uncompensated '++−' AF structure with $q=\pm(0, 0, 1/3)$; (6) ferromagnetic state; (7) paramagnetic phase. All magnetic structures are collinear with U magnetic moments parallel to the c-axis and ferromagnetically coupled within the basal plane of the hexagonal ZrNiAl-type structure of UNiGa. In the ground state all uranium atoms carry the same magnetic moment of 1.4 μ_B.

at 600°C. The cooling rate down to room temperature was 6 K/h and quality of the crystal was checked by X-ray diffraction and by microprobe analysis. The top and the bottom of the crystal were found single crystalline, single phase and homogeneous with composition deviating from the ideal stoichiometry by less than 1 at.%.

The thermal expansion at ambient pressure along the principal axes was first measured with a parallel-plate capacitance method in the temperature range 1.5–210 K on a cube-shaped sample. Measurements under external hydrostatic pressures up to 2.2 GPa were performed on a sample on which also the magnetostriction data reported in ref. [10] have been taken. Prior to the experiments, the sample was exposed to an additional heat treatment at 250°C for 16 h in order to remove internal stresses. This treatment led to a positive shift of magnetic phase transition temperatures of about 1.5 K. The linear thermal expansion and the magnetostriction were measured by strain gauge glued on the single crystal which was placed under the hydrostatic-pressure in a magnetic field applied along the c-axis.

To identify the magnetic phases microscopically, we have performed neutron diffraction experiments under hydrostatic pressure up to 0.9 GPa with the field applied along the c-axis. The crystal was investigated on the normal-beam diffractometer D15 at ILL Grenoble. The crystal was glued at the end of an Al rod and mounted inside a Cu-Be pressure cell together with a crystal of NaCl which was used as a pressure-calibrating sensor. Hydrostatic pressure was generated by a mixture of kerosene and transformer oil.

3. Results and discussion

The temperature dependence of the linear thermal-expansion coefficients, along the a- and c-axes, ($\alpha_i=L_i^{-1} dL_i/dT$) are shown in Fig. 2 together with the coefficient of the volume expansion. As can be seen, the thermal expansion of UNiGa is highly anisotropic. Along the a-axis, the lattice monotonously expands with increasing temperature (α_a is always positive). At low temperatures we observe a considerable shrinking of the c-axis with increasing temperature. It becomes most pronounced around T_N and may be attributed to the loss of long range antiferromagnetic order. Around 70 K, where the antiferromagnetic correlations vanish (also reflected by termination of the negative slope of the $\rho(T)$ curve [6]), $\alpha_c(T)$ changes sign and at higher temperatures the lattice expands also in the c direction. Several sharp anomalies observed in both $\alpha_a(T)$ and $\alpha_c(T)$ curves between 34 and 39 K reflect magnetic phase transitions. Since the absolute value of α_c is approximately twice as large as α_a, the coefficient of volume expansion α_V is at low temperatures rather small.

Fig. 3a shows the linear thermal expansion along the c-axis in zero magnetic field and in the critical temperature interval. The data taken at several pressures between 0.1 and 2.2 GPa reveal that (a) the phases 1 and 2 are destabilised in pressures above 0.6 GPa, (b) the transition temperatures $T_{7\leftrightarrow3}$ and $T_{3\leftrightarrow4}$ are reduced at a rate of 1.1 and 2.6 K GPa^{-1}, respectively, and (c) at pressures ≥1.8

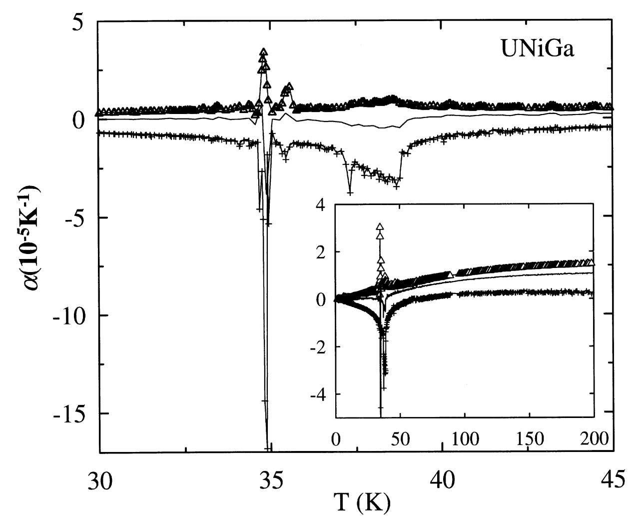

Fig. 2. Temperature dependence of the linear thermal expansion coefficients ($\alpha=L^{-1}(dL/dT)$) along the a-axis (△) and along the c-axis (+). The coefficient of the volume expansion $\alpha_V/3$ is given by the solid line.

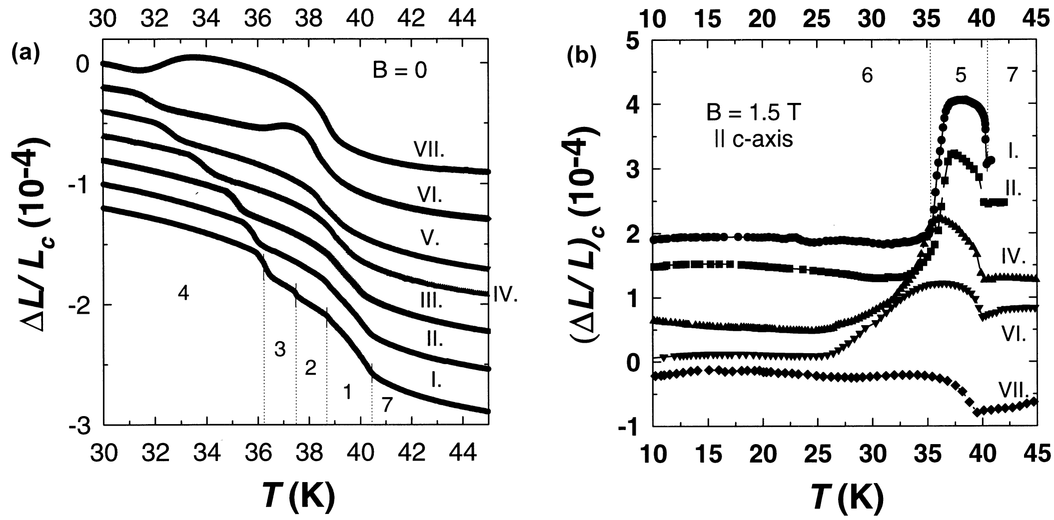

Fig. 3. Temperature dependence of the change of the relative length along the *c*-axis of the UNiGa in zero magnetic field (a) and in a field of 1.5 T$\|c$ (b) at (I) ambient pressure, (II–VII) pressures of 0.3, 0.6, 1.0, 1.4, 1.8 and 2.2 GPa. The magnetic phases are denoted as in Fig. 1.

GPa a new phase (presumably AF because of the behavior of the *c*-axis expansion which resembles other AF phases) is induced instead of the phase 3. A relevant *p*-*T* diagram of UNiGa in zero field determined from elastic experiments is shown in Fig. 4.

The thermal expansion along the *c*-axis in the magnetic field of 1.5 T at various pressures up to 2.2 GPa is shown in Fig. 3b. The sharp increase of $(\Delta L/L)_c$ and the simultaneous decrease of $(\Delta L/L)_a$ at ambient pressure reflects the magnetic phase transition from paramagnetic to the uncompensated AF phase 5. Opposite and more pronounced effects are observed at lower temperatures for the transition to the ferromagnetic phase 6. Application of pressure apparently enhances AF coupling which is documented by the gradually expanding existence range of the phase 5.

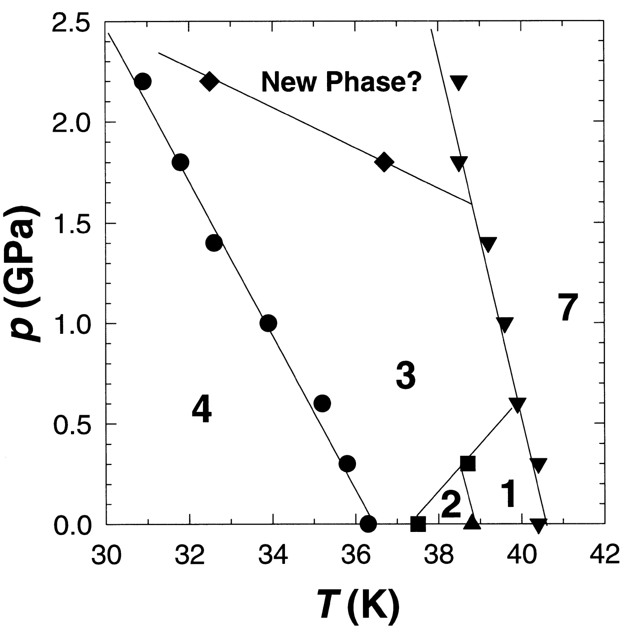

Fig. 4. A schematic *p*-*T* phase diagram of UNiGa in zero field determined from the thermal-expansion data. The magnetic phases are denoted as in Fig. 1.

The magnetic phase diagram at hydrostatic pressure of 0.9 GPa determined by slow field and temperature scans of intensities of relevant magnetic reflections in neutron diffraction is depicted in Fig. 1. As can be seen, it is generally similar to the one determined at ambient pressure and no new magnetic phases have been detected. However, some important modifications (consistent with results of elastic measurements under pressure displayed in Fig. 3 and Fig. 4) can be recognized by closer inspection: (i) T_N becomes reduced; (ii) the transition towards the field-induced ferromagnetic phase occurs at higher critical fields, confirming that antiferromagnetic interactions are promoted by hydrostatic pressure and (iii) the zero-field '1/8' phase, reveals strong history dependence. It can be traced, only in increasing field up to about 0.3 T. Above this field, the field induced '1/3' phase occurs. In the descending field, the '1/3' phase is stable down to zero field. Negligible reduction of magnetic intensities with application of pressure points to an important conclusion that not only the magnetic periodicity of individual phases, but also the U magnetic moment is rather inert with respect to pressures up to at least 0.9 GPa.

The results obtained of neutron diffraction experiments of UNiGa under pressure confirm conclusions drawn from previous electrical-resistivity and magnetoelastic measurements [7,8]. Pressures up to at least 2 GPa are desirable for the future neutron experiment to identify the new pressure-induced phase detected in the thermal-expansion studies (see Fig. 4). In order to address different components of strongly anisotropic exchange interactions (ferromagnetic within the basal plane and predominantly antiferromagnetic along the *c*-axis) experiments on crystals exposed to uniaxial pressure are needed.

The huge uniaxial magnetocrystalline anisotropy in UNiGa apparently originates from considerable uranium 5f-orbital moments and from the rather strong coupling of U 5f orbitals in the U-*T* planes where also a strong ferromagnetic interaction between U moments takes place. The interaction along the *c*-axis is considerably weaker and provides various (antiferromagnetic) couplings of the ferromagnetic basal-plane sheets. Since the value of the ordered U moment is independent of actual magnetic structure at ambient pressure and under hydrostatic pressures up to 0.9 GPa, we can definitely conclude that the pressure effects in UNiGa can be primarily attributed to variations in lattice parameters between individual magnetic phases, arising due to the strong spin-orbit interaction, i.e. not due to pressure effects on the size of U-moments.

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