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Antiferromagnetic ordering in the ternary uranium germanide UNi_{1-x}Ge₂: neutron diffraction and physical properties studies

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Abstract

The compound UNi_{0.45}Ge₂ was studied by means of X-ray diffraction (XRD), magnetization, neutron diffraction, specific heat and electrical resistivity measurements performed in wide ranges of temperature and magnetic field. The single-crystal XRD experiment indicated that the material crystallizes in the orthorhombic CeNiSi₂-type structure with the lattice parameters $a = 4.1062(2)$ Å, $b = 15.9272(8)$ Å and $c = 4.0461(2)$ Å. The physical properties measurements showed that it orders antiferromagnetically at $T_N = 47$ K. The magnetic ordering was confirmed by the neutron diffraction experiment, which revealed that the ordered magnetic moments are aligned along the crystallographic b -axis in a sequence $-++-$. In strong magnetic fields, a distinct field-induced first-order metamagnetic phase transition occurs, characterized by a large magnetic hysteresis. Temperature and field variations of the electrical resistivity are strongly affected by the atom disorder in the crystallographic unit cell of the investigated compound.

Keywords

Intermetallics; crystal chemistry; site occupancy; magnetic properties; electrical properties.

Introduction

The presence of zig-zag chains of uranium atoms in crystal structures of ferromagnetic UGe₂, URhGe and UCoGe, which locally break their inversion symmetry, seems to be of uttermost importance for the emergence of unconventional superconductivity in these three germanides [1]. In this context, investigation of other materials bearing U-U chains is a possible

route towards understanding microscopic mechanisms which govern the coexistence of long-range ferromagnetism and superconductivity in $5f$ -electron compounds.

One of the crystal structures harboring zig-zag chains of heavy atoms is that of the CeNiSi₂ type [2]. This structure is adopted by a few ternary uranium germanides UT_{1-x}Ge₂ with T = Fe [3], Co [4], Ni [5-8], or Ru [9]. Remarkably, the CeNiSi₂-type structure is closely related to the ZrGa₂-type structure of UGe₂ [10]. In the unit cells of both UGe₂ and the UT_{1-x}Ge₂ series, uranium atoms form a planar network of [U6] trigonal prisms centered by Ge-atoms, this network being separated from the other by a planar layer of [Ge4] lozenges. In the ternaries, the transition metal atoms fill partly the intercalation sites between the layers (see Ref. [9] for a graphical comparison). Interestingly, interatomic distances and angles in the U-U chains, as well as distances between such chains in the ac plane are nearly the same in UGe₂ and UT_{1-x}Ge₂. In contrast, distances between two consecutive U-U chains along the b -axis are significantly larger (by about 1 Å) in the ternaries.

The amount of experimental data available in the literature for the UT_{1-x}Ge₂ phases is fairly small. The compounds URu_{0.29}Ge₂ and UCo_{0.45}Ge₂ order ferromagnetically below the Curie temperature T_C of 63(1) K [9] and 18(1) K [4], respectively. In the ordered state, URu_{0.29}Ge₂ exhibits an unusual increase in the electrical resistivity [9,11], resembling analogous feature of UGe₂ that remains a subject of intense debate [12–16]. It might be argued that comparable physical behavior in the two systems is a direct consequence of their structural similarities. However it should also be noted that the electronic band structure of URu_{0.29}Ge₂ is likely strongly influenced by contribution of $4d$ electrons due to the Ru constituent. In addition, when interpreting the thermodynamic and electrical transport properties of URu_{0.29}Ge₂, the presence of vacancies on the Ru site must be taken into account.

The formation of UNi_{1-x}Ge₂ ($x = 0.5$) with the orthorhombic CeNiSi₂-type structure (space group Cmcm) was first reported by Akselrud [5]. More recent studies on splat-cooled and arc-melted samples of this germanide indicated an antiferromagnetic order below the Neel temperature T_N of 54 K [7] and 45 K [8], respectively. The difference in the reported values of T_N can be attributed to disparate Ni content in various specimens. Indeed, our investigation of the ternary system U–Ni–Ge [6] established a homogeneity domain for the UNi_{1-x}Ge₂ phase with $0.35 \leq x \leq 0.6$ at 1173 K, and in our sample with $x = 0.45$ (cell parameters $a = 4.1062(2)$ Å, $b = 15.9272(8)$ Å and $c = 4.0461(2)$ Å) the antiferromagnetic ordering was found below $T_N = 50$ K. Moreover, our studies indicated that each synthesis starting from the ideal stoichiometry “UNiGe₂” results in the formation of at least two secondary phases, namely UNi₂Ge₂ and UNi_{1-x}Ge_{1+x}.

In order to get deeper insight into the electronic ground state of $\text{UNi}_{1-x}\text{Ge}_2$ and inspect the role of structural features on the physical properties of this compound, a high-quality polycrystalline sample ($x = 0.45$) was prepared and thoroughly studied by means of X-ray diffraction, magnetization, electrical resistivity and specific heat measurements. Furthermore, the magnetic structure of $\text{UNi}_{0.45}\text{Ge}_2$ was determined via neutron diffraction experiment.

Experimental details

Polycrystalline sample of $\text{UNi}_{0.45}\text{Ge}_2$ was synthesized by conventional arc-melting the corresponding amounts of high-purity elemental components (at least 99.5 at.%) under Ti/Zr-gettered argon atmosphere. To ensure good homogeneity, the pellet was turned over and re-melted several times. Subsequently, the button was sealed in an evacuated quartz tube and annealed at 1173 K for 2 weeks.

Small single crystals suitable for X-ray diffraction (XRD) were selected from crushed annealed polycrystalline ingots. The XRD intensities were collected on a Nonius Kappa CCD diffractometer with Mo $K\alpha$ radiation ($\lambda = 0.71073 \text{ \AA}$). The data collected on a half of Ewald sphere was integrated using the Denzo-Scalepack software [17]. The crystal structure was solved by direct methods using SHELXS-97 [18] and refined employing SHELXL-97 [18] after empirical absorption correction [19].

In order to examine the bulk sample quality, powder XRD experiment was performed in reflection mode on a Bruker D8 Advance diffractometer equipped with a LynxEye fast detector and working with monochromatized Cu $K\alpha_1$ radiation ($\lambda = 1.5406 \text{ \AA}$). The XRD patterns were indexed and refined using the Fullprof Suite software [20]. The sample was also checked by energy dispersive spectroscopy (EDS, Oxford EDS SDD X-max spectrometer) using a Jeol JSM 7100F scanning electron microscope. The EDS measurements were performed on a polished surface of the ingot.

Neutron diffraction experiments were performed on the 2-axes G4.1 diffractometer at the Laboratoire Léon Brillouin (Centre d'Etudes Nucléaires, Saclay, France) between 300 and 1.4 K, with a $\lambda = 2.426 \text{ \AA}$ wavelength. Approximately 1 cm^3 of powdered sample in vanadium holders was used for the experiments. The magnetic structure was refined using the Fullprof Suite software [20].

Magnetic measurements were carried out in the temperature range 2–400 K in external magnetic fields up to 70 kOe using a Quantum Design MPMS-XL magnetometer. In addition, magnetization isotherms were measured at a few constant temperatures from the interval 4.2–

10 K and in magnetic fields up to 140 kOe employing a home-made vibrating-sample magnetometer equipped with a step motor and installed inside a Bitter-type magnet [21]. The electrical resistivity was measured from room temperature down to 1.8 K in magnetic fields up to 140 kOe using a Quantum Design PPMS platform. The measurement was performed employing a conventional four-point ac technique, on a bar-shaped specimen with electrical leads attached to its surface by means of silver epoxy paste. Heat capacity measurements were done in the temperature range 2–300 K in magnetic fields up to 90 kOe using a relaxation method implemented in the same PPMS system.

Results and discussion

Crystal structure

The powder XRD pattern of UNi_{0.45}Ge₂ was indexed within the orthorhombic CeNiSi₂-type structure and with the lattice parameters $a = 4.1057(1)$ Å, $b = 15.9314(1)$ Å and $c = 4.0449(1)$ Å. In addition to the Bragg peaks of the main phase, a few reflections due to two impurity phases, *viz.* UGe₃ and UO₂ were detected. Their amounts, estimated via the Rietveld refinement, were about 1 and 0.5 wt.%, respectively. The presence of both secondary phases was corroborated in the EDS analysis (fig. 1a). The composition of the main phase in the sample investigated, derived from the EDS data, was 29(1)U-13(1)Ni-58(1)Ge that implies the chemical formula UNi_{0.45(4)}Ge₂.

The XRD peaks collected for a small single crystal of UNi_{0.45}Ge₂ were indexed with the orthorhombic unit cell with the dimensions $a = 4.1062(2)$ Å, $b = 15.9272(8)$ Å and $c = 4.0461(2)$ Å, in good agreement with the powder XRD study. The reflection conditions were compatible with the *Cmcm* space group (Table 1). Further crystal structure solution converged to the expected CeNiSi₂-type structure with four independent *4c* positions: one occupied by U atoms, one by Ni atoms and two by Ge atoms (Table 2). In concert with the previous reports [6-8], a partial occupancy was found on the Ni site, and the refined crystallographic formula UNi_{0.452(8)}Ge₂ perfectly agrees with the EDS result.

The crystal structure of UNi_{0.45}Ge₂ is shown in Figure 1b. The interatomic U-U distance within the uranium atoms chains is equal to 3.8814(4) Å, which is slightly longer than in UFe_{0.5}Ge₂ (3.863 Å [3]) or URu_{0.29}Ge₂ (3.868 Å [9]), yet significantly longer than in UGe₂ (3.816 Å [10]). The angles within the chains follow an opposite trend: they are larger in UGe₂ (63.85°) and gradually decrease going from URu_{0.29}Ge₂ (63.06°) to UFe_{0.5}Ge₂ (62.91°) and to UNi_{0.45}Ge₂ (62.83°).

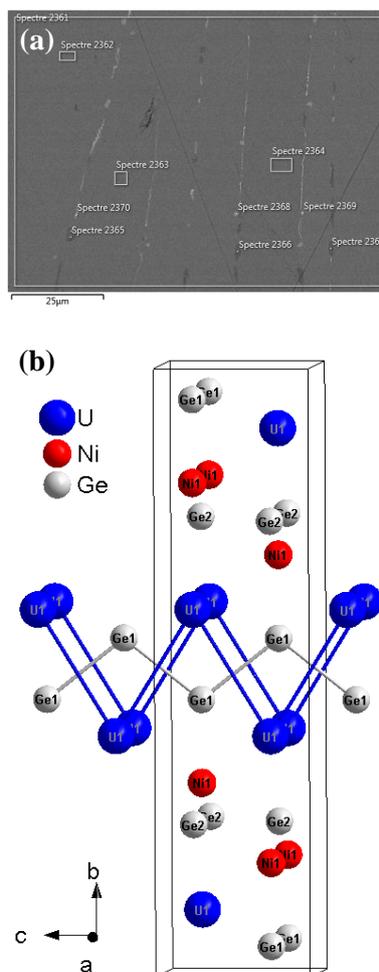


Figure 1 (a) Backscattered electron SEM image of a $UNi_{0.45}Ge_2$ sample with EDS-analyzed spots: $UNi_{0.45(4)}Ge_2$ appears as majority grey phases while the UGe_3 and UO_2 impurities appear as dark and light grey areas at the grain boundaries, respectively. (b) Crystal structure of $UNi_{0.45}Ge_2$ determined from single crystal XRD. The UGe_2 -like uranium chains are highlighted by blue bonds.

Further details of the crystal structure investigations are listed in Table 1 and may be obtained from the Fachinformationszentrum Karlsruhe, 76344 Eggenstein-Leopoldshafen, Germany (Fax: +49-7247-808-666; E-Mail: crysdata@fiz-karlsruhe.de, http://www.fiz-karlsruhe.de/request_for_deposited_data.html) on quoting the depository numbers CSD 2044292.

Table 1 – Single crystal X-ray diffraction data collection and refinement parameters for $UNi_{0.45}Ge_2$

Empirical formula	$UNi_{0.45}Ge_2$
Formula weight ($g\ mol^{-1}$)	409.63
Structure-type	CeNiSi ₂
Space group	Cmcm (no. 63)
Unit cell parameters (\AA)	$a = 4.1062(2)$
	$b = 15.9272(8)$
	$c = 4.0461(2)$
Unit cell volume (\AA^3)	264.62(2)
Z / calculated density ($g\ cm^{-3}$)	4 / 10.282
Absorption coefficient (mm^{-1})	86.339
Theta range	$1.00^\circ - 34.97^\circ$
Limiting indices	$-5 \leq h \leq 6$
	$-25 \leq k \leq 25$
	$-6 \leq l \leq 6$
Collected/unique reflections	2379 / 365
Crystal size (μm^3)	$60 \times 50 \times 50$
Absorption correction	Semi-empirical
R(int)	0.0868
Data / restraints / parameters	365 / 0 / 19
Goodness of fit on F^2	1.035
R indices [$I > 2\sigma(I)$]	$R1 = 0.0221$
	$wR2 = 0.0563$
Extinction coefficient	0.0024(4)
Largest difference peak and hole ($e\ \text{\AA}^{-3}$)	+1.93 / -2.29

Table 2 – Refined standardized atomic coordinates, occupancy rates and isotropic displacement parameters for $UNi_{0.45}Ge_2$

Atoms	Wyckoff position	X	Y	z	Occupancy	U_{eq} (\AA^2)
U1	$4c$	0	0.3960(1)	$\frac{1}{4}$	1	0.020(1)
Ni1	$4c$	0	0.1877(2)	$\frac{1}{4}$	0.452(8)	0.030(1)
Ge1	$4c$	0	0.0506(1)	$\frac{1}{4}$	1	0.035(1)
Ge2	$4c$	0	0.7495(1)	$\frac{1}{4}$	1	0.028(1)

Table 3 – Selected interatomic distances in UNi_{0.45}Ge₂ (Å)

U1	-4 Ge1	3.0052(5)	Ge1	-1 Ni1	2.183(3)
	-2 Ge2	3.076(2)		-2 Ge1	2.587(2)
	-2 Ge2	3.108(2)		-4 U1	3.0052(5)
	-4 Ni1	3.175(2)		-2 U1	3.206(2)
	-2 Ge1	3.206(2)			
	-1 Ni1	3.318(3)	Ge2	-2 Ni1	2.257(2)
	-2 U1	3.8814(4)		-2 Ni1	2.277(2)
				-4 Ge2	2.882(2)
Ni1	-1 Ge1	2.183(3)		-2 U1	3.076(2)
	-2 Ge2	2.257(2)		-2 U1	3.109(2)
	-2 Ge2	2.277(2)			
	-4 U1	3.175(2)			

Magnetic properties

Figure 2 gathers the main results of the magnetic properties measurements carried out on the polycrystalline sample of UNi_{0.45}Ge₂. As can be inferred from panel (a) of the figure, above about 100 K, the inverse magnetic susceptibility $\chi^{-1}(T)$ can be described by a modified Curie-Weiss law:

$$\chi(T) = \chi_0 + \frac{C}{T - \theta_p} \quad (1)$$

with the Curie constant $C = 0.94(3)$ emu K mol⁻¹, the paramagnetic Curie–Weiss temperature $\theta_p = 22(4)$ K and the temperature independent term $\chi_0 = 2.9(6) \times 10^{-4}$ emu mol⁻¹ as fitting parameters. The effective magnetic moment $\mu_{\text{eff}} = \sqrt{8C}$ is equal to $2.74(1) \mu_B$ per U atom that is a typical value for uranium based intermetallics, which usually exhibit partial delocalization of the 5f electrons. The large and positive value of θ_p hints at predominantly ferromagnetic character of exchange interactions between the U magnetic moments. While this finding suggests similarity to ferromagnetic URu_{0.29}Ge₂ and UCu_{0.45}Ge₂, a distinct sharp maximum in the $\chi(T)$ curve (see the upper inset to Figure 2a) clearly indicates an antiferromagnetic phase transition at the Néel temperature T_N of about 47 K (determined from a position of a maximum in $d\chi/dT$ [22]), in line with the previous report by Ohashi *et al.* [8].

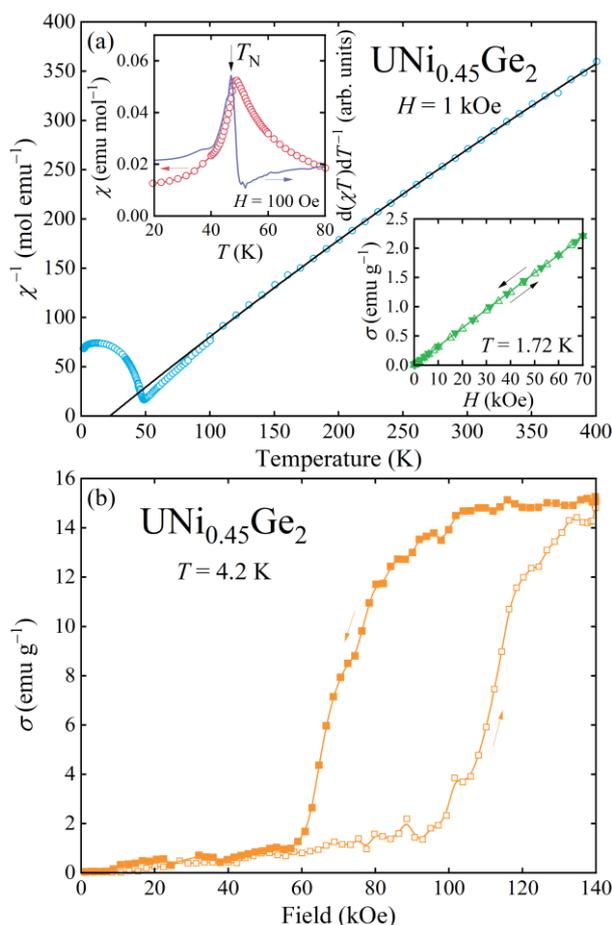


Figure 2 (a) Temperature dependence of the inverse magnetic susceptibility of $\text{UNi}_{0.45}\text{Ge}_2$. The black solid line is a fit of the modified Curie-Weiss law to the experimental data. The upper inset shows the magnetic susceptibility taken at low temperatures in a small field of 100 Oe and the derivative $d(\chi T)dT^{-1}$ (blue line). The lower inset displays the magnetization isotherm measured at $T = 1.72$ K with increasing (open symbols) and decreasing (filled symbols) magnetic field strength. (b) Magnetization isotherm measured at $T = 4.2$ K with increasing (open symbols) and decreasing (filled symbols) magnetic field.

The antiferromagnetic ground state in $\text{UNi}_{0.45}\text{Ge}_2$ was corroborated by measuring the magnetization as a function of magnetic field. As displayed in the lower inset to Figure 2a, the $M(H)$ isotherm taken at $T = 1.72$ K is linear and fully reversible up to $H = 70$ kOe. In turn, the experiment performed at $T = 4.2$ K revealed a sharp increase of the magnetization near 100 kOe that signals a metamagnetic-like phase transition (Figure 2b). In the strongest field applied, *i.e.* $H = 140$ kOe, the magnetization reached a value of $14.5(5)$ emu g^{-1} that corresponds to about $0.9 \mu_B$. On decreasing magnetic field, a large hysteresis was found, namely $M(H)$ returned to a linear behavior characteristic of antiferromagnetic state near 60 kOe that is at a field much

smaller than the metamagnetic critical field. Such a pronounced irreversibility in $M(H)$ suggests a first-order character of the metamagnetic transition.

Neutron diffraction

The neutron diffraction patterns were collected at selected temperatures from 70 K down to 1.4 K using neutron wavelength $\lambda = 2.426 \text{ \AA}$. For this experiment, an alloy slightly richer in nickel content was used, namely $\text{UNi}_{0.5}\text{Ge}_2$, in order to avoid any presence of ferromagnetic UGe_2 that could obfuscate determination of the magnetic structure of $\text{UNi}_{1-x}\text{Ge}_2$. As shown in Figure 3, the neutron patterns measured in the paramagnetic state was easily indexed with the proper structural model. The Rietveld refinement yielded the unit cell parameters $a = 4.1103(5) \text{ \AA}$, $b = 15.973(2) \text{ \AA}$ and $c = 4.0590(5) \text{ \AA}$, the atomic coordinates $y_{\text{U}} = 0.3969(5)$, $y_{\text{Ni}} = 0.1829(6)$, $y_{\text{Ge}1} = 0.0531(3)$, $y_{\text{Ge}2} = 0.7478(4)$, and the Ni-site occupancy equal to 0.499(1).

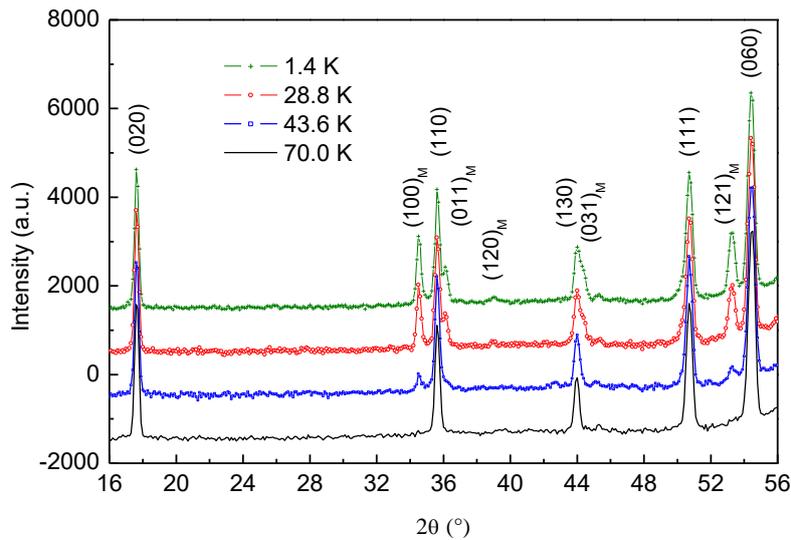


Figure 3 Selected angular domain of the neutron diffraction patterns of $\text{UNi}_{0.5}\text{Ge}_2$ measured at four different temperatures above and below T_N . The indexation is given for the main Bragg and magnetic peaks.

Below T_N , additional diffraction peaks of magnetic origin appeared, which could not be indexed within the C -mode orthorhombic cell and required lowering the symmetry to a P -mode orthorhombic cell with similar lattice parameters. Moreover, (hkl) reflections were observed for $h + k = 2n \pm 1$ only, highlighting an antiferromagnetic coupling of the magnetic moments carried by the atoms linked by a $(\frac{1}{2} \frac{1}{2} 0)$ translation. Using the propagation vector formalism,

the magnetic reflections were indexed with a $\vec{k} = (100)$ propagation vector. Additionally, the (010) magnetic peak was not observed at any temperature down to 1.4 K, implying that the magnetic moments are aligned along the crystallographic b -axis. While two different magnetic structures (Table 4) were compatible with these observations, after simulation of the neutron diffraction patterns, only model No. 1 was found to properly describe the experimental data. The Rietveld refinement of the experimental pattern measured at $T = 1.4$ K (Figure 4), based on the latter model, yielded the uranium magnetic moment of $1.95(3) \mu_B$.

Table 4 Hypothetical magnetic structures of UNi_{0.5}Ge₂ fitting the extinction rules observed on neutron diffraction patterns. Only the model No. 1 fits to the experimental pattern with refined $m_y = 1.95(3) \mu_B U^{-1}$ (see text for details)

Magnetic atom	Atomic coordinates			Model No.1			Model No. 2		
	X	Y	Z	M_x	M_y	M_z	M_x	M_y	M_z
U1	0	0.396	$\frac{1}{4}$	0	m_y	0	0	m_y	0
U2	$\frac{1}{2}$	0.896	$\frac{1}{4}$	0	$-m_y$	0	0	$-m_y$	0
U3	0	0.604	$\frac{3}{4}$	0	m_y	0	0	$-m_y$	0
U4	$\frac{1}{2}$	0.104	$\frac{3}{4}$	0	$-m_y$	0	0	m_y	0

As visualized in Figure 5, the magnetic structure of UNi_{0.5}Ge₂ consists of ferromagnetically ordered (a,c) planes, which are stacked antiferromagnetically along the b -axis in a sequence $-++-$. The uranium magnetic moments are aligned along the b -axis. The strong ferromagnetic ordering within the (a,c) planes rationalizes the positive sign of the paramagnetic Curie–Weiss temperature found for the antiferromagnetic compound studied. Furthermore, it is worth underlining that the coupling between the magnetic moments within the U-atoms chains is purely ferromagnetic, alike in the ferromagnetic superconductors mentioned in the Introduction. In turn, the magnetic moments alignment along the b -axis resembles the magnetic ordering in UCoGe and URhGe [1, 23], while the magnetic moments in UGe₂ point along the a -axis [10]. Interestingly, also in UNiSi₂ which is isostructural to UNi_{0.5}Ge₂, the U magnetic moments order parallel to the c -axis [24,25].

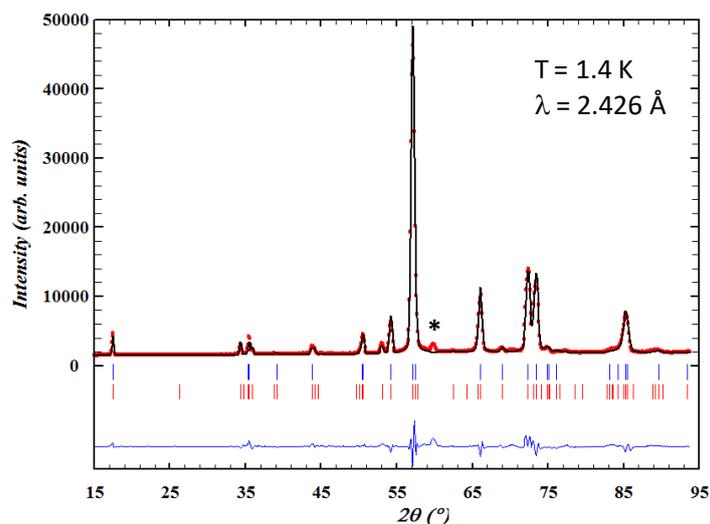


Figure 4 Neutron diffraction pattern of $UNi_{0.5}Ge_2$ measured at 1.4 K. The experimental data are plotted using red symbols and the calculated pattern is drawn as black line. The difference pattern is shown at the bottom (blue line). The vertical ticks mark the crystal lattice (upper row) and magnetic (lower row) Bragg peak positions. The asterisk indicates an impurity peak that was found also in the pattern taken in the paramagnetic state.

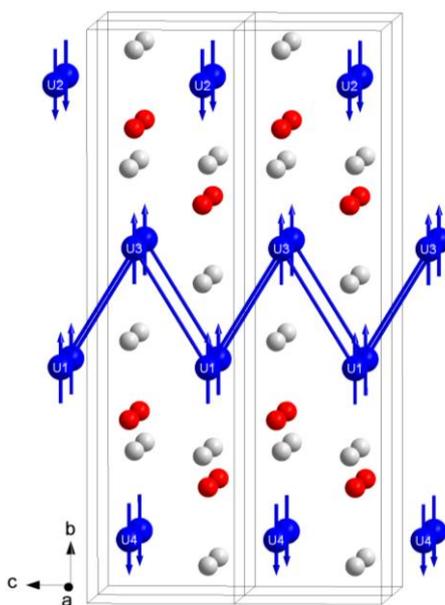


Figure 5. Magnetic structure of $UNi_{0.5}Ge_2$ at 1.4 K determined from neutron diffraction experiments. For clarity, only the U-atoms are labelled according to table 4. Ni-atoms are presented in red and germanium atoms in light grey. Two U-zig-zag chains are highlighted by blue lines.

Figure 6 shows the temperature variation of the ordered magnetic moment in $UNi_{0.5}Ge_2$ computed via Rietveld refinement from the $(100)_M$ magnetic peak intensities. The so-estimated

Néel temperature is about 50 K, in good agreement with the value of T_N derived from the magnetic susceptibility data of $\text{UNi}_{0.45}\text{Ge}_2$.

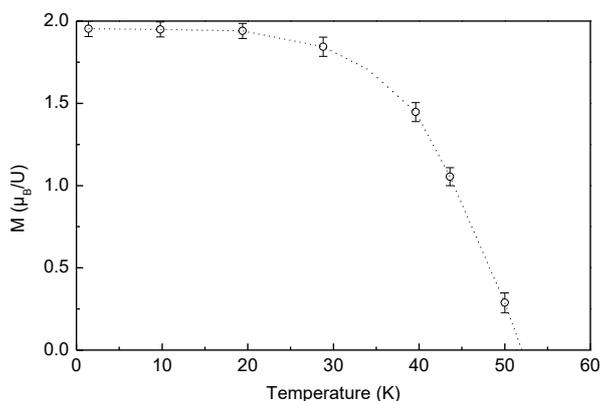


Figure 6 Temperature variation of the magnetic moment carried by U atoms in $\text{UNi}_{0.5}\text{Ge}_2$. The dashed line serves as a guide for the eyes.

Specific heat

Figure 7a displays the temperature dependence of the specific heat of $\text{UNi}_{0.45}\text{Ge}_2$. At 300 K, the specific heat is close to the Dulong-Petit limit of about $86 \text{ JK}^{-2}\text{mol}^{-1}$, calculated for one mole of molecules containing 3.45 atoms each. The antiferromagnetic ordering manifests itself as a pronounced mean-field type peak located near $T_N = 47 \text{ K}$. In applied magnetic field, this anomaly gradually broadens and shifts to lower temperatures with increasing the field strength (see Figure 7b), in concert with the antiferromagnetic character of the ordered state.

As shown in the inset to Figure 7a, below about 30 K, the experimental data can be described by the formula:

$$C_p(T) = C_{\text{el}}(T) + C_{\text{sw}}(T) + C_{\text{ph}}(T), \quad (2)$$

where:

$$C_{\text{el}}(T) = \gamma T \quad (3)$$

represents the electronic contribution with the Sommerfeld coefficient $\gamma = 54.2(6) \text{ mJ K}^{-2}\text{mol}^{-1}$,

$$C_{\text{sw}}(T) = \alpha T^{-0.5} e^{-\Delta/T} \quad (4)$$

describes the magnon contribution [26] with the coefficient $\alpha = 486(11) \text{ JK}^{-0.5}\text{mol}^{-1}$ and the energy gap $\Delta = 78.7(2) \text{ K}$ in the spin-waves spectrum, and

$$C_{\text{ph}}(T) = 9rR \left(\frac{T}{\Theta_D}\right)^3 \int_0^{\Theta_D/T} \frac{x^4 e^x}{(e^x - 1)^2} dx \quad (5)$$

is the conventional Debye formula for $r = 3.45$ atoms in the formula unit, the universal gas constant R and the Debye temperature $\Theta_D = 267(2) \text{ K}$. The obtained values of γ and Θ_D are close to those reported for the structurally related compound $\text{URu}_{0.29}\text{Ge}_2$

($\gamma = 60.8(4) \text{ mJ K}^{-2} \text{ mol}^{-1}$ and 240 K, respectively [9]), with the former one suggesting moderate electronic correlations in $\text{UNi}_{0.45}\text{Ge}_2$.

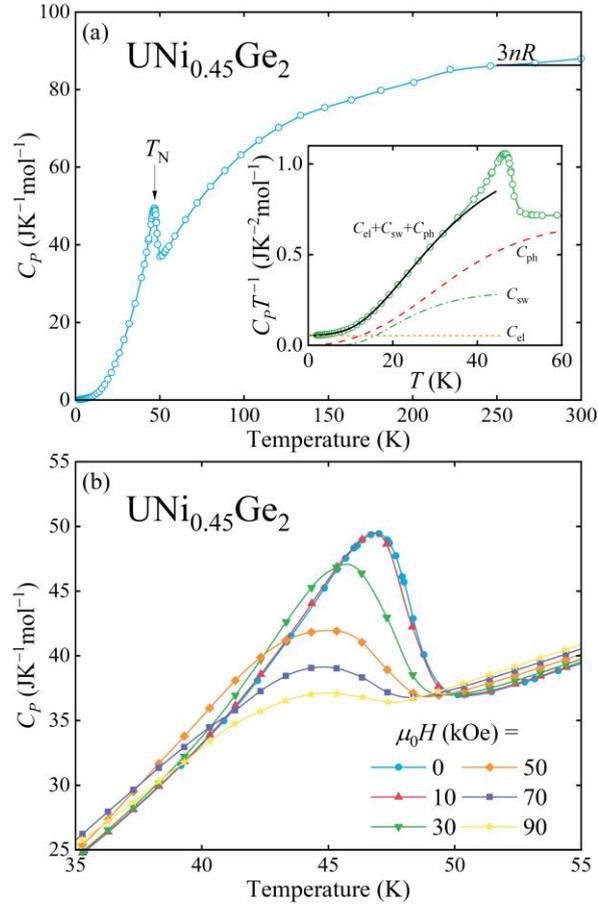


Figure 7 (a) Temperature variation of the specific heat of $\text{UNi}_{0.45}\text{Ge}_2$. The arrow marks the ordering temperature, and the horizontal line represents the Dulong-Petit limit $3nR$. The inset shows the low temperature region plotted as C_p/T vs T . The black solid line is the fit of Eq. 2 to the experimental data with its electronic (Eq. 3), magnon (Eq. 4) and phonon (Eq. 5) contributions plotted as orange dotted, green dash-dotted and red dashed lines, respectively. (b) Low-temperature dependencies of the specific heat measured in external magnetic field.

Electrical resistivity

The temperature dependence of the electrical resistivity of $\text{UNi}_{0.45}\text{Ge}_2$ is shown in Figure 8a. In the paramagnetic region, the resistivity hardly varies and $\rho(T)$ exhibits a faint, strongly smeared maximum near 170 K. Below T_N , the resistivity rapidly increases. The Néel temperature, defined as a minimum on the $d\rho/dT(T)$ curve (see the inset to Figure 8a), equals 47 K, in perfect agreement with the magnetic and heat capacity data.

The observed upturn of the resistivity below T_N is often observed in antiferromagnets as a result of opening an energy gap in the Fermi surface at the onset of the ordered state. However, in contrast to the regular behavior, $\rho(T)$ of $\text{UNi}_{0.45}\text{Ge}_2$ does not form any maximum at lower temperatures but transforms into a quasi-linear rise of the resistivity on decreasing temperature below about 20 K. The latter feature probably arises due to crystallographic disorder, inherently present in the Ni-deficient unit cell of $\text{UNi}_{0.45}\text{Ge}_2$. Another indication of the structural disorder in this compound is the rather large magnitude of electrical resistivity in the entire temperature range covered, along with its weak temperature dependence in the paramagnetic state.

Figure 8b presents the effect of external magnetic field (aligned perpendicular to the electric current) on the $\rho(T)$ curve below 100 K. With increasing the magnetic field strength, the singularity associated with the antiferromagnetic phase transition in $\text{UNi}_{0.45}\text{Ge}_2$ notably broadens and moves towards lower temperatures, in concert with the behavior of the specific heat. At the same time, the quasi-linear variation of the resistivity at the lowest temperatures stays almost intact in magnetic fields as strong as 90 kOe. This finding corroborates our conjecture of the crystallographic disorder being the principal source of that particular feature.

Another interesting effect apparent from Figure 8b is rather substantial magnetic field dependence of the resistivity at temperatures much above T_N . Such a behavior implies a significant magnetic contribution to the electrical resistivity in the paramagnetic state, originating from short-range magnetic exchange interactions and scattering conduction electrons on disordered magnetic moments, with possible input due to Kondo effect and/or crystalline electric field effect.

Furthermore, as can also be inferred from Figure 8b, deeply in the ordered state, the magnitude of the electrical resistivity becomes sharply dependent on magnetic field if its strength is large enough. In order to examine the latter effect, the resistivity of $\text{UNi}_{0.45}\text{Ge}_2$ was measured as a function of magnetic field at a few different temperatures $T < T_N$. The results are displayed in Figure 8c, in a form of the magnetoresistivity isotherms $\frac{\Delta\rho}{\rho}(H) = 100\% \cdot [\rho(H) - \rho(H = 0)]/\rho(H = 0)$. At each temperature, the magnetoresistivity abruptly drops down in strong fields, and the $\frac{\Delta\rho}{\rho}(H)$ curves exhibit hysteretic behavior. The hysteresis loops are very broad; their width reaches as much as 40 kOe at $T = 2$ K. Undoubtedly, the observed feature is an indicator of the metamagnetism in $\text{UNi}_{0.45}\text{Ge}_2$, which has a character of first-order magnetic field induced phase transition, as anticipated also from the magnetization data of the compound (see above).

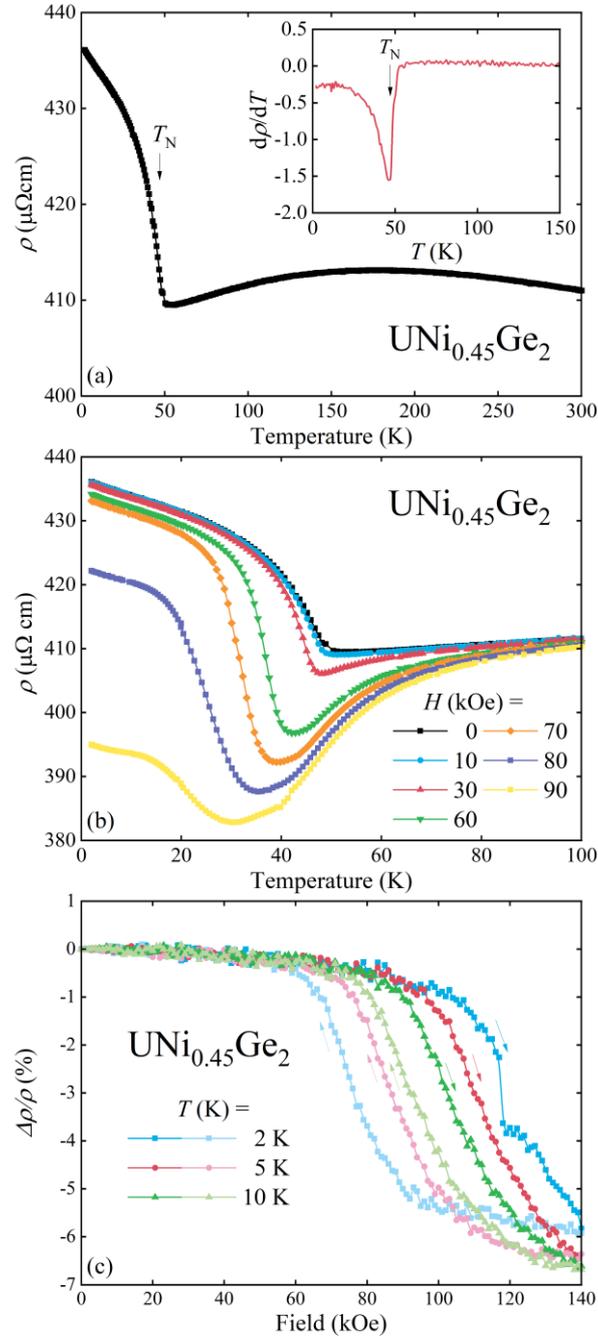


Figure 8 (a) Temperature dependence of the electrical resistivity of $\text{UNi}_{0.45}\text{Ge}_2$. The inset presents the temperature derivative of the low-temperature resistivity. The arrow marks the antiferromagnetic phase transition. (b) The electrical resistivity measured at low temperatures in different magnetic fields applied perpendicular to the electric current. (c) Transverse magnetoresistivity of $\text{UNi}_{0.45}\text{Ge}_2$ measured as a function of magnetic field at three different temperatures from the antiferromagnetically ordered region with increasing (dark symbols) and decreasing (light symbols) magnetic field strength.

Summary

In agreement with the literature reports for the UNi_{1-x}Ge₂ alloys [5-8], the compound UNi_{0.45}Ge₂ was found to crystallize with the CeNiSi₂-type structure and to exhibit a long-range antiferromagnetic ordering due to the magnetic moments carried on the U atoms. The Neel temperature determined from the magnetic susceptibility, heat capacity and electrical resistivity data equals 47 K. The magnetic structure revealed from the neutron diffraction experiment consists of ferromagnetically coupled U-atom layers stacked in a sequence $-++-$. In strong magnetic fields, the compound undergoes a field-induced first-order metamagnetic phase transition. The structural disorder associated with Ni atoms vacancies in the crystallographic unit cell influences the electrical transport in UNi_{0.45}Ge₂ and its behavior in external magnetic field.

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