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Ferromagnetism in structurally disordered $\text{UFe}_{0.39}\text{Ge}_2$

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ABSTRACT

A polycrystalline sample of the uranium ternary germanide $\text{UFe}_{0.39}\text{Ge}_2$ was examined by means of X-ray powder diffraction, DC magnetization and AC magnetic susceptibility, heat capacity and electrical resistivity measurements performed in wide ranges of temperature and magnetic fields. The experiments confirmed that the compound crystallizes with orthorhombic crystal structure of the CeNiSi_2 -type that is closely related to that of the ferromagnetic superconductor UGe_2 and orders ferromagnetically at 37 K. Moreover, it exhibits some features characteristic of ferromagnetic cluster glasses. The electrical transport in $\text{UFe}_{0.39}\text{Ge}_2$ is strongly influenced by structural disorder arising from the partly occupied Fe-sites in its crystallographic unit cell.

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1. Introduction

Uranium-based intermetallic phases often exhibit unique physical properties that result in most cases from interactions of electrons from conduction band with those from partly occupied $5f$ shells of periodically arranged uranium ions. In that context, uranium germanides have attracted special attention, ignited by a seminal discovery in 2000 of unconventional superconductivity in the ferromagnetically ordered compound UGe_2 [1] that crystallizes with an orthorhombic unit cell of the ZrGa_2 type (space group $Cmmm$) [2,3]. This finding has been followed by an intense search for other materials harboring the coexistence of ferromagnetism and superconductivity that are commonly considered as antagonistic physical phenomena. As a result, two ternary germanides have been identified as superconducting ferromagnets, namely URhGe [4] and UCoGe [5], both crystallizing with the orthorhombic TiNiSi -type unit cell (space group $Pnma$) [6,7]. Interestingly, a common feature of the crystal structures of those ternaries and that of UGe_2 is the presence of zig-zag chains of the U atoms with the U-U distances close to the Hill limit for uranium compounds (i.e. 3.5 Å) [8].

Recently, we began systematic re-investigation of another family of ternary uranium germanides, namely $\text{UTE}_{1-x}\text{Ge}_2$, where TE stands for a transition-metal element. Our research revealed the formation of two novel ferromagnets, $\text{URu}_{0.29}\text{Ge}_2$ ($T_C = 63$ K) [9] and $\text{UOs}_{0.25}\text{Ge}_2$

($T_C = 54$ K, unpublished), crystallizing with a new type of monoclinic structure (space group $C2/c$). Moreover, we confirmed the existence of antiferromagnetic $\text{UNi}_{0.45}\text{Ge}_2$ ($T_N = 47$ K) [10] that crystallizes with the orthorhombic CeNiSi_2 -type unit cell (space group $Cmcm$). It is worth noting, that the monoclinic $\text{URu}_{0.29}\text{Ge}_2$ -type structure and the orthorhombic CeNiSi_2 -type structure can be considered as close derivatives of the orthorhombic ZrGa_2 type structure adopted by UGe_2 . In turn, an important distinction between those three structures is significantly different distances between the U-atom chains [9,10].

The compound $\text{UF}_{1-x}\text{Ge}_2$ is another representative of the $\text{UTE}_{1-x}\text{Ge}_2$ family, reported by Henriques *et al.* to form with an extended homogeneity range $0.58 < 1-x < 0.78$, [11]. It crystallizes with a TE -deficient orthorhombic structure of the CeNiSi_2 type, alike $\text{UNi}_{0.45}\text{Ge}_2$. In this paper, we report the results of our study of the physical properties of $\text{UF}_{0.39}\text{Ge}_2$, performed in wide ranges of temperature and magnetic field.

2. Experimental details

Several polycrystalline samples of $\text{UF}_{1-x}\text{Ge}_2$ ($0.55 \leq x \leq 0.80$) were obtained by arc-melting (Bühler MAM1 furnace) the corresponding amounts of the elemental components (purity of each > 99.5 at%) in purified argon atmosphere. To ensure homogeneity of the products, the buttons were turned over and remelted several times. Subsequently, they were enclosed in an evacuated quartz tube and annealed at 900 K for four weeks.

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Quality of the samples was checked using powder X-ray diffraction (XRD) in reflection mode on a Bruker D8 Advance diffractometer operating with monochromatized Cu K α 1 radiation ($\lambda = 1.5406 \text{ \AA}$), equipped with a LynxEye fast detector to select X-ray energy and thus remove the Fe-fluorescence signal. The diffraction patterns were analyzed by the Rietveld method using the Fullprof Suite software [12]. The composition of the samples was verified using energy dispersive spectroscopy (SDD X-Max 50 mm² Oxford Instruments EDS) on a scanning electron microscope (JEOL JSM 7100F SEM).

DC magnetic measurements were performed on a selected sample in the form of bulk piece (see below) in the temperature range 1.72–400 K and in magnetic fields up to 70 kOe. AC magnetic susceptibility was measured at 32–40 K in zero external DC magnetic field using an alternate probing magnetic field with an amplitude of 3.7 Oe and a frequency from 1 Hz to 1 kHz. Both experiments were carried out using a Quantum Design MPMS-7 SQUID (superconducting quantum interference device) magnetometer. Heat capacity was measured from 2 to 300 K and in magnetic fields up to 10 kOe using a thermal relaxation technique. Electrical resistivity of a bar-shaped sample was measured in the temperature range 2–300 K and in magnetic fields up to 90 kOe using a conventional four-probe AC method in the field cooling regime and upon decreasing the temperature. The heat capacity and electrical transport experiments were carried out employing a Quantum Design PPMS-9 platform.

3. Results and discussion

3.1. Chemical composition and crystal structure

Due to a non-congruent character of the formation of UFe_{1-x}Ge₂ and a large number of phases being in equilibrium in the ternary phase diagram U-Fe-Ge [11], several samples with different compositions were synthesized along the solid-solution line UFe_{1-x}Ge₂. It allowed to control the formation of secondary phases that was generally aimed at decreasing their amount as much as possible. In particular, we successfully avoided the contamination of the main phase by ferromagnetic UGe₂ ($T_C = 52 \text{ K}$ [13]), U₃Ge₅ ($T_C = 94 \text{ K}$ [14]), U₃Fe₄Ge₄ ($T_C = 17 \text{ K}$ [15]) and U₃₄Fe₄Ge₃₃ ($T_C = 28 \text{ K}$ [16]). In turn, in each sample we found some amount of the ferromagnetically ordered impurity U₃Fe₂Ge₇ ($T_C = 62 \text{ K}$ [17]). In our attempts, we allowed for the presence of paramagnetic phases, like e.g. UGe₃ [18], U₉Fe₇Ge₂₄ [19] or UFe₂Ge₂ [20,21], expecting them to be less deleterious to investigate the physical properties of the desired phase UFe_{1-x}Ge₂.

As the final result of all these efforts, we selected for the physical properties measurements a sample with the initial composition 1U-0.45Ge-2Ge, which (besides the major phase) contained only two additional phases in small amounts. The SEM-EDS analysis (see the inset to Fig. 1) revealed that the majority phase in this specimen forms large and homogeneous grains with an average composition 29.89U-11.54Fe-58.57Ge (in at%) that corresponds to UFe_{0.39(2)}Ge₂. This Fe-atom content is slightly lower than the lower limit ($1-x = 0.42$) derived by Henriques *et al.* for the solid solution UFe_{1-x}Ge₂ at 1173 K [11]. The two secondary phases were found exclusively at grain boundaries and had the compositions: 26.02U-15.77Fe-58.21Ge, which corresponds to U₃Fe₂Ge₇, and 22.91U-22.21Fe-54.88Ge. The latter phase significantly deviates from any other known ternary alloy in the system U-Fe-Ge and might be a novel ternary compound. Alternatively, it can be attributed to a Fe/Ge solubility domain in U₉Fe₇Ge₂₄ (note that 22.5U-17.5Fe-60.0Ge is expected for U₉Fe₇Ge₂₇).

Based on those findings, the powder XRD pattern (Fig. 1) was easily indexed with the orthorhombic unit cells of UFe_{1-x}Ge₂ and U₃Fe₂Ge₇. The remaining small extra diffraction peaks seen in the

pattern were found to appear at 2θ values near the Bragg peaks positions generated for U₉Fe₇Ge₂₄, yet with significant deviation from the expected intensities. Nevertheless, the Rietveld refinement was performed considering these three phases and the results are shown in Fig. 1 and summarized in Tab. 1. In particular, UFe_{1-x}Ge₂ has been confirmed to adopt the transition-metal-deficient orthorhombic CeNiSi₂-type structure [11] with the cell parameters $a = 4.0886(1) \text{ \AA}$, $b = 15.8339(3) \text{ \AA}$ and $c = 4.0505(1) \text{ \AA}$. It should be noted that the absence of splitting of the main Bragg peak (131) rules out the monoclinic distortion observed in URu_{0.29}Ge₂ [9]. The refined atomic coordinates agree with the structural model proposed by Henriques *et al.* [11], and the derived occupancy of the Fe-atom site leads to a chemical formula UFe_{0.32(1)}Ge₂, slightly poorer in iron than that obtained from the EDS experiments. The refinements showed that the content of the two impurities were 2(1) wt% of U₃Fe₂Ge₇ and about 3(1) wt% of "U₉Fe₇Ge₂₄".

3.2. Magnetic properties

Fig. 2 (a) shows the temperature dependence of the inverse molar magnetization divided by the magnetic field, $(M/H)^{-1}(T)$, of UFe_{0.39}Ge₂. Above about 130 K, it exhibits slightly curvilinear behavior, which can be described by the modified Curie-Weiss law $M/H(T) = \chi_0 + (\mu_{\text{eff}}^2/8)/(T - \theta_p)$, where μ_{eff} denotes the effective magnetic moment and θ_p stands for the paramagnetic Curie-Weiss temperature. Least-square fitting of that formula to the experimental data yielded $\mu_{\text{eff}} = 2.59 \mu_B$, $\theta_p = 30 \text{ K}$, and $\chi_0 = 1 \times 10^{-4} \text{ emu mol}^{-1}$. The so-obtained μ_{eff} is much smaller than the values predicted for free U³⁺ and U⁴⁺ ions (3.62 and 3.58 μ_B , respectively), but it is large enough to ascribe the observed magnetic properties of the compound exclusively to the uranium ions. The discrepancy can be attributed to partial delocalization of 5f electrons, magnetocrystalline anisotropy and/or crystal field effect. The positive sign of θ_p hints at predominance of ferromagnetic exchange interactions.

Below about 37 K, the $M(T)$ variation of UFe_{0.39}Ge₂ displays an anomaly with a Brillouin-like curvature (see the upper inset to Fig. 2(a)) that manifests a ferromagnetic phase transition. Pronounced difference between the magnetization curves measured in zero-field-cooled (ZFC) and field-cooled (FC) regimes results from formation and reorientation of ferromagnetic domains. Also the magnetization isotherm $M(H)$ measured at 1.7 K, the lowest temperature available in the experiment, is typical for long-range ferromagnetic ordering (see the lower inset to Fig. 2(a)). At 70 kOe, $M(H)$ achieves a value of about 9.9 emu g⁻¹, which corresponds to the magnetic moment of 0.72 μ_B per formula unit, being a reasonable value for uranium intermetallics.

As can be seen in Fig. 2(b), the difference between the $M(T)$ curves in the ordered region systematically diminishes with increasing magnetic field, and in 10 kOe it is visible only at the lowest temperature studied. At the same time, the anomaly associated with the magnetic ordering smears out and moves towards higher temperature. Both behaviors are characteristic of ferromagnets.

The temperature variation of the AC magnetic susceptibility χ_{AC} of UFe_{0.39}Ge₂ is plotted in Fig. 3. The real and imaginary components of the susceptibility (χ' and χ'' , respectively) show distinct maxima at T_{max} , which can be associated with the magnetic ordering at $T_C = 37 \text{ K}$.

The value of T_{max} in $\chi'(T)$ equals 36.1 K for the probing AC magnetic field frequency $f_{AC} = 1 \text{ Hz}$. With increasing f_{AC} , T_{max} increases up to 36.3 K in $f_{AC} = 1 \text{ kHz}$. Such a behavior of $\chi'(T)$ is often considered as a fingerprint of spin-glass-like ground state [22]. For spin glasses, one can identify T_{max} as a freezing temperature T_f and calculate its relative shift upon change of f_{AC} , $\delta T_f = \frac{\Delta T_f}{T_f \Delta \log f_{AC}}$. For typical non-magnetic metallic spin glasses with strong structural disorder, this parameter is of the order of 10^{-2} (e.g., $\delta T_f = 0.036$ in

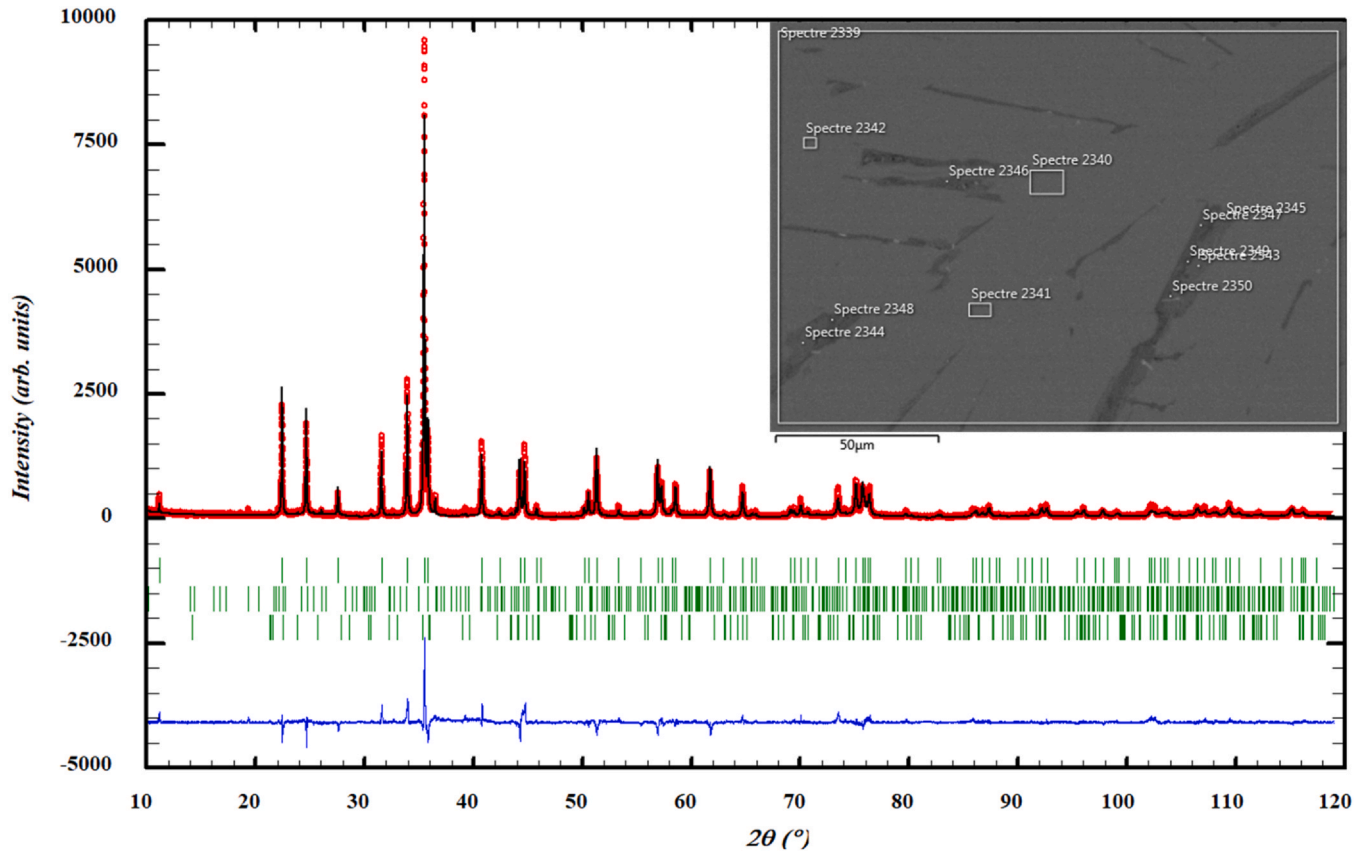


Fig. 1. Rietveld-refined powder XRD pattern of the sample "UFe_{0.45}Ge₂". Red circles display the experimental data, black line represents the theoretical pattern, and blue curve shows the difference between them. Vertical ticks mark the Bragg positions calculated for UFe_{1-x}Ge₂, U₀Fe₇Ge₂₄ and U₃Fe₂Ge₇ (top, middle and bottom ticks, respectively). The inset shows a backscattered electron SEM image of the sample studied with the examined areas marked; a majority phase (UFe_{0.39(2)}Ge₂) is visible as dominating gray area, while the two secondary phases (i.e. U₃Fe₂Ge₇ and a phase with the composition 22.91U-22.21Fe-54.88Ge, in at%) are represented as dark gray inclusions. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

Table 1
Crystal structure data of "UFe_{0.45}Ge₂" obtained by Rietveld analysis of the powder XRD pattern.

Formula	UFe _{1-x} Ge ₂						
Structure type	CeNiSi ₂						
Space group	Cmcm (no. 63)						
Cell parameters (Å)	a = 4.0886(1); b = 15.8339(3); c = 4.0505(1)						
Atomic positions	Atom	Wyckoff	x	y	z	Occ.	B _{iso} (Å ²)
	U1	4c	0	0.3966(1)	1/4	1	0.83(2)
	Fe1	4c	0	0.1978(9)	1/4	0.32(2)	2.3(4)
	Ge1	4c	0	0.0681(3)	1/4	1	4.5(2)
	Ge2	4c	0	0.7495(2)	1/4	1	0.69(7)
Reflections/ refined parameters	146/24						
Reliability factors	χ ² = 4.75 R _p = 25.2; R _{wp} = 27.8; R _{exp} = 12.78 R _{Bragg} = 12.1; R _f = 6.49						

Pr₂NiSi₃ [23], and $\delta T_f = 0.025$ in or URh₂Ge₂ [24]). The value of δT_f estimated for UFe_{0.39}Ge₂ is as small as 0.002, i.e. one order of magnitude lower than expected for spin glasses. Nevertheless, it is worth noting that it is similar to those reported for ferromagnetic cluster glasses with large spin clusters, such as Pr₂CuSi₃ ($\delta T_f = 0.001$) and Nd₂CuSi₃ ($\delta T_f = 0.002$) [25].

3.3. Heat capacity

Fig. 4 (a) shows temperature dependence of the specific heat C_p of UFe_{0.39}Ge₂ measured in zero magnetic field. As seen, at room temperature C_p attains a value of about 88 J K⁻¹ mol⁻¹, which is close to the Dulong-Petit limit $3nR$ (where n is a number of atoms per

molecule and R is the universal gas constant) calculated for the compound studied ($n = 3.39$), i.e. 85 J K⁻¹ mol⁻¹. The ferromagnetic phase transition at $T_c = 37$ K manifests itself in the $C_p(T)$ curve as a λ -shaped anomaly. However, the peak in the heat capacity data is rather broad and its shape is similar to that reported for the cluster-glass systems U₂RhSi₃ [26], U₂IrSi₃ [27,28], and U₂CoSi₃ [29].

The inset to Fig. 4(a) displays C_p/T of UFe_{0.39}Ge₂ plotted as a function of T^2 . Below about 6 K, the experimental curve exhibits nearly linear behavior, which can be described as a sum $C_p(T) = \gamma T + \beta T^3$, where the first term represents the Sommerfeld contribution, while the second term accounts for the phonon contribution approximated by the Debye law. The numerical analysis yielded $\gamma = 123(1)$ mJ K² mol⁻¹ and $\beta = 3.9(2) \times 10^{-4}$ mJ K⁴ mol⁻¹. Using the formula

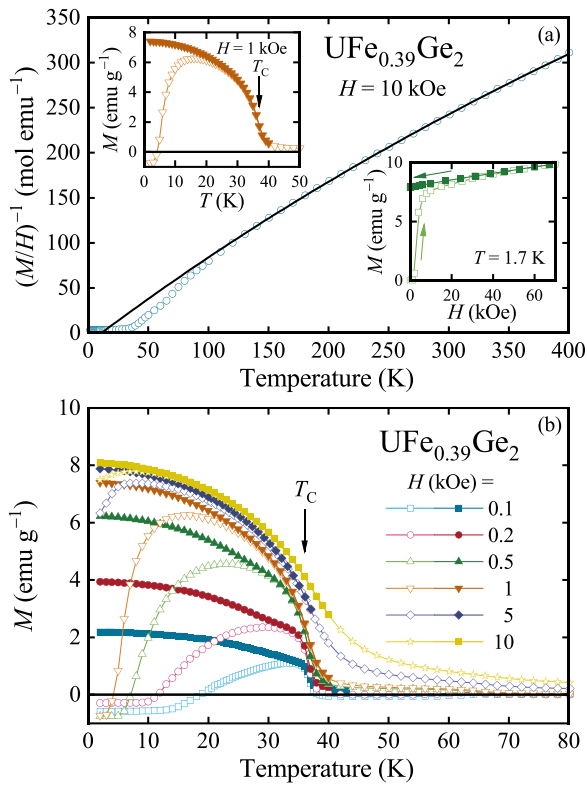


Fig. 2. (a) Temperature dependence of inverse molar magnetization divided by the magnetic field $(M/H)^{-1}$ of $\text{UFe}_{0.39}\text{Ge}_2$; solid line is a fit of the modified Curie-Weiss law to the experimental data (for details see the text). Upper inset: low-temperature variation of magnetization M measured in zero-field-cooling (closed symbols) and field-cooling (open symbols) regimes; an arrow marks temperature of magnetic ordering T_C . Lower inset: M measured at the lowest temperature available as a function of increasing (open symbols) and decreasing (closed symbols) external magnetic field H . (b) $M(T)$ studied in various magnetic fields.

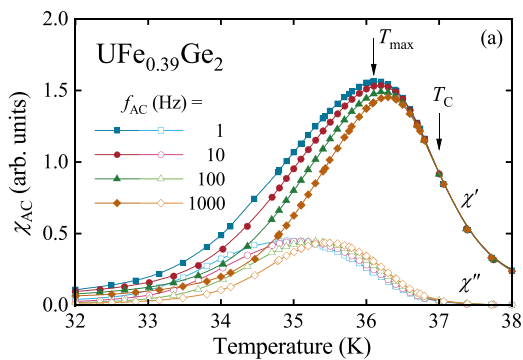


Fig. 3. Dynamic magnetic susceptibility χ_{AC} of $\text{UFe}_{0.39}\text{Ge}_2$ measured as a function of temperature with various frequencies f ; χ' and χ'' stand for the real and imaginary part of the susceptibility, respectively.

$\Theta_D = \sqrt[3]{1944n/\beta}$, one finds the Debye temperature $\Theta_D = 257(5)$ K that is of the order of magnitude characteristic of intermetallic compounds.

Remarkably, the Sommerfeld coefficient derived for $\text{UFe}_{0.39}\text{Ge}_2$ has a significantly higher value than usually observed in U-bearing systems. Similar enhancement of the γ coefficient in uranium intermetallics is often considered as originating from the presence of strong electronic correlations. However, an enlarged linear contribution to the low-temperature specific heat is also a characteristic feature of structurally disordered systems [22]. The excess of specific heat results from the magnetic entropy surviving to low temperatures due to frustrated and degenerate nature of the ground state. In

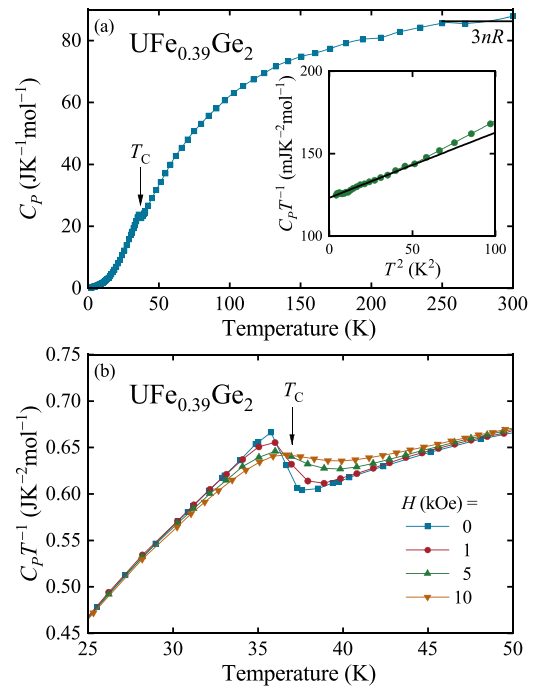


Fig. 4. (a) Temperature variation of specific heat C_p of $\text{UFe}_{0.39}\text{Ge}_2$; an arrow marks the ordering temperature T_C and the horizontal line points at the Dulong-Petit limit calculated for the compound. Inset: low temperature C_p/T -ratio as a function of T^2 ; solid line is a fit of a sum of the Debye- T^3 law and the Sommerfeld coefficient to the experimental data (for details see the text). (b) $C_p T^{-1}$ vs. T measured in magnetic field.

some special cases such enhancement in C/T could be a false indicator of heavy-fermion behavior [30].

In this context, it is worth recalling that the γ value reported for the crystallographically ordered ferromagnet UGe_2 equals $33 \text{ mJ K}^{-2} \text{ mol}^{-1}$ (see Ref. [31] and references therein), while that derived for the closely related ferromagnetic compound $\text{URu}_{0.29}\text{Ge}_2$ with partly occupied Ru-sites is twice larger ($61 \text{ mJ K}^{-2} \text{ mol}^{-1}$ [9]). By analogy, given the inherent crystallographic disorder in $\text{UFe}_{0.39}\text{Ge}_2$, the obtained large value of γ seems to originate from its structural features, and rather unlikely from strong electronic correlations.

As can be seen in Fig. 4(b), upon applying the magnetic field, the anomaly in $C_p(T)$ associated with the magnetic phase transition at T_C significantly broadens and moves towards higher temperature, which confirms the ferromagnetic-like character of the ordering. Similar behavior was observed in ferromagnetic $\text{URu}_{0.29}\text{Ge}_2$ [9].

3.4. Electrical resistivity

Fig. 5 (a) displays the temperature dependence of the electrical resistivity of $\text{UFe}_{0.39}\text{Ge}_2$. Overall, the magnitude of ρ is rather large and hardly varies with changing temperature. Both features are typical for metals with strong crystallographic disorder.

Near 190 K, the $\rho(T)$ curve forms a broad maximum characteristic of semimetallic systems. At $T_C = 37$ K, $\rho(T)$ exhibits a sharp dip, followed by rapid upturn in the ordered state. The latter behavior hints at more complex spin arrangement in $\text{UFe}_{0.39}\text{Ge}_2$ than a simple collinear ferromagnetism. Interestingly, a very similar deep minimum at the respective T_C was observed before for U_2CoSi_3 [29] and U_2IrSi_3 [27,28], where it was identified as a signature of cluster glass state in metallic systems with disordered crystal structures. It seems likely that the electronic transport behavior in $\text{UFe}_{0.45}\text{Ge}_2$ can also be attributed to the emergence at low temperature of ferromagnetic cluster-glass regime.

Most remarkably, the upturn in $\rho(T)$ can be entirely suppressed by as small magnetic field as 1 kOe (see Fig. 5(b)). The resistivity

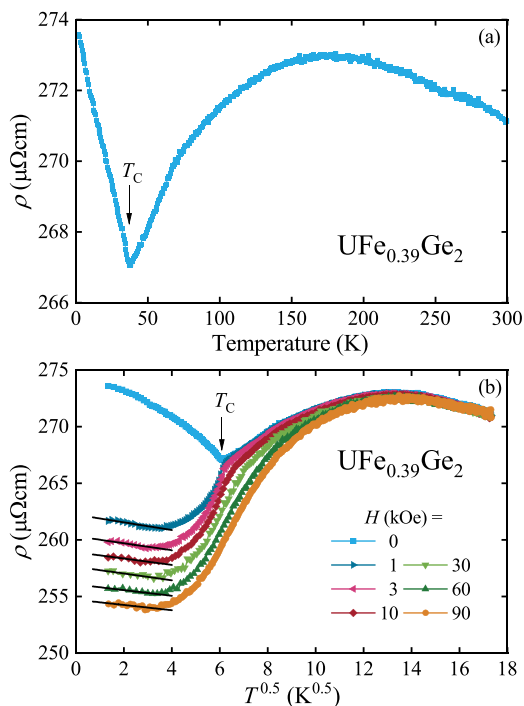


Fig. 5. (a) Electrical resistivity ρ of $\text{UFe}_{0.39}\text{Ge}_2$ as a function of temperature. (b) ρ vs. $T^{0.5}$ measured in various magnetic fields; solid lines show $\rho(T) \propto -T^{0.5}$ dependence at low temperature.

measured in applied field varies with temperature in a manner expected for ferromagnets, i.e. $\rho(T)$ shows a kink at T_C , and its position shifts towards higher temperatures with increasing the field strength. Also the magnitude of the resistivity diminishes with increasing field, signaling gradual suppression of ferromagnetic fluctuations.

Below about 16 K, the resistivity measured in finite external magnetic field increases with decreasing temperature. As can be inferred from Fig. 5(b), $\rho(T)$ has there a $-T^{0.5}$ slope that is independent of the strength of magnetic field at least up to 90 kOe. The relative change of the resistivity in this temperature range is only +0.3%. Such a small and field-independent change of the resistivity with the $-T^{0.5}$ variation points at possible presence of quantum interference effects, such as electron-electron interactions or weak localization. Quantum correction to $\rho(T)$ of $\text{UFe}_{0.39}\text{Ge}_2$ can be expected to occur because of distinct crystallographic disorder in its unit cell, and would manifest itself in a similar manner as reported for several disordered U-based intermetallics, such as U_2CoSi_3 [29] or U_2NiSi_3 [32].

4. Summary

Our studies confirmed that $\text{UFe}_{0.39}\text{Ge}_2$ crystallizes with the Ni-deficient orthorhombic CeNiSi_2 -type structure and orders ferromagnetically at $T_C = 37$ K. The phase transition manifests itself as pronounced anomalies in the temperature variations of magnetization, AC magnetic susceptibility, specific heat and electrical resistivity of the compound, and the evolution of that phase transition in external magnetic field is in accord with its ferromagnetic character. The vacancies on the Fe-sites are a source of significant crystallographic disorder that leads to some additional features in the temperature characteristics studied, i.e. some frequency dependence of the dynamic susceptibility, enlarged electronic contribution to the specific heat, large magnitude of the resistivity and its small, field-independent increase at the lowest temperatures. Another possible explanation of the anomalous behavior of the thermodynamic and

transport properties of $\text{UFe}_{0.39}\text{Ge}_2$ near T_C could be the formation of ferromagnetic cluster-glass state, but this conjecture needs to be verified by further experimental studies.

CRedit authorship contribution statement

Maria Szlawka: Investigation, Formal analysis, Writing – original draft. **Mathieu Pasturel:** Investigation, Formal analysis, Writing – original draft. **Dariusz Kaczorowski:** Formal analysis, Writing – review & editing. **Adam Pikul:** Formal analysis, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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