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Highlights

Ferromagnetic Ordering in UFe_{0.40}Ge₂ Studied by ⁵⁷Fe Mössbauer Spectroscopy

Adam P. Pikul, Rafał Idczak, Piotr Sobota, Wojciech Nowak, Mathieu Pasturel, Vinh Hung Tran

• UFe_{0.40}Ge₂ has been confirmed to order ferromagnetically below $T_{\rm C}$ = 37 K.

• The compound has been studied by Mössbauer spectroscopy.

• The experiments have confirmed that the ferromagnetism in $UFe_{0.40}Ge_2$ is purely long-range in nature.

• The hypothesis of a ferromagnetic cluster glass below $T_{\rm C}$ has been discarded.

Ferromagnetic Ordering in UFe_{0.40}Ge₂ Studied by ⁵⁷Fe Mössbauer Spectroscopy

Adam P. Pikul^{a,*}, Rafał Idczak^{b,a}, Piotr Sobota^{b,a}, Wojciech Nowak^{b,a}, Mathieu Pasturel^c, Vinh Hung Tran^a

^aInstitute of Low Temperature and Structure Research, Polish Academy of Sciences, ul. Okólna 2, 50-422 Wrocław, Poland ^bInstitute of Experimental Physics, University of Wrocław, pl. Maksa Borna 9, 50-204 Wrocław, Poland ^cUniv Rennes, CNRS, Institut des Sciences Chimiques de Rennes - UMR 6226, 35042 Rennes, France

Abstract

A polycrystalline ⁵⁷Fe-isotope-enriched sample of UFe_{0.40}Ge₂ was investigated by X-ray powder diffraction, magnetization and Mössbauer spectra measurements. The compound was confirmed to crystallize with the orthorhombic CeNiSi₂-type structure (space group *Cmcm*) and with the lattice parameters a = 4.0759(2) Å, b = 15.8107(7) Å, and c = 4.0379(2) Å, being close to those reported previously for a very similar composition. Magnetic properties measurements showed that UFe_{0.40}Ge₂ orders ferromagnetically at $T_{\rm C} = 37$ K with the effective magnetic moment of 2.57(3) $\mu_{\rm B}$ and the ordered magnetic moment at 70 kOe of about 0.64(1) $\mu_{\rm B}$, being fully consistent with the earlier report on physical properties of UFe_{0.39}Ge₂. ⁵⁷Fe Mössbauer spectra measurements performed both in the field-cooled and zero-field-cooled regimes rulled out the formation of ferromagnetic cluster-glass or spin-glass state, and confirmed that the ferromagnetism in the studied system is purely long-range in nature. A small contribution of about 0.24–0.36 $\mu_{\rm B}$ to the total magnetic moment from the iron atoms was observed.

Keywords: A. Uranium intermetallics, A. Uranium germanides, D. Ferromagnetic ordering, E. X-ray powder diffraction, E. Magnetization measurements, E. Mössbauer spectroscopy

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1. Introduction

Uranium germanides have attracted much attention from the 29 scientific community, following the discovery of unconven- 30 tional superconductivity in the ferromagnetically ordered com- 31 pound UGe₂ [1], which crystallizes in the orthorhombic ZrGa₂- ³² type structure (space group Cmmm) [2, 3]. As a result of in- 33 tense search for other compounds exhibiting the coexistence 34 of ferromagnetism and superconductivity, which so far were 35 8 considered to be mutually exclusive, two ternary uranium ger- 36 9 manides, namely URhGe and UCoGe, have been reported to be 37 10 superconducting ferromagnets [4, 5]. They crystallize with the 11 orthorhombic unit cell of the TiNiSi type (space group Pnma), 12 and their common feature with UGe2 is the presence of uranium ³⁸ 13 zig-zag chains within the uranium layers. 14

Very similar uranium zig-zag chains have also been found in ³⁹ 15 another group of uranium germanides, namely the compounds 16 $UT_{1-x}Ge_2$ (where T = Ru, Os, Ni and Fe), which all crystallize 17 in certain derivatives of the same orthorhombic type of crystal 18 structure. In particular, URu_{0.29}Ge₂ and UOs_{0.25}Ge₂ form with 19 a monoclinically distorted CeNiSi₂-type unit cell (space group 20 C2/c [6, 7]), while UNi_{0.45}Ge₂ and UFe_{0.39}Ge₂ crystallize in a 21 Ni-deficient CeNiSi₂ structure type (space group *Cmcm*) [8, 9]. 22 The compounds with Ru, Os and Fe order ferromagnetically at 47 23 63 K [6], 45 K [7] and 37 K [9], respectively, and only the Ni- 48 24 bearing phase was found to order antiferromagnetically at 47 K $^{\scriptscriptstyle 49}$ 25 [8]. 26 51

*Corresponding author *Email address:* a.pikul@intibs.pl (Adam P. Pikul) The ferromagnetic order in UFe_{0.39}Ge₂ at $T_{\rm C} = 37$ K occurs in the presence of significant crystallographic disorder. That disorder strongly influences the low-temperature physical properties of the compound (especially its transport properties), suggesting the formation of a ferromagnetic cluster glass [9]. To verify the latter hypothesis, we have investigated that compound by measuring its ⁵⁷Fe Mössbauer spectra at various temperatures from the magnetically ordered range and in two different cooling regimes. In the following, we present the results of those experiments along with their analysis for the formation of glassy states.

2. Material and Methods

A polycrystalline sample with a nominal composition of UFe_{0.40}Ge₂ (that is, to the nearest 0.3%, identical to the previous composition) was prepared by conventional arc melting the elemental components (with purity higher than 99.5% in each case) with about 5 at.% iron substitution by the ⁵⁷Fe isotope in purified argon atmosphere. During synthesis, the ingot was turned over and remelted several times to ensure good homogeneity. Subsequently, the specimen was annealed for four weeks at 900 K in an evacuated quartz tube to ensure good homogeneity. The mass loss at the end of the procedure was negligible.

Quality of the product was checked by X-ray powder diffraction (XRD) using a PANalytical X'Pert Pro diffractometer equipped with a Cu K_{α} radiation source. The experimental XRD pattern was analyzed by the Rietveld method using the Fullprof software [10]. Magnetic properties of the sample were

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Figure 1: Powder X-ray diffraction pattern of UFe_{0.40}Ge₂ (red circles) plotted along with results of the Rietveld refinement of its crystal structure (black curve). Upper and lower green ticks indicate Bragg positions for the major (UFe_{0.40}Ge₂) and the impurity (U₉Fe₇Ge₂₄) phases, respectively. Blue curve shows the difference between the experimental and the theoretical patterns.

studied using a Quantum Design MPMS-XL SQUID magne tometer in the temperature range 1.8 – 300 K and in applied ⁸⁶
 magnetic field up to 70 kOe.

The ⁵⁷Fe Mössbauer spectra of the powdered UFe_{0.40}Ge₂ ⁸⁸ 58 sample were measured in transmission geometry (TMS) with a 89 59 conventional constant-acceleration spectrometer, using a ⁵⁷Co- ⁹⁰ 60 in-Rh standard source with a full width at half maximum 91 61 (FWHM) of 0.22 mm/s. The temperature in the range of 4.3 - 9262 300 K was controlled using a variable-temperature insert in an 93 63 Oxford Instruments Spectromag cryostat. The obtained TMS ⁹⁴ 64 spectra were analyzed using a least-squares fitting procedure 95 65 to determine the parameters of the hyperfine interactions, such 96 66 as the magnetic hyperfine field $B_{\rm hf}$, the isomer shift IS, the ⁹⁷ 67 quadrupole splitting QS, the quadrupole shift for the mag-98 68 netic sextet ϵ , the absorption spectral areas C, and the spectral 99 69 linewidths Γ . The IS values presented in this paper refer to the 70 100 α -Fe standard at room temperature. 71

72 **3. Results and Discussion**

73 3.1. Crystal structure

The experimental XRD pattern of the ⁵⁷Fe-isotope-enriched 74 UFe_{0.40}Ge₂ sample (Fig. 1) was readily indexed within the Fe-75 deficient orthorhombic CeNiSi2-type structure, already solved 76 for that germanide [9], with a small addition of the tetrag-77 onal paramagnetic $U_9Fe_7Ge_{24}$ phase [11]. (The presence of 10178 that secondary phase was deliberately allowed, as discussed in102 79 Ref. [9]). The Rietveld refinement of those two crystal struc-103 80 tures, performed using the crystallographic data reported in₁₀₄ 81 Refs. [9] and [11], gave relative amounts of the two phases of 105 82 about 95 and 5 wt.%, respectively, indicating good quality of 106 83 84 the prepared sample. The elevated background due to iron flu-107 85 orescence could hide Bragg reflections from other secondary₁₀₈



Figure 2: Temperature dependence of inverse magnetic susceptibility χ^{-1} of UFe_{0.40}Ge₂; 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 mass magnetization σ measured in field-cooling regime; an arrow marks the ferromagnetic ordering temperature $T_{\rm C}$. Lower inset: σ as a function of increasing (open symbols) and decreasing (closed symbols) applied magnetic field *H*.

phases (if present at all), but their amount would still be less than 1-2 wt%.

The refined cell parameters for UFe_{0.40}Ge₂ are a = 4.0759(2) Å, b = 15.8107(7) Å, and c = 4.0379(2) Å. These values are somewhat closer to those reported by Henriques *et al.* for a similar chemical composition (a = 4.083(5) Å, b = 15.805(5) Å, and c = 4.030(5) Å[12]) than our previous results reported for a polycrystalline sample with the initial composition 'UFe_{0.45}Ge₂' (a = 4.0886(1) Å, b = 15.8339(3) Å, and c = 4.0505(1) Å), suggesting the existence of a small homogeneity domain for the studied phase. Nevertheless, the results show that the sample synthesized for the study described in this paper is essentially identical to the previous sample described in Ref. [9].

3.2. Magnetic properties

Fig. 2 shows results of the magnetic properties measurements carried out for $UFe_{0.40}Ge_2$. As can be seen, above about 130 K the inverse magnetic susceptibility exhibits featureless and slightly curvilinear behavior characteristic of paramagnets. The experimental data can be easily described by the modified Curie-Weiss law:

$$\chi(T) = \frac{1}{8} \frac{\mu_{\text{eff}}^2}{T - \theta_{\text{p}}} + \chi_0, \qquad (1)$$

where μ_{eff} is the effective magnetic moment, θ_{p} stands for the Curie-Weiss temperature, and χ_0 denotes temperature independent Pauli-like contribution to the total magnetic susceptibility. The least-squares fitting of Eq. (1) to the experimental data resulted in the parameter values: $\mu_{\text{eff}} = 2.57(3) \,\mu_{\text{B}}$, $\theta_{\text{p}} = 23(2)$ K, and $\chi_0 = 8.6(5) \times 10^{-4}$ emu/mol, being fully consistent with those obtained previously for UFe_{0.39}Ge₂ ($\mu_{\text{eff}} = 2.59\mu_{\text{B}}$, $\theta_{\text{p}} = 30$ K, and $\chi_0 = 1 \times 10^{-4}$ [9]). The so-estimated value of

¹⁰⁹ μ_{eff} is lower than that expected for free U³⁺ and U⁴⁺ ions (3.62 ¹¹⁰ and 3.58 μ_{B} , respectively), but it is large enough to be attributed ¹¹¹ mainly to the uranium ions. The observed discrepancy may be ¹¹² due to partial delocalization of 5f electrons, magnetocrystalline ¹¹³ anisotropy or crystal field effects. The large and positive value ¹¹⁴ of θ_{p} indicates the presence of predominantly ferromagnetic in-¹¹⁵ teractions between the magnetic moments.

The pronounced Brillouin-like-shaped anomaly that is vis-116 ible in the temperature variation of the magnetization, $\sigma(T)$, 117 at about 37 K (see the upper inset to Fig.2) manifests ferro-118 magnetic ordering of UFe_{0.40}Ge₂, being in full agreement with 119 our previous finding [9]. The field dependence of the magne-120 tization, $\sigma(H)$, measured at the lowest temperature studied, *i.e.* 121 1.8 K (see the lower inset to Fig.2) confirms the ferromagnetic 122 character of the ordering. In the highest field σ reaches a value 123 of 8.83 emu/g, which corresponds to the ordered magnetic mo-124 ment of about 0.64 $\mu_{\rm B}$. It is close to the value reported for 125 $UNi_{0.39}Ge_2$ [9] and is within the range of values observed for 126 uranium intermetallics. 127

The above findings show that the magnetic properties of the sample used in the present study are well defined, and the UFe_{1-x}Ge₂ system under study exhibits reproducible magnetic properties.

132 3.3. TMS results

Selected TMS spectra measured for the UFe_{0.40}Ge₂ sample 133 are presented in Figs. 3 and 4. As can be seen, the spectra mea-134 sured at $T \ge 35$ K (Fig. 3), corresponding approximately to the 135 paramagnetic region of the studied compound, can be described 136 using two paramagnetic components. The major one, with the 137 relative intensity of about 95%, is a well-resolved symmetric 138 doublet. Given the XRD results described in Sec.3.1 as well 139 as in Ref. [9], this component can be ascribed to the compound¹⁶⁴ 140 UFe_{0.40}Ge₂, in which Fe atoms occupy a single crystallographic¹⁶⁵ 141 site with symmetry lower than cubic. Deep in the magnetically¹⁶⁶ 142 ordered region (Fig. 4), *i.e.* at $T \leq 30$ K, this component can no¹⁶⁷ 143 longer be described by the doublet, and it must be described by168 144 a magnetic sextet with a small hyperfine field $B_{\rm hf} = 2.10(3) \,\mathrm{T}_{,169}$ 145 3.39(1) T and 3.55(2) T for the spectra measured at 30 K, 10 K¹⁷⁰ 146 and 4.3 K, respectively. 171 147

The second, minor component of the deconvoluted TMS172 148 spectra, with relative intensity being close to 5%, can¹⁷³ 149 be described by a single line component (a singlet) with¹⁷⁴ 150 IS = 0.06(5) mm/s at 300 K. As can be seen in Figs. 3 and 4,¹⁷⁵ 151 this component remains a singlet at every other temperature¹⁷⁶ 152 studied, thus showing the absence of a magnetic phase tran-177 153 sition at least down to 4.3 K. It can be attributed to the presence¹⁷⁸ 154 of the impurity phase U₉Fe₇Ge₂₄ (see Sec. 3.1 and Ref. [11]),¹⁷⁹ 155 which contains iron atoms and is known to remain paramag-180 156 netic down to the lowest measured temperature. It should be181 157 noted here that since the hyperfine parameters for the com-182 158 pound U₉Fe₇Ge₂₄ are not available in the literature, the assign-183 159 ment of the singlet to the impurity phase is based on the XRD184 160 results (Sec. 3.1)) and the fact that TMS measurements per-185 161 162 formed for some other U-Fe-Ge ternaries revealed similar IS values at 300 K. In particular, IS at 300 K for U₂Fe₃Ge and 163



Figure 3: TMS spectra of UFe_{0.40}Ge₂ measured at several temperatures from the range 35-300 K and in zero applied magnetic field, fitted by a sum (black curve) of one doublet and one singlet component (blue and red lines, respectively). For details of the fits see the text.

 $U_{34}Fe_{4-x}Ge_{33}$ is -0.03 mm/s [13] and 0.13 mm/s [14], respectively.

The TMS spectra shown in Figs. 3 and 4 can alternatively be described by only one component: an asymmetric doublet at $T \ge 35$ K or a Zeeman sextet below 35 K. It should be emphasized, however, that if one excludes the possibility of two components in the measured spectra, then the asymetry of the quadrupole doublet should be related to the preferred crystal orientation of the sample or to the presence of the Goldanskii-Karyagin effect [15, 16]. The first possibility is very unlikely since the TMS measurements were made on a powdered polycrystalline sample. The second possibility (the Goldanskii-Karyagin effect) is related to vibrational anistropy in the studied crystal. This effect is temperature dependent, so the spectral asymmetry should decrease with decreasing temperature, which is not the case. In Fig. 3 it is clearly seen that the shapes of the spectra measured at 300 K and 35 K are almost identical, so the Goldanskii-Karyagin effect cannot be responsible for the observed asymmetry of the quadrupole doublet. Taking the above into account, it is more likely that the spectra shown in Fig. 3 consist of two components rather than one doublet with an asymmetry of unknown origin.

The IS(T) dependence obtained from the above fits to the TMS spectra of UFe_{0.40}Ge₂ are shown in Fig. 5(a). In general,



Figure 4: TMS data collected for $UFe_{0.40}Ge_2$ at several temperatures from the magnetically ordered region and in zero applied magnetic field, fitted by a sum (black line) of one sextet and one singlet component (blue and red curves, respectively). FC marks the spectrum measured in zero magnetic field, after cooling the sample in the field of 3 T. For details of the fits see the text.

the isomer shift is dependent on the s-electrons density and is given by the relation [15]:

$$IS = \frac{4\pi}{5} Z e^2 R^2 \left(\frac{\delta R}{R}\right) \left[|\Psi_{\rm A}(0)|^2 - |\Psi_{\rm S}(0)|^2 \right], \tag{2}$$

where Z is the nuclear charge of the Mössbauer absorber, e is 186 the elementary charge, R is the radius of the probe nucleus, δR is 187 the difference between the radii in the ground and excited states, 188 and $|\Psi_{\rm S}(0)|^2$ and $|\Psi_{\rm A}(0)|^2$ are the s-electrons densities at the 189 probe nuclei in the source and absorber, respectively. At 300 K, 190 the IS parameter determined for the UFe_{0.40}Ge₂ compound is¹⁹⁸ 191 equal to 0.238(4) mm/s. This value lies between that reported199 192 earlier for U₃Fe₂Ge₇ (0.21(2) mm/s) [17] and that found for²⁰⁰ 193 $U_3Fe_4Ge_4$ (0.27(2) mm/s) [18], indicating a very similar elec-201 194 tron density at the iron nuclei in these three systems. 202 195

As expected from the second-order Doppler (SOD) effect,²⁰³ the *IS* values obtained for UFe_{0.40}Ge₂ increase concavely with²⁰⁴ decreasing temperature. Since the second-order Doppler shift²⁰⁵ depends on the lattice vibrations of the Fe atoms, the *IS*(*T*)²⁰⁶ dependence can be expressed in terms of the Debye approxi-²⁰⁷ mation of the lattice vibrations as [15]: ²⁰⁸

$$IS(T) = IS(0) + IS_{SOD}(T) =$$
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$$= IS(0) - \frac{9}{2} \frac{k_{\rm B}T}{Mc} \left(\frac{T}{\Theta_{\rm D}}\right)^3 \int_0^{\Theta_{\rm D}/T} \frac{x^3 dx}{e^x - 1}, \qquad (3)_{21}$$

where IS(0) denotes the temperature-independent isomer shift,²¹⁴ M is the mass of the nuclei of the absorber (here: ⁵⁷Fe), $k_{B^{215}}$



Figure 5: Temperature variations of the *IS* (a) and *QS* (b) parameters derived from the TMS spectra of UFe_{0.40}Ge₂. Black curves are fits of Eqs. (3) and (4) to the experimental data shown in panels (a) and (b), respectively (for details see the text).



Figure 6: TMS spectrum of $UFe_{0.40}Ge_2$ measured at $40\,K$ and in external magnetic field of $3\,T.$

is the Boltzmann constant and *c* is the speed of light in vacuum. Fitting of Eq. 3 to the experimental IS(T) values gave IS(0) = 0.357(2) mm/s and $\Theta_D = 434(23)$ K. Here, it should be noted that Θ_D determined from the TMS data is higher than the corresponding value of $\Theta_D = 257(5)$ K obtained from heat capacity measurements [9]. This discrepancy can be explained by the fact that the Mössbauer spectroscopy is only sensitive to the immediate neighbourhood of the Mössbauer probe atom (⁵⁷Fe) and for this reason the estimated Θ_D values from the TMS spectra should be treated as the local Debye temperature of the iron sublattice.

The QS(T) dependence obtained from the TMS spectra of UFe_{0.40}Ge₂ are shown in Fig. 5(b). The observed quadrupole splitting fully identify the presence of the electric field gradient (EFG) which is mainly proportional to the asymmetric charge distribution around the ⁵⁷Fe nucleus. The *QS* parameter at 300 K is equal to 0.342(2) mm/s, which is comparable to QS = 0.32(2) mm/s reported previously for U₃Fe₂Ge₇ [17].

Since the structure of U₃Fe₂Ge₇ can be seen as stacking of the²⁶⁹
 UFeGe₂ and UGe₃ layers along the *b*-axis, the charge distribu-²⁷⁰
 tion around the Fe nuclei in the U₃Fe₂Ge₇ system should indeed

 $_{219}$ be similar to that observed in UFe_{0.40}Ge₂.

In many intermetallic systems, the QS(T) dependence can be described by the empirical formula [19, 20]:

$$QS(T) = QS(0)(1 - AT^{3/2}), \qquad (4)_{272}$$

where QS(0) is the value of QS at 0 K and A is a²⁷³ constant. Fitting Eq. 4 to the experimental data yields²⁷⁴ QS(0) = 0.353(2) mm/s and $A = 6(1) \times 10^{-6} \text{ K}^{-3/2}$. The determined fitting parameters can be treated as material constants,²⁷⁶ since their physical meaning is not clearly defined.²⁷⁷

According to the results of our magnetic properties measure-278 225 ments (see Sec. 3.2 and Ref. [9]), the observed magnetic prop-226 erties of the $UFe_{0.40}Ge_2$ compound can be ascribed mainly to $^{\scriptscriptstyle 280}$ 227 the uranium ions. However, the appearance of the Zeeman sex-281 228 tets in the TMS spectra measured at $T \,{\leqslant}\, 30\,K$ may suggest that $^{^{282}}$ 229 the Fe sublattice establishes long-range magnetic correlations²⁸³ 230 below $T_{\rm C}$. This behavior is similar to that observed in other²⁸⁴ 231 uranium-iron ferromagnets, e.g. UFe2 [21], U2Fe3Ge [22] and²⁸⁵ 232 $U_3Fe_2Ge_7$ [17]. In all of them, the Mössbauer spectra collected²⁸⁶ 233 below their magnetic ordering temperatures indicate the pres-287 234 ence of small hyperfine field ($B_{\rm hf} < 5 \,\mathrm{T}$) at the ⁵⁷Fe nuclei. 235

On the other hand, the magnetic ⁵⁷Fe spectra observed be-²⁸⁹ 236 low 35 K may be due to the hyperfine magnetic field trans-290 237 ferred from the ferromagnetically ordered uranium sites. To²⁹¹ 238 shed more light on this problem, we measured the TMS spec-²⁹² 239 trum of UFe_{0.40}Ge₂ at 40 K, which is slightly above $T_{\rm C}$, and²⁹³ 240 in external magnetic field $B_{\text{ext}} = 3 \text{ T}$, applied parallel to the²⁹⁴ 241 propagation of the γ radiation beam. As can be seen in²⁹⁵ 242 Fig. 6, the collected spectrum reveals the presence of two Zee-²⁹⁶ 243 man components that consist of four lines, since in such a²⁹⁷ 244 configuration the observed line intensity ratio for the sextet298 245 should be 3:0:1:1:0:3 [16]. The first component, with a rel-299 ative intensity of 7(2)%, IS = 0.13(2) mm/s and effective hy-³⁰⁰ 247 perfine magnetic field $B_{\rm eff} = 2.8(2)$ T, can be attributed to the³⁰¹ 248 impurity phase. The second, major component corresponds³⁰² 249 to UFe_{0.40}Ge₂ and can be described by the Zeeman sextet³⁰³ 250 with $B_{\rm eff} = 3.78(2)$ T, IS = 0.333(7) mm/s and $\epsilon = 0.01(4)$ mm/s.³⁰⁴ 251 Since $B_{\text{eff}} = B_{\text{hf}} + B_{\text{ext}}$, the determined $B_{\text{eff}} = 3.78(2) \text{ T con}^{-305}$ 252 firms that the iron atoms in UFe_{0.40}Ge₂ have a small magnetic³⁰⁶ 253 moment. Assuming that the hyperfine coupling constant for³⁰⁷ 254 intermetallic compounds is in the range from 10 T/ μ_B to 15³⁰⁸ 255 T/μ_B [17, 22, 23], the hyperfine magnetic field $B_{hf} = 0.78(2) T^{309}_{,309}$ 256 obtained from the TMS spectrum measured for UFe_{0.40}Ge₂ at³¹⁰ 257 40 K and in $B_{\text{ext}} = 3$ T, corresponds to the magnetic moment of³¹¹ 258 iron $\mu_{\rm Fe}$ of about $0.05 - 0.08 \,\mu_{\rm B}$. This estimated value of $\mu_{\rm Fe}^{312}$ 259 is comparable with $\mu_{\rm Fe} \approx 0.2 \,\mu_{\rm B}$ inferred from the Mössbauer³¹³ 260 spectra measurements carried out for U₃Fe₂Ge₇ [17], and is³¹⁴ 261 small compared to the effective magnetic moment found for 262 UFe_{0.40}Ge₂. Thus, it does not change our conclusions about 263 the origin of magnetism in the latter compound (see Sec. 3.2³¹⁵ 264 and Ref. [9]). 265

The values of the *QS* and ϵ parameters obtained for the para-³¹⁶ magnetic and magnetically ordered states of UFe_{0.40}Ge₂, re-³¹⁷ spectively, can be used to determine the angle φ between *B*_{hf}³¹⁸ and the principal axis V_{zz} of the main electrical field gradient (EFG). Using the commonly used formula [17, 24]:

$$\varepsilon = \frac{1}{2}QS(3\cos^2\varphi - 1),\tag{5}$$

and taking QS = 0.353 mm/s and $\epsilon = 0.08$ mm/s, the angle $\varphi = 45.3^{\circ}$ was obtained.

Finally, the TMS technique can be used to confirm or rule out the formation of a ferromagnetic cluster-glass state in the UFe_{0.40}Ge₂ system [9]. For this purpose, we measured the Mössbauer spectra of the compound at 4.3 K, that is, deep in the magnetically ordered region, and in two different regimes. The first measurement (denoted as ZFC in Fig. 4) was carried out in zero applied magnetic field and after cooling the sample in zero external magnetic field. The second one (labelled as FC in Fig. 4) was also performed in zero magnetic field, but after cooling the sample in the external field of 3 T, *i.e.* in the nearly-saturated magnetization range.

For the ZFC spectrum, the sextet attributed to UFe_{0.40}Ge₂ could be described by IS = 0.353(4) mm/s, $\epsilon = 0.08(1)$ mm/s and $B_{\rm hf} = 3.55(2)$ T while for FC, the hyperfine parameters are IS = 0.349(2) mm/s, $\epsilon = 0.08(1)$ mm/s and $B_{\rm hf} = 3.52(2)$ T. Comparing these two sets of values, it can be seen that the two spectra are virtually identical. Moreover, the line intensity ratio estimated for both Zeeman sextets is close to 3:2:1:1:2:3. This means that the magnetic domains in the sample cooled in both the ZFC and FC regimes are randomly oriented, which is expected for the TMS measurements performed on polycrystalline or powdered samples in the absence of an external magnetic field [16].

Let us now consider the expected response from the hypothetical cluster-glass formation. In the FC regime, the sample was cooled down to 4.3 K in magnetic field applied parallel to the γ -ray beam propagation. Again, in this configuration, the observed line intensity ratio for the sextet should be 3:0:1:1:0:3 [16]. If we assume that below T_{rmc} a ferromagnetic cluster-glass state really forms, then in the FC regime the magnetic moments should be frozen at 4.3 K and aligned parallel to the applied field. And then also the line intensity ratio for the magnetic sextet in the FC spectrum should be close to the 3:0:1:1:0:3 mentioned above, not 3:2:1:1:2:3 as inferred from the experimental spectrum. This means that the magnetic moments are already magnetically ordered within randomly oriented domains, so the external magnetic field does not affect their local arrangement. Thus, it can be concluded that no behavior indicating the freezing of the magnetic moments (as should be the case in cluster glass or spin glass) is observed, and the compound UFe_{0.40}Ge₂ orders purely ferromagnetically at $T_{\rm C}$.

4. Conclusions

We have measured the transmission Mössbauer spectra on a polycrystalline, ⁵⁷Fe-isotope-enriched sample of UFe_{0.40}Ge₂, which has been positively verified for the crystal structure and 384

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magnetic properties described previously for UFe0.39Ge2. Anal-373 319 ysis of the collected TMS spectra have confirmed that the com-374 320 pound orders ferromagnetically below $T_{\rm C}$ of about 37 K, while 321 they have shown no sign of the previously postulated forma-377 322 tion of magnetic glassy states. This suggests that the crystallo-378 323 graphic disorder present in UFe_{1-x}Ge₂ mainly affects the trans-³⁷⁹ 324 port properties of the system rather than its magnetic ground³⁸⁰₃₈₁ 325 state. Further experiments, preferably performed on a single-382 crystalline sample, are needed to confirm this conclusion. 383 327

328 CRediT authorship contribution statement

Adam Pikul: Conceptualization, Formal analysis, Writing –³⁸⁸ Original Draft, Review & Editing; **Rafał Idczak:** Investigation,³⁸⁹ Formal analysis, Writing – Original Draft, Review & Editing;₃₉₁ **Piotr Sobota:** Investigation; **Wojciech Nowak:** Investigation;³⁹² **Vinh Hung Tran:** Writing – Review & Editing; **Mathieu Pas**-³³³ **turel:** Formal analysis; Writing – Original Draft, Review &³⁹⁴ Editing.

336 Declaration of Competing Interest

The authors declare that they have no known competing fi-401 nancial interests or personal relationships that could have ap-402 peared to influence the work reported in this paper.

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- UFe_{0.40}Ge₂ has been confirmed to order ferromagneticallybelow T_c = 37 K.
- The compound has been studied by Mössbauer spectroscopy.
- The experiments have confirmed that the ferromagnetism in UFe_{0.40}Ge₂ is purely long-range in nature.
- The hypothesis of a ferromagnetic cluster glass below T_c has been discarded.

Journal Pre-proof

Adam Pikul: Conceptualization, Formal analysis, Writing -- Original Draft, Review & Editing; Rafał Idczak: Investigation, Formal analysis, Writing -- Original Draft, Review & Editing; Piotr Sobota: Investigation;

Wojciech Nowak: Investigation;

Vinh Hung Tran: Writing -- Review & Editing;

Mathieu Pasturel: Formal analysis; Writing -- Original Draft, Review & Editing.

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