================== **MAGNETISM** ==================

Specific Features of Magnetic Ordering in the SmFeGe$_2$O$_7$ Compound

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Abstract—The results of the experimental investigation of the magnetic properties of the SmFeGe$_2$O$_7$ compound have been presented. It has been found that the temperature dependence of the susceptibility exhibits two features that coincide with the anomalies in the temperature dependence of the specific heat and indicate magnetic phase transitions in SmFeGe$_2$O$_7$. The external magnetic field induces a magnetic transition, the critical field of which depends on the temperature.

1. INTRODUCTION

It is known that RFeGe$_2$O$_7$ compounds are characterized by a monoclinic crystal structure, whereas their space symmetry group depends on the radius of the rare-earth cation R. The following groups of compounds can be separated according to the type of the space group: first, the compounds with rare-earth ions of the cerium subgroup (La–Eu) and Gd, which have the space group $P2_1/c$ [1, 2]; second, the compounds with rare-earth ions of the yttrium subgroup (Gd–Lu), except Gd, with the space group $P2_1/m$ [3, 4]. The unit cell of the crystals of both groups of compounds contains four formula units ($Z = 4$).

The magnetic properties of the system RFeGe$_2$O$_7$ were studied in [1–4]. According to the results of these studies, the temperature dependences of the susceptibility of the samples of the first group including SmFeGe$_2$O$_7$ exhibit one anomaly associated with simultaneous antiferromagnetic ordering of Fe^{3+} and R^{3+} ions [1, 2]. The temperature dependences of the susceptibility of the samples of the second group exhibit two anomalies, the characteristic temperatures T_{N1} and T_{N2} of which are associated with the ordering of Fe^{3+} and R^{3+} ions, respectively [3, 4]. With the use of Mössbauer measurements, the Néel temperature T_N of the SmFeGe$_2$O$_7$ sample was found to be $T_N = 6.5$ K [1]. Kazei et al. [3] also reported on the discovery of metamagnetic transitions in RFeGe$_2$O$_7$ (R = Tb–Tm) compounds.

In this work, we have presented the results of studying specific features of the magnetic ordering of SmFeGe$_2$O$_7$.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

The SmFeGe$_2$O$_7$ samples were synthesized by the solid-state reaction from a mixture of the oxides Fe$_2$O$_3$, Sm$_2$O$_3$, and GeO$_2$ taken in accordance with stoichiometry of the compound. The samples were molded under pressure to form tablets with a diameter of 10 mm and a thickness of 1.5–2.0 mm, annealed in air in two stages at a temperature of 1100°C with intermediate wet grinding in an alcoholic medium, and again molded. The chemical and phase compositions of the samples were controlled by X-ray diffraction analysis.

The X-ray diffraction pattern of a polycrystalline SmFeGe$_2$O$_7$ sample at room temperature was measured on a D8-ADVANCE diffractometer (CuK_α radiation) with the use of a VALTEC linear detector. The angle 2θ was scanned with steps of 0.016° and an exposure of 0.6 s per step.

The magnetic measurements were performed on a MPMS-XL magnetometer at the Siberian Federal University in the temperature range of 2–300 K in the magnetic fields up to 5 T.

3. EXPERIMENTAL RESULTS

The X-ray diffraction investigation of SmFeGe$_2$O$_7$ showed that its crystal structure is isostructural to the structure of PrFeGe$_2$O$_7$, which was determined earlier in [2]. Therefore, the coordinates of the atoms of the latter compound were taken as the starting model for the Rietveld refinement with the TOPAS 4.2 software [5]. The refinement gave low discrepancy factors (Fig. 1, Table 1). The X-ray diffraction pattern exhib-
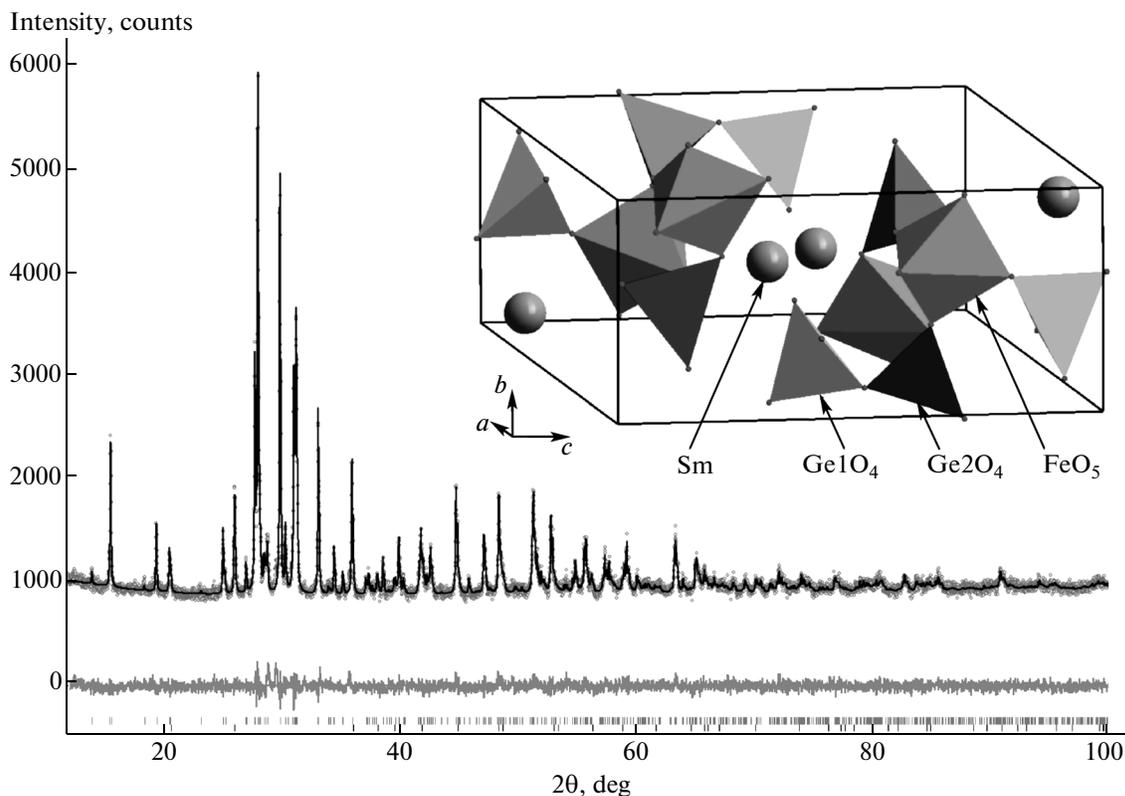
Fig. 1. Difference X-ray diffraction pattern of polycrystalline $SmFeGe_2O_7$. The inset shows the structure of the compound.

ited reflections corresponding to the GeO_2 impurity. The refinement showed that the weight fraction of the impurity was 3.7(2)%. The coordinates of the atoms of $SmFeGe_2O_7$ and the selected bond lengths are listed in Tables 2 and 3, respectively. The dependence of the unit cell volume on the ionic radius of the element R^{3+}

in the compounds $RFeGe_2O_7$ [1, 2, 6] is linear, and the data for the compound under investigation also fall in this linear dependence (Fig. 2).

Figure 3 shows the temperature dependence of the susceptibility of $SmFeGe_2O_7$ measured in the mag-
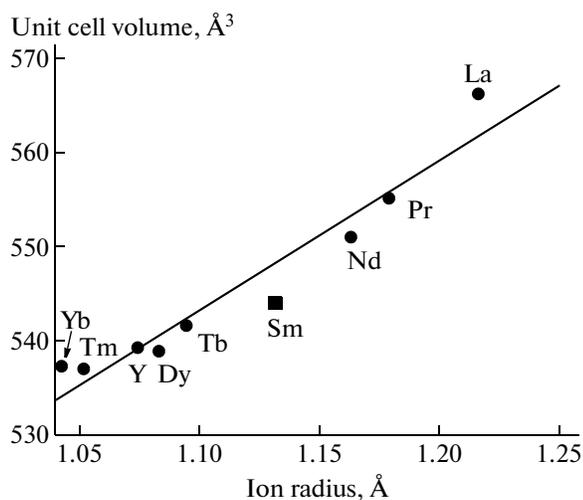
Fig. 2. Unit cell volume versus the ionic radius of the element R^{3+} in the compounds $RFeGe_2O_7$. The compound of interest with $R^{3+} = Sm^{3+}$ is shown by the square.
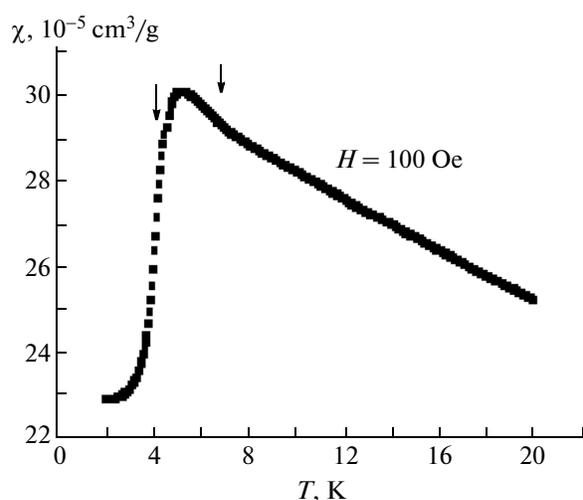
Fig. 3. Temperature dependence of the susceptibility of $SmFeGe_2O_7$ in a magnetic field of 100 Oe.

Table 1. Data collection and structure refinement parameters of SmFeGe$_2$O$_7$ (space group $P2_1c$)

Parameter	Value
a, Å	7.1752(2)
b, Å	6.6080(2)
c, Å	12.8912(3)
β, deg	117.110(1)
V, Å^3	544.07(2)
Angle range 2θ, deg	5−100
Number of Bragg reflections	573
Number of refined parameters	67
R_B, %	0.82
R_{wp}, %	3.65
R_p, %	2.85
R_{exp}, %	3.11
χ^2	1.17

Designations: a, b, c, and β are the unit cell parameters; V is the unit cell volume; R_B, R_{wp}, R_p, and R_{exp} are the integrated, weighted profile, profile, and expected discrepancy factors, respectively; and χ^2 is the goodness-of-fit.

Table 2. Coordinates of atoms and isotopic thermal parameters (B_{iso}) of the SmFeGe$_2$O$_7$ structure

Atom	x	y	z	B_{iso}
Sm	0.7621(7)	0.1496(6)	0.0243(3)	0.4(2)
Fe	0.7958(13)	0.4017(16)	0.2698(7)	0.4(3)
Ge1	0.7822(12)	0.6521(12)	0.0373(7)	0.5(3)
Ge2	0.2951(11)	0.4096(13)	0.2214(6)	0.5(3)
O1	0.575(5)	0.833(6)	0.016(2)	0.6(4)
O2	0.766(4)	0.127(7)	0.219(2)	0.6(4)
O3	0.554(5)	0.385(6)	0.302(2)	0.6(4)
O4	0.007(5)	0.354(6)	0.423(2)	0.6(4)
O5	0.728(6)	−0.005(4)	0.415(3)	0.6(4)
O6	0.797(6)	0.483(5)	0.131(3)	0.6(4)
O7	0.154(5)	0.166(5)	0.189(3)	0.6(4)

netic field $H = 100$ Oe. The following features can be seen: a kink at the temperature $T_1 = 6.5$ K and a steepest variation of the susceptibility near the temperature $T_2 = 4.3$ K.

The specific heat of SmFeGe$_2$O$_7$ in the temperature range from 2 K to room temperature was measured in our previous work [7]. The thermal properties were studied on a Quantum Design PPMS 6000 setup in the temperature range of 2.0−300 K. According to the results of thermal measurements presented in Fig. 4, the temperature dependence of the specific heat C_p exhibits two anomalies at the temperatures T_1 = 6.5 K and T_2 = 4.3 K, which are typical for thermodynamic phase transitions.

According to [1], the compound SmFeGe$_2$O$_7$, which has the crystal structure of the space group $P2_1/c$ in the paramagnetic state, preserves this structure under cooling of the sample to the temperature $T = 5$ K. Thus, the phase transitions discovered in SmFeGe$_2$O$_7$ at the temperatures $T_1 = 6.5$ K and $T_2 = 4.3$ K (Fig. 4) presumably occur without a change in the symmetry of the crystal lattice.

It is noteworthy that the temperatures of the anomalies in the temperature dependence of the specific heat coincide with the temperatures of the anomalies seen in the temperature dependence of the susceptibil-

Fig. 4. Temperature dependence of the specific heat of SmFeGe$_2$O$_7$ [7].
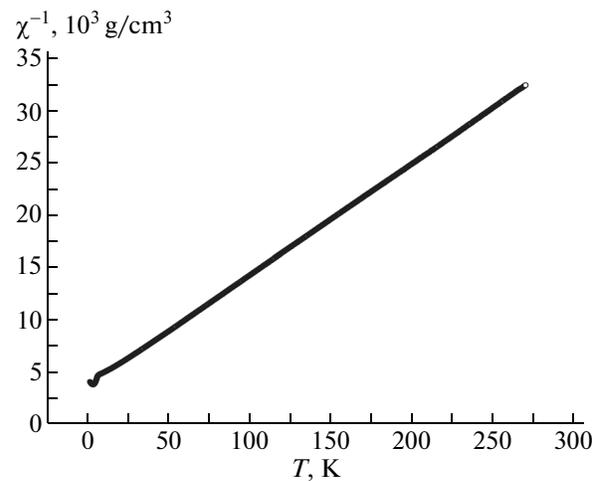
Fig. 5. Temperature dependence of the inverse susceptibility of SmFeGe$_2$O$_7$ in a field of 30000 Oe (the asymptotic Néel temperature is $\theta = -38.6$ K).
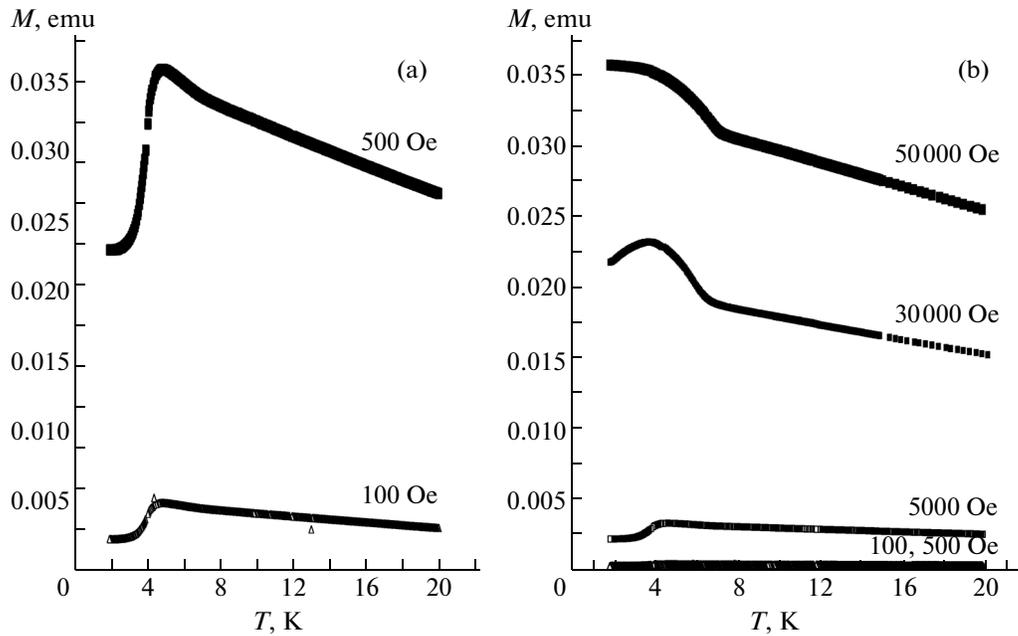
Fig. 6. Evolution of the temperature dependence of the magnetic moment of $SmFeGe_2O_7$ in the magnetic fields (a) $H = 100$, 500 Oe and (b) $H = 100$, 500, 5000, 30000, 50000 Oe. The sample weight is $m = 0.030$ g.

ity (Figs. 3, 4). Taking into account the results of determining the Néel temperature $T_N = 6.5$ K by the Young's method [1] and the results of magnetic measurements shown in Fig. 3, we conclude that the phase transition at the temperature $T_1 = T_N = 6.5$ K corresponds to magnetic ordering of the sample (the order–disorder phase transition). The anomalies in the specific heat and magnetization at the temperature $T_2 = $

Table 3. Selected bond lengths (Å) in the $SmFeGe_2O_7$ structure

Bond length	Value	Bond length	Value
Sm–O1[(i)]	2.46(3)	Fe–O4[(vi)]	1.89(3)
Sm–O1[(ii)]	2.23(2)	Fe–O6	1.87(4)
Sm–O2	2.50(2)	Fe–O7[(vii)]	1.81(4)
Sm–O3[(iii)]	2.57(3)	Ge1–O1	1.83(3)
Sm–O4[(iv)]	2.44(4)	Ge1–O4[(vii)]	1.90(3)
Sm–O4[(v)]	2.63(2)	Ge1–O5[(iii)]	1.74(3)
Sm–O5[(iii)]	2.69(3)	Ge1–O6	1.61(3)
Sm–O6	2.55(3)	Ge2–O2[(vii)]	1.78(4)
Sm–O7[(vi)]	2.64(3)	Ge2–O3	1.67(2)
Fe–O2	1.91(4)	Ge2–O5[(vii)]	1.78(3)
Fe–O3	1.96(2)	Ge2–O7	1.85(3)

The symmetry elements are: (i) $x, y - 1, z$; (ii) $-x + 1, -y + 1, -z$; (iii) $x, -y + 1/2, z - 1/2$; (iv) $-x + 1, y - 1/2, -z + 1/2$; (v) $x + 1, -y + 1/2, z - 1/2$; (vi) $x + 1, y, z$; (vii) $-x + 1, y + 1/2, -z + 1/2$.

4.3 K seemingly indicate the presence of a transition between different types of magnetic structures, which occurs in a number of magnets with different types of magnetic ordering (see, e.g., [8–10]).

Since the only direct method of determining the magnetic structure of compounds is magnetic neutron diffraction analysis, we attempted experiments on neutron scattering with the wavelength $\lambda = 2.4576$ Å in the temperature range to 1.6 K on a DMC diffractometer at the Paul Scherrer Institute (Villigen, Switzerland) to investigate the nature of the phase transitions in $SmFeGe_2O_7$ under the temperature variation. However, investigation of $SmFeGe_2O_7$ by elastic neutron scattering unfortunately did not give results owing to a large neutron absorption by samarium [7].

Figure 5 presents the temperature dependence of the inverse magnetic susceptibility $\chi^{-1}(T)$ in a magnetic field of 3 T. As follows from the experimental data, the behavior of $\chi^{-1}(T)$ in the high-temperature region ($T > 20$ K) is described by the Curie–Weiss law.

The asymptotic Néel temperature found as the crossing point of the T axis with the asymptote to the curve $\chi^{-1}(T)$ in the high-temperature region is $\theta = -38.6$ K and witnesses predominantly antiferromagnetic interaction in the magnetic subsystem of $SmFeGe_2O_7$ formed by the magnetic ions Sm^{3+} and Fe^{3+}.

The behavior of the $SmFeGe_2O_7$ sample in the magnetic field was also studied. Figure 6 presents the temperature dependences of the magnetic moment in the magnetic fields $H = 100$ and 500 Oe (Fig. 6a) and

Fig. 7. Isothermal magnetization curves of SmFeGe$_2$O$_7$. The sample weight is $m = 0.030$ g.
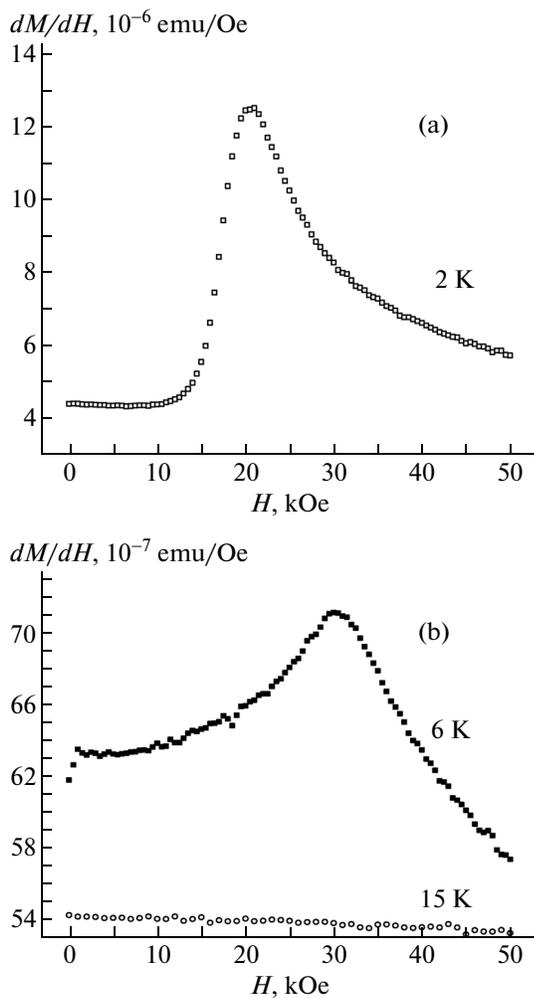
Fig. 8. Isothermal differential susceptibility of SmFeGe$_2$O$_7$ in static magnetic fields at T = (a) 2, (b) 6 and 15 K.

$H = 100$, 500, 5000, 30000, and 50000 Oe (Fig. 6b), which illustrate its evolution under the external action. The field dependences of the magnetic moment of SmFeGe$_2$O$_7$ at temperatures of 2, 6, and 15 K are shown in Fig. 7. The magnetic moment depends linearly on the magnetic field at the temperatures $T \geq$ 15 K. The linearity is violated at low temperatures. A sharp increase in the magnetic moment occurs at the temperature $T = 2$ K in a magnetic field of ~20000 Oe. Figure 8 shows isotherms of the differential susceptibility of SmFeGe$_2$O$_7$ in static magnetic fields. As is seen, the maximum of the differential susceptibility at the temperatures $T = 2$ and 6 K corresponds to the magnetic fields $H_c = 20300$ and 30000 Oe, respectively. Thus, as follows from the experiment, the critical field H_c of the transformation of the magnetic structure of SmFeGe$_2$O$_7$ depends on temperature. In the paramagnetic region, the anomaly of the susceptibility disappears (Fig. 8b, $T = 15$ K).

Figure 9 presents the temperature dependence of the magnetic moment of SmFeGe$_2$O$_7$ measured in a magnetic field of 0.05 T under different conditions of sample cooling: zero-field cooling (ZFC) and cooling in a magnetic field of 0.05 T (FC). There is a noticeable hysteresis in the low-temperature region, which is typical for the first-order phase transition.

Thus, investigation of the specificity of magnetization of SmFeGe$_2$O$_7$ also revealed the presence of a phase transition induced by the magnetic field.
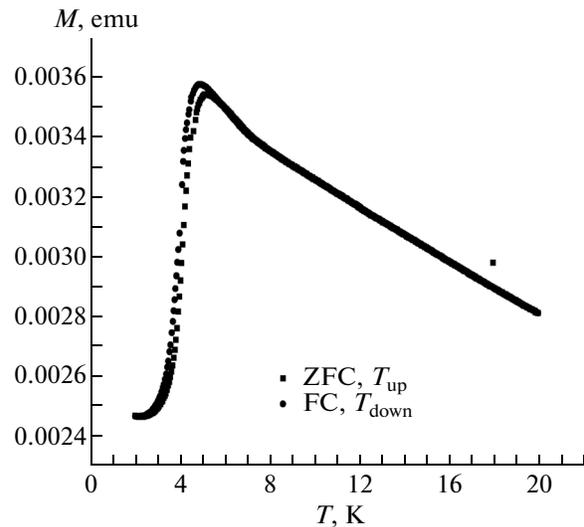
Fig. 9. Temperature dependence of the magnetic moment of SmFeGe$_2$O$_7$ in a magnetic field of 0.05 T. T_{up} and T_{down} correspond to the directions of the temperature variation during the measurements. ZFC is the cooling of the sample without a magnetic field, and FC is the cooling in a magnetic field of 0.05 T.

4. CONCLUSIONS

The specific features of the magnetic ordering of the $SmFeGe_2O_7$ compound, the magnetic structure of which is characterized by the antiferromagnetic exchange interaction, have been investigated. Two features in the temperature dependence of the susceptibility and the corresponding anomalies in the temperature dependence of specific heat have been discovered at the temperatures $T_1 = 6.5$ K and $T_2 = 4.3$ K. Based on the whole set of the experimental data, we can conclude that the following magnetic phase transitions occur in $SmFeGe_2O_7$: (i) a magnetic phase transition from a high-temperature paramagnetic state to a low-temperature magnetically ordered state — the order−disorder transition — at the temperature $T = 6.5$ K; (ii) a change in the magnetically ordered state, i.e., the order−disorder transition, at the temperature $T = 6.5$ K; (ii) a change in the magnetically ordered state, i.e., the order−disorder transition presumably corresponding to a change in the spin orientation in the sample, at the temperature $T_2 = 4.3$ K; and (iii) a magnetic-field induced magnetic transition (the critical magnetic field at $T = 2$ K is $H_c = 20\,300$ Oe).

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