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Abstract

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Keywords

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Comments

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INTRODUCTION

As early as 1936, Néel¹ predicted that in a uniaxial anisotropic antiferromagnet where magnetic moments are coupled with the crystal lattice, a first order spin-flop transition should occur at a certain critical magnetic field, H_{sf} , when the magnetic field is applied along the antiferromagnetic (AFM) coupling direction, i.e., along the AFM easy axis. As a result of the spin-flop transition, the directions of spin moments of the two sublattices abruptly change from their antiparallel configuration in which the moments are collinear with the external field to a new spin-flop configuration in which the moments remain antiparallel but become perpendicular or nearly perpendicular to the magnetic field vector. The spin-flop transition was later experimentally observed in $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$.²⁻⁵ The phenomenon perhaps is best manifested in several transition metal systems such as MnF_2 (Refs. 6–8) and FeF_2 ,⁹ and current research activities on spin-flop transitions are extended toward many low dimensional systems.¹⁰⁻¹² As in the past, spin-flop transformations remain an interesting subject for both theoretical and experimental research.

Gd_5Ge_4 is one of the two parent compounds of the fundamentally important and potentially useful pseudobinary magnetocaloric alloys $\text{Gd}_5(\text{Si}_x\text{Ge}_{1-x})_4$.¹³⁻¹⁵ This binary gadolinium germanide has been broadly studied because of its intriguing physical properties that are intimately related to a peculiar low dimensional crystallography. The distinctly layered crystal structure that the system adopts in the paramagnetic state can be described as a stacking of weakly interacting identical slabs that are infinite in the ac plane but are less than 1 nm thick along the b axis.¹⁶⁻¹⁸ In the original study by Holtzberg *et al.*,¹⁹ the authors suggested that the ground state of Gd_5Ge_4 is AFM, although the positive Weiss temperature determined from the Curie-Weiss behavior points to a ferromagnetic ground state. Recently, this apparently contradic-

tory result has been explained by the fact that the Gd moments are coupled ferromagnetically within the slabs, while the Gd moments between the slabs are coupled antiferromagnetically along the c axis.²⁰⁻²⁵ This AFM configuration was identified both by macroscopic dc magnetization measurements^{26,27} and microscopic x-ray resonant magnetic scattering experiments,²⁸ thus showing that zero-field-cooled Gd_5Ge_4 is a uniaxial antiferromagnet. In the AFM state, the compound preserves the same crystal structure as in the paramagnetic state.^{29,30} However, when Gd_5Ge_4 is magnetized at low temperatures by a magnetic field exceeding 12 kOe,^{20-24,26,27,29,30} or is exposed to a hydrostatic pressure in excess of 10 kbar,³¹ it becomes ferromagnetic (FM). The FM ordering is accompanied by a simultaneous martensitic-like change of its crystal structure. The latter occurs by shearing of the neighboring slabs along a in opposite directions,^{29,30} hence creating a new layered crystal structure where the slabs themselves remain unchanged but the inter-slab exchange interactions become much stronger.²⁵ These magnetic field and pressure induced transformations are unusually sharp, which is related to the existence of a nonergodic kinetically retarded magnetic glassy state at low temperature.³²

Unlike the interplay between the paramagnetic (PM), AFM, and FM states of Gd_5Ge_4 , which is relatively well studied,²⁰⁻³² the nature of the spin-flop transition, which was first reported by Levin *et al.*,²⁶ is much less understood. This transition occurs in the AFM state only when the magnetic field is applied along the c axis. Interestingly, the spin-flop transformation remains completely unaffected by the kinetic retardation that affects the AFM-FM transition.^{27,33} The magnetic phase diagram with the magnetic field vector parallel to the c axis (Fig. 1) shows that the PM phase orders into the normal AFM phase below $T_N=128$ K in low magnetic fields, but it is converted into a spin-flop AFM phase in fields higher than ~ 8 kOe. Levin *et al.*²⁶ suggested that in the

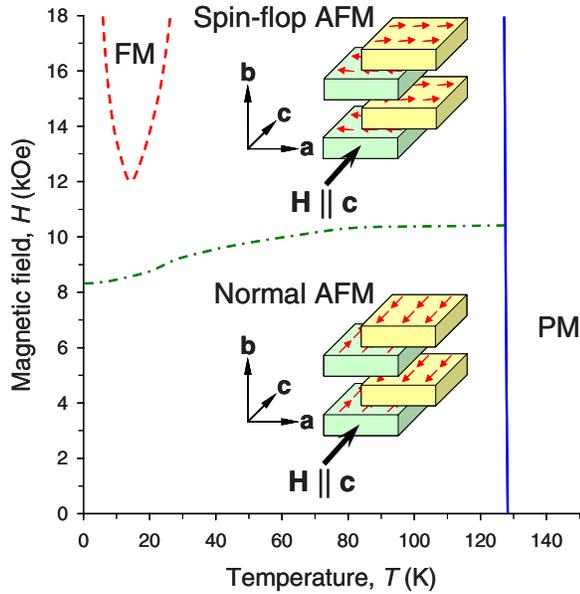


FIG. 1. (Color online) The low temperature, low magnetic field corner of the magnetic phase diagram of Gd_5Ge_4 taken from Ref. 27. This diagram is for the initial magnetization of the zero-field-cooled material with the magnetic field vector parallel to the c axis. The microscopic magnetic structure of the normal AFM phase is illustrated schematically taking into account both the macroscopic and microscopic data of Refs. 26 and 28. The schematic of the spin-flop AFM phase is shown following an assumption of Ref. 26.

spin-flop state, the AFM coupling axis coincides with the a axis, which was logical considering that the moments then have to rotate in the planes of the slabs rather than out of the planes of the slabs. The AFM coupling axis is perpendicular to the field vector in the spin-flop phase, but the magnetization of each sublattice is slightly tilted away from the spin coupling axis (a) due to the application of the magnetic field. To date, this model has not been verified experimentally.

In a uniaxial antiferromagnet, where the anisotropy energy is usually small compared to the generally isotropic exchange energy, the first order spin-flop transition can be observed only over a small range of tilt angles when the field vector remains perpendicular to the hard axis but is tilted away from the AFM easy axis toward the next easiest axis (i.e., toward the spin-flop axis).^{4,5,7,34} Here, we investigate angular dependence of the spin-flop transition in Gd_5Ge_4 . In addition, owing to the distinctly layered crystal structure of Gd_5Ge_4 , a different behavior is expected when the magnetic field is tilted away from the c axis (AFM easy axis) toward the b axis when compared to the magnetic field tilted away from the c axis toward the a axis. These experiments should verify an earlier prediction (by Levin *et al.*²⁶) that the microscopic magnetic structure of the spin-flop AFM phase is the result of rotating the AFM coupled moments in the ac plane, as shown schematically in Fig. 1, rather than in the ab plane.

EXPERIMENTAL DETAILS

A cubelike single crystal of Gd_5Ge_4 ($0.9 \times 1.0 \times 1.0 \text{ mm}^3$) with its faces normal to the principal crystallo-

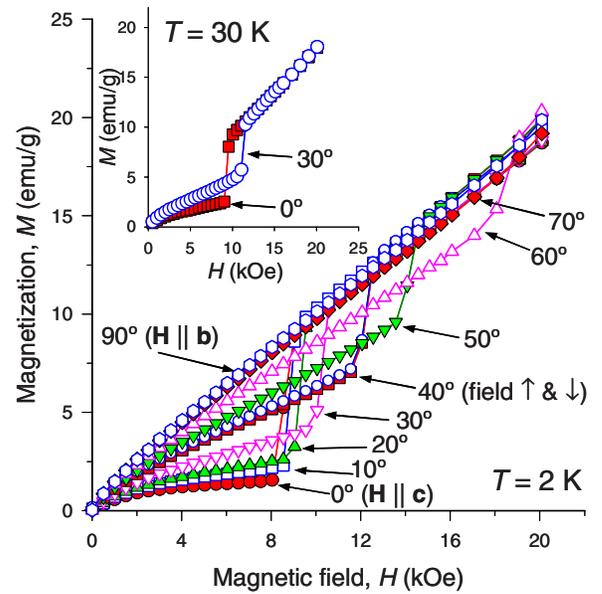


FIG. 2. (Color online) The magnetization isotherms of a Gd_5Ge_4 single crystal measured at $T=2 \text{ K}$ with the magnetic field vector gradually tilted away from the c axis toward the b axis (the cb plane isotherms). The inset shows two isotherms measured at $T=30 \text{ K}$ with $\mathbf{H} \parallel \mathbf{c}$ and with $\mathbf{H} \angle \mathbf{c} = 30^\circ$ in the cb plane.

graphic directions was extracted from a crystal grown using the tri-arc pulling technique.³⁵ Details about the basic characterization, orientation, and preparation of the specimen are given elsewhere.²⁷ The magnetization measurements were performed using a superconducting quantum interference device magnetometer, MPMS-XL manufactured by Quantum Design, Inc. Before each measurement, the sample was zero-field cooled from the paramagnetic state at 300 K to the desired temperature. The misalignment between the crystallographic axes and directions of magnetic field vectors was estimated to be less than 5° .

EXPERIMENTAL RESULTS

Figures 2 and 3 show the field dependencies of the magnetization of a Gd_5Ge_4 single crystal measured at 2 K when the field vector was gradually tilted away from the c axis toward the b axis (the cb plane isotherms) and away from the c axis toward the a axis (the ca plane isotherms), respectively. All of the cb plane $M(H)$ curves (Fig. 2) exhibit nonlinearity in low fields (less than $\sim 4 \text{ kOe}$), revealing the presence of a small amount of the FM Gd_5Ge_4 phase that precipitates in the AFM matrix during zero-field cooling.³⁶ Around 20 kOe, the $M(H)$ curves show signs of an upturn, indicating the onset of the well-known and well-characterized field induced AFM \rightarrow FM phase transformation. For $\theta=0$, i.e., when the magnetic field vector is applied along the c axis, a nearly discontinuous steplike transition is observed at $H_{\text{sf}} \cong 8.3 \text{ kOe}$, which is a signature of a first order spin-flop transition²⁶ and whose temperature behavior was established in an earlier report²⁷ (also see Fig. 1). With an increase of the tilt angle θ between \mathbf{H} and \mathbf{c} , the magne-

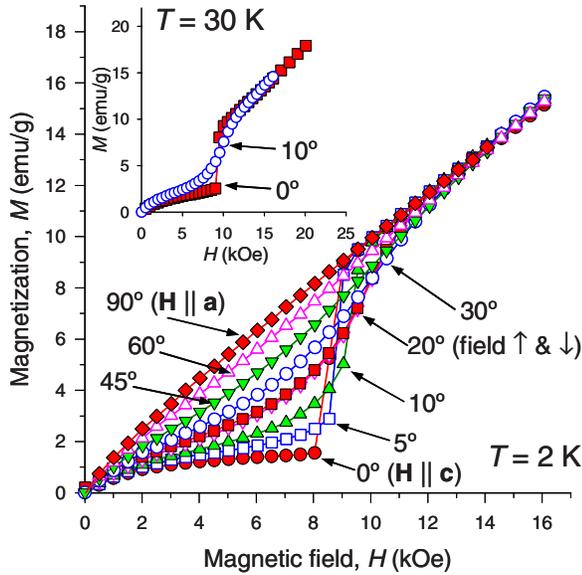


FIG. 3. (Color online) The magnetization isotherms of Gd_5Ge_4 single crystal measured at $T=2$ K with the magnetic field vector gradually tilted away from the c axis toward the a axis (the ca plane isotherms). The inset shows two isotherms measured at $T=30$ K with $\mathbf{H}\parallel c$ and with $\mathbf{H}\angle c=10^\circ$ in the ca plane.

tization in the AFM state is increased, but in the spin-flop state, the magnetization remains nearly equal to that of $\theta=0$. This leads to a gradual reduction of the magnetization discontinuity at H_{sf} . However, judging from the sharpness of the jump of the magnetization, the spin-flop transition maintains its first order character. Regardless of the angle θ , the H -decreasing $M(H)$ curves nearly follow the H -increasing ones, showing the nearly complete reversibility of the spin-flop transition (see, for example, the two curves shown for $\theta=40^\circ$). We also note that the first order reversible spin-flop transition is observed at higher temperatures, as illustrated in the inset of Fig. 2 for $T=30$ K.

At 2 K, H_{sf} increases nonlinearly with θ as long as the field vector is confined in the cb plane, as shown in Fig. 4(a). Extrapolation of the H_{sf} vs θ curve to 25 kOe, which is the critical field for the AFM \rightarrow FM transition with the field vector parallel to the b axis,²⁷ leads to $\theta\cong 65^\circ$. This suggests that the spin-flop transition should disappear when $\theta>65^\circ$, which is consistent with the results shown in Fig. 2. The components of the vector \mathbf{H}_{sf} are plotted in Fig. 4(b), indicating a weak and nearly linear (from 8.3 kOe at $\theta\cong 0$ to 9.8 kOe at $\theta\cong 60^\circ$) dependence of its projection along the c axis.

When the magnetic field vector is tilted away from the c axis toward the a axis, i.e., when it is confined in the ca plane and therefore is coplanar with the slabs, a sharp spin-flop transition is only observed within a narrow range of the tilt angles, i.e., $0<\theta<20^\circ$ (Fig. 3). Upon further increasing the tilt angle, the magnetization begins to exhibit a gradual and smooth transition from the normal AFM state to the spin-flop AFM state, i.e., the spin-flop transition becomes continuous and occurs over a relatively wide range of fields. Nearly the same behavior is observed at 30 K (see the inset

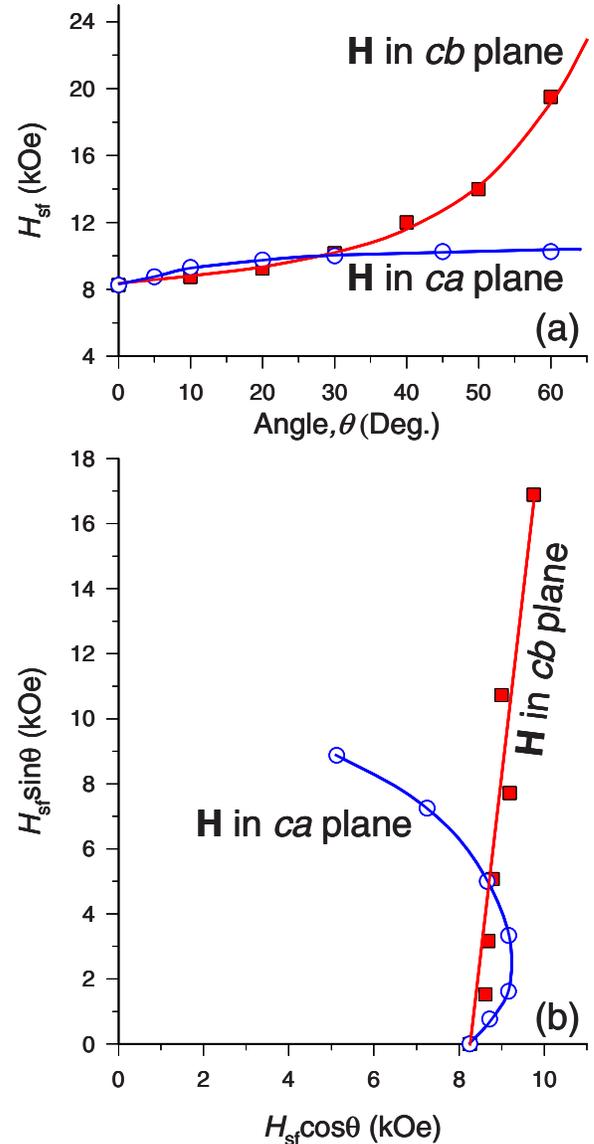


FIG. 4. (Color online) (a) Angle dependencies of H_{sf} in Gd_5Ge_4 at $T=2$ K and (b) projections of the H_{sf} vector along the c and b axes and the c and a axes.

of Fig. 3). All of these are consistent with a changeover from a first order transformation at low θ to a second order transformation at large values of θ , which is quite different from the behavior observed when the field was applied in the cb plane. When θ exceeds 60° , the spin-flop transition is no longer recognizable, and all magnetization curves merge into that of $\theta=90^\circ$ (i.e., when $\mathbf{H}\parallel a$) (see Fig. 3). Obviously, the second order spin-flop transition is also reversible, as shown in Fig. 3 for $\theta\cong 20^\circ$.

For a continuous spin-flop transition, we define the field where the dM/dH exhibits a maximum as H_{sf} . A plot of H_{sf} vs θ is shown in Fig. 4(a). With increasing θ , the value of H_{sf} initially increases slowly and then nearly saturates around 10 kOe [Fig. 4(a)], never approaching the critical field $H_c\cong 31$ kOe (Ref. 27) for the AFM \rightarrow FM transition with the magnetic field vector parallel to a . The projection of the vector H_{sf} along the c axis increases first for $0<\theta<20^\circ$ and

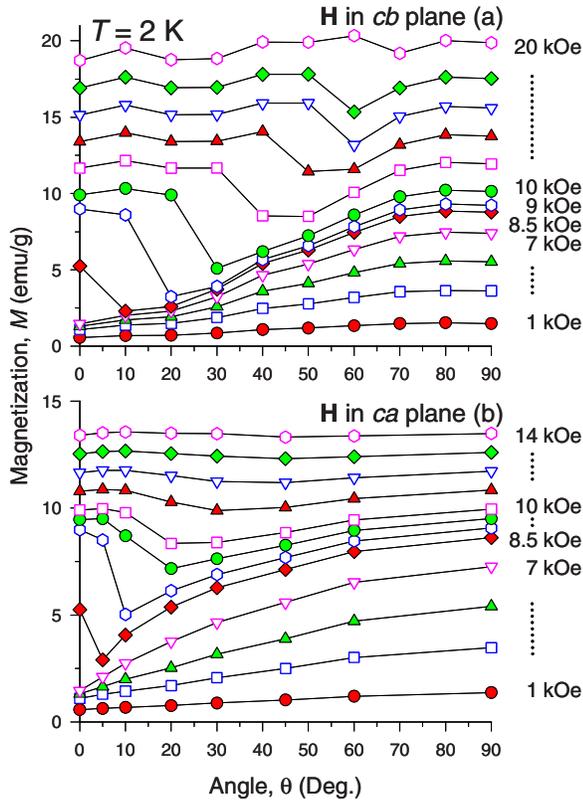


FIG. 5. (Color online) Angle dependencies of the magnetization of Gd_5Ge_4 at selected magnetic fields extracted from $M(H)$ measurements in the cb plane (a) and in the ca plane (b).

then decreases rapidly when $\theta > 20^\circ$ [see Fig. 4(b)], which is quite different from the cb behavior, also shown in Fig. 4(b).

A clear picture of the angular dependencies of the spin-flop transition may also be seen in Fig. 5, where we plot the magnetization versus θ extracted at selected fields from the data shown in Figs. 2 and 3. In Fig. 5(a) (the cb curves), there are no anomalies for $H < 8.3$ kOe, i.e., as long as the magnetic field remains below the critical spin-flop field for $\theta \equiv 0$. When the magnetic field increases to 8.5 kOe and higher, a plateau in magnetization develops starting from $\theta \equiv 0$ and the length of the plateau increases (in units θ) as the field increases [Fig. 5(a)]. The plateau is followed by a spin-flop transition, which is characterized by a sharp drop of the magnetization. The first order spin-flop transition is recognizable up to $\theta \approx 65^\circ$. However, in the ca curves [Fig. 5(b)], the sharp spin-flop transition is observed only within $0 < \theta < 20^\circ$.

Based on the experimental results presented above, we conclude that the nature of the spin-flop transition in Gd_5Ge_4 is strongly dependent on the orientational relationships between the direction of the magnetic field vector and the plane of the slabs. As long as the magnetic field vector is confined within the cb plane (i.e., it is rotated in the plane normal to the slabs), the spin-flop transition always remains first order. However, in the ca plane (the field vector is coplanar with the slabs), the spin-flop transition is only first order within a small range of angles between the magnetic field vector and the AFM easy (c) axis ($0 < \theta < 20^\circ$). The data shown in Fig.

5 compare well to that found in $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, which also has an orthorhombic structure.⁴ In $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$, the a axis is the AFM easy axis, the b axis is the next easiest axis (spin-flop axis), and the c axis is the hard axis. The magnetization of $\text{CuCl}_2 \cdot 2\text{H}_2\text{O}$ measured as a function of θ with the magnetic field vector confined in the ac plane exhibits a constant value at low θ (when $\mathbf{H} \parallel \mathbf{a}$) followed by a sharp drop, whereas both of these features are absent in the ab plane measurements.³ Hence, our results for Gd_5Ge_4 are consistent with the AFM easy axis being the c axis, the next easiest axis (the spin-flop axis) is the a axis, and the hard axis is the b axis. This means that with the occurrence of the spin-flop transition, the Gd moments that were coupled antiferromagnetically along the c axis are rotated within the plane of the slab and become nearly collinear with the a axis. In the cb plane measurements, the spin rotation is discontinuous because the spin-flop axis is always perpendicular to the magnetic field vector, whereas in the ca plane measurements, the spin rotation is continuous because the spin-flop axis is no longer perpendicular to the field vector.

Even though data about microscopic magnetism of $R_5\text{Si}_x\text{Ge}_{4-x}$ compounds with heavy lanthanides are limited to only a few representatives with $R = \text{Gd}$, Tb , and Er ,^{28,37-40} the spin-flop AFM configuration of Gd_5Ge_4 with the magnetic moments coplanar with the slabs is quite reasonable. In all known cases, there is a clear tendency toward ferromagnetism of the individual slabs. Furthermore, the moments are coplanar or nearly coplanar with the slabs in compounds formed by heavy lanthanides with small orbital contribution to the total magnetic moment (Gd and Tb), while the moments become normal to the slabs in Er, where the orbital contribution dominates. We believe that this is related to a gradual reduction of the strength of the isotropic exchange interactions and to an increasing anisotropic contribution from the magnetoelastic coupling that arises from the competition between the anisotropy of the layered crystal structure and single ion anisotropy. Thus, the prediction by Levin *et al.*,²⁶ which was made without direct evidence, was correct, and the results presented above do confirm that the a axis is indeed the spin-flop axis of Gd_5Ge_4 .

It is worth noting that the theoretical treatment using the molecular field approximation³⁴ of the phase transition between a normal AFM phase and a spin-flop AFM phase in a uniaxial antiferromagnet shows that the range of angles over which a first order spin-flop transition may be observed is quite small because the anisotropy energy is usually small compared to the isotropic exchange energy. This conclusion is in agreement with some other uniaxial antiferromagnets, where the maximum tilt angle at which a first order spin-flop transition between the AFM easy axis and the spin-flop axis can be observed (θ_{max}) has a magnitude of less than 1° .^{4,5,7,34} This is indeed much smaller than the value of $\theta_{\text{max}} = 20^\circ \pm 5^\circ$ seen in Gd_5Ge_4 , especially considering the possible misalignment of the field vector with respect to the crystallographic direction(s). As pointed out by Rohrer and Thomas,³⁴ the maximum tilt angle can be expressed as $\theta_{\text{max}} = 28.6^\circ \times H_{\text{an}}/H_{\text{ex}}$ when $K_1 = 0$. Here, K_1 is the anisotropy constant between the two sublattices, and H_{an} and H_{ex} are the anisotropy and exchange fields, respectively. This indicates that although the isotropic exchange interactions are

always dominant in uniaxial antiferromagnets, the value of $H_{\text{an}}/H_{\text{ex}}$ is large in Gd_5Ge_4 when compared to some other transition metal systems, thus leading to a relatively large θ_{max} .

The unusually large $H_{\text{an}}/H_{\text{ex}}$ is a reflection of the fact that anisotropic exchange interactions in Gd_5Gd_4 are stronger than in many other Gd-based systems. The magnetic anisotropy of the title compound is probably dominated by the anisotropic exchange interactions due to its distinctly layered²⁹ and, therefore, anisotropic orthorhombic crystal structure. This behavior is similar to that observed in GdRu_2Si_2 and GdRu_2Ge_2 ,⁴¹ where the low temperature anisotropy may originate from anisotropic exchange interactions caused by the anisotropy of the tetragonal lattice. Anisotropic exchange interactions are also a likely origin of the anisotropy observed in GdCu_2 , where a model based on the anisotropy of the classical dipole-dipole exchange fails to describe the easy magnetization direction in this noncollinear amplitude-modulated magnetic structure,⁴² even though this model works well for other Gd-based compounds, e.g., GdAg , GdAu_2Si_2 , and GdCu_2In .⁴³

CONCLUSIONS

The angular dependence of the spin-flop transition in single crystal Gd_5Ge_4 has been investigated by magnetization measurements. When the magnetic field vector is tilted away from the antiferromagnetic easy axis (c axis) toward the b axis, a first order spin-flop transition is observed over a wide range of tilt angles $0 < \theta < 65^\circ$. However, when the field vector is tilted away from the c axis toward the a axis, the first order spin-flop transition is only observed over a small range of tilt angles ($0 < \theta < 20^\circ$). When θ exceeds 20° , the spin-flop transition becomes a second order transformation, suggesting that in the spin-flop AFM phase, the Gd moments are antiferromagnetically coupled along the a axis.

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