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Magnetic properties and magnetic phase diagrams of intermetallic compound $GdMn_2Ge_2$*

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A modified Yafet–Kittle model is applied to investigate the magnetic properties and magnetic phase transition of the intermetallic compound $GdMn_2Ge_2$. Theoretical analysis and calculation show that there are five possible magnetic structures in $GdMn_2Ge_2$. Variations of external magnetic field and temperature give rise to the first-order or second-order magnetic transitions from one phase to another. Based on this model, the magnetic curves of $GdMn_2Ge_2$ single crystals at different temperatures are calculated and a good agreement with experimental data has obtained. Based on the calculation, the H–T magnetic phase diagrams of $GdMn_2Ge_2$ are depicted. The Gd–Gd, Gd–Mn, intralayer Mn–Mn and interlayer Mn–Mn exchange coupling parameters are estimated. It is shown that, in order to describe the magnetic properties of $GdMn_2Ge_2$, the lattice constant and temperature dependence of interlayer Mn–Mn exchange interaction must be taken into account.

Keywords: rare earth intermetallics, magnetic phase transition, magnetic structure
PACC: 7500, 7530K, 7550D

1. Introduction

In recent years, the ternary intermetallic compounds RMn_2Ge_2, where R is rare earth element or Y, have been extensively studied due to their unusual magnetic properties. RMn_2Ge_2 compounds crystallize in the body-centred-tetragonal $ThCr_2Si_2$-type structure (space group $I4/mmm$), which consists of R, Ge and Mn layers stacked in the sequence R-Ge-Mn-Ge-R- along the c-axis.[1] From the magnetic point of view, RMn_2Ge_2 compounds have two different magnetic subsystems: R- and Mn-subsystems, and their magnetic properties are mainly determined by these two subsystems and their interactions. There are different kinds of exchange interactions in RMn_2Ge_2. A number of research works[1] indicate that the intralayer Mn–Mn exchange interaction is the strongest and gives rise to the magnetic order temperature of RMn_2Ge_2 as high as 350–450 K, depending on R. The interlayer Mn–Mn and R-Mn exchange interactions have the same order but they are substantially less than the intralayer Mn–Mn exchange interaction; however, their interplay is reflected in the magnetic properties of magnetically ordered state. The smallest one is the R–R exchange interaction. Many studies[2,3] have shown that the interlayer Mn–Mn exchange interaction is very sensitive to the intralayer Mn–Mn distance, and consequently the lattice constant a. When a is larger than a critical value a_k, the interlayer Mn–Mn exchange coupling is ferromagnetic, otherwise it is antiferromagnetic. For this reason, the Mn-sublattice in most of light rare-earth compounds has a ferromagnetic structure, and the intrinsic magnetic structure of Mn-sublattice in the compounds with heavy rare earths is antiferromagnetic. Venturini and co-workers[3–6] have performed extensively neutron diffraction studies on RMn_2Ge_2. Their studies indicated that in the light rare-earth compounds the Mn magnetic structure of the same sublayer is not collinear ferromagnetic, but there is a large antiferromagnetic component perpendicular to the c-axis. Their investigations have also shown that the intralayer Mn–Mn exchange interaction is ferromagnetic for the intralayer Mn–Mn distance d_{Mn-Mn} lower than 0.248 nm and antiferromagnetic for d_{Mn-Mn} greater than 0.289 nm, whereas the mixed planes, where ferromagnetic and antiferromagnetic components coexist, are observed for in-

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termediate distances. Recently, we have studied the spontaneous magnetic phase transition by using the x-ray diffraction method.[7,8] Our studies show that the high-temperature paramagnetism-antiferromagnetism magnetic transitions in light rare-earth compounds will accompany the negative magnetoelastic anomaly of lattice constant c, whereas a does not change, indicating that the antiferromagnetic component of intralayer Mn–Mn exchange interaction is correlated to c. Just because of these peculiar properties of exchange interactions, the $R\mathrm{Mn_2Ge_2}$ compounds display abundant magnetic properties.

In $R\mathrm{Mn_2Ge_2}$ with heavy rare-earth elements, both the interlayer Mn–Mn exchange interaction and R-Mn exchange interaction are antiferromagnetic, thus these compounds can be considered as two-sublattice ferrimagnet with antiferromagnetic exchange interaction inside one of the sublattices. The competition between R-Mn and interlayer Mn–Mn exchange interaction makes the magnetic structure of Mn-subsystem unstable; when the magnetic parameters and external conditions (temperature, magnetic field, etc.) change, there may occur a series of magnetic structures and different kinds of magnetic transitions. In accordance with the magnetic and neutron diffraction studies,[9–13] the heavy rare-earth $R\mathrm{Mn_2Ge_2}$ compounds (R=Gd, Tb and Dy) have collinear ferrimagnetic or antiferromagnetic structures. When $T_\mathrm{t} < T < T_\mathrm{N}$ (for $\mathrm{GdMn_2Ge_2}$, T_t=97 K, T_N=365 K), the Mn subsystem has a collinear antiferromagnetic structure, the Mn magnetic moments couple ferromagnetically to each other in the same sublayer, where the interlayer Mn–Mn exchange interaction is antiferromagnetic, and the Mn moments are along the c-axis. As temperature decreases to T_t, the Mn-subsystem undergoes a first-order magnetic transition from antiferromagnetic to ferromagnetic state; at the same time, the Gd sublattice becomes ferromagnetically ordered and couples antiferromagnetically with Mn-subsystem. In an external magnetic field metamagnetic transition also occurs in $\mathrm{GdMn_2Ge_2}$.[10]

In Refs.[12] and [13], we have discussed the magnetic properties of the systems $\mathrm{Gd_{1-x}Y_xMn_2Ge_2}$ at low temperatures (T <100 K) and $\mathrm{Gd_{1-x}La_xMn_2Ge_2}$, based on a modified Yafet–Kittle model and given a good description. In this paper we will use this model to study the magnetic properties and magnetic phase transition of $\mathrm{GdMn_2Ge_2}$ single crystals by taking into account the lattice constant and temperature dependence of interlayer Mn–Mn exchange coupling.

2. Theoretical model

As stated in the introduction, the R-Mn and interlayer Mn–Mn exchange couplings in $\mathrm{GdMn_2Ge_2}$ are all antiferromagnetic, hence it can be considered as a two-sublattice ferrimagnet with a negative exchange interaction inside one of the sublattices. In accordance with the Yafet–Kittle model it should be expected that the Mn-subsystem is split into two sub-sublattices with the same magnetic moments, but with different orientations to the crystal axis, signalling the formation of a triangular magnetic structure. In as much as Gd is an S-ion with no orbital moment, the whole magnetic anisotropy of $\mathrm{GdMn_2Ge_2}$ can be attributed to the Mn-subsystem. According to the Yafet–Kittle model,[14] the magnetic energy of the $\mathrm{GdMn_2Ge_2}$ system can be written as

$$\begin{aligned} E = & -\lambda_{\mathrm{Gd\text{–}Mn}}\boldsymbol{M}_{\mathrm{Gd}}\cdot(\boldsymbol{M}_{\mathrm{Mn1}}+\boldsymbol{M}_{\mathrm{Mn2}}) \\ & -\lambda_{\mathrm{Mn\text{–}Mn}}\boldsymbol{M}_{\mathrm{Mn1}}\cdot\boldsymbol{M}_{\mathrm{Mn2}}-\lambda'_{\mathrm{Mn\text{–}Mn}}M^2_{\mathrm{Mn}} \\ & -\frac{1}{2}\lambda_{\mathrm{Gd\text{–}Gd}}M^2_{\mathrm{Gd}}+k(M^2_{\mathrm{Mnc1}}+M^2_{\mathrm{Mnc2}}) \\ & -\boldsymbol{H}\cdot(\boldsymbol{M}_{\mathrm{Gd}}+\boldsymbol{M}_{\mathrm{Mn1}}+\boldsymbol{M}_{\mathrm{Mn2}}), \end{aligned} \tag{1}$$

where the first term describes the exchange interaction between Gd and Mn subsystems ($\lambda_{\mathrm{Gd\text{–}Mn}} < 0$), the second term corresponds to the interlayer Mn–Mn interaction ($\lambda_{\mathrm{Mn\text{–}Mn}} < 0$ for heavy rare earth compounds), the third term is the intralayer Mn–Mn exchange ($\lambda'_{\mathrm{Mn\text{–}Mn}} > 0$), the fourth term is the exchange interaction in the Gd sublattice, and the fifth term is associated with the magnetic anisotropy of the Mn subsystem (M_{Mnc} is the magnetization component along the tetragonal axis c and k is related to the uniaxial anisotropy constant K as $K=-2kM^2_{\mathrm{Mn}}$). The final term reflects the Zeeman energy.

From the equilibrium conditions one can find five possible magnetic phases, as shown in Fig.1. In the first phase the magnetic moments of the two manganese sub-subsystems are parallel to each other and antiparallel to the gadolinium moment. This case means the formation of the collinear ferrimagnetic structure (Fi). For next two phases, the magnetic moments of Mn sublattices are split apart from each other and form an angle with Gd moment, giving rise to a triangular magnetic structure (T and T′). The difference between these two phases consists in the Gd moment orientation with respect to the c-axis: parallel and orthogonal, respectively. The fourth phase presents the antiferromagnetic ordering of Mn

subsystem (AF). Finally, when the magnetic field is strong enough, the magnetic moment of every sublattice direct along the field, signalling the formation of collinear ferromagnetic structure (F). The energy of each phase depends on the magnitude of exchange interactions of the subsystems, the energy of magnetic anisotropy and Zeeman energy. Variations of the parameters or external field may result in the possibility of different phase transitions. For example, at zero magnetic field, when the Gd–Mn exchange interaction exceeds the interlayer Mn–Mn exchange, the phase Fi is stable. Reduction of the Gd–Mn exchange interaction leads to the second-order phase transition for the triangle structure T. Further decrease of this interaction stimulates the increment of the angle between the c-axis and the Mn magnetic moment. As a result, the energy of magnetic anisotropy increases, provoking a first-order phase transition for the triangle phase T′, because in this phase the angle between the easy axis c and the Mn moment direction is less than that in T phase and, consequently, this phase has a smaller energy of magnetic anisotropy.

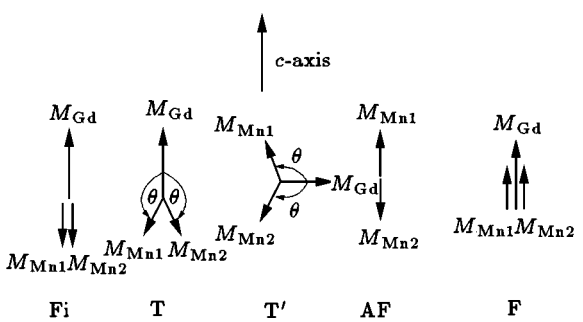


Fig.1. Possible magnetic phases in the intermetallic compound $GdMn_2Ge_2$.

At a finite temperature it is necessary to consider the temperature and field dependence of magnetization of the subsystems. In this case, in order to decide the magnetic structure one needs to calculate the equilibrium free energies of all possible phases and find the lowest one. Such numerical calculations were proposed earlier in Ref.[15]. We have extended and modified this approach on the basis of the molecular field model. According to the molecular field approximation of exchange interaction, the molecular fields acting on the Gd and Mn ions can be written as follows:

$$H_{\mathrm{ex,Gd}} = \lambda_{\mathrm{Gd-Gd}} M_{\mathrm{Gd}} + \lambda_{\mathrm{Gd-Mn}} M_{\mathrm{Mn1}} + \lambda_{\mathrm{Gd-Mn}} M_{\mathrm{Mn2}},$$

$$H_{\mathrm{ex,Mn}i} = \lambda_{\mathrm{Gd-Mn}} M_{\mathrm{Gd}} + \lambda_{\mathrm{Mn-Mn}} M_{\mathrm{Mn}j} + \lambda'_{\mathrm{Mn-Mn}} M_{\mathrm{Mn}i},$$

$$i = 1,2, \quad j = 2,1. \tag{2}$$

Here M_{Gd} and M_{Mn} represent the thermal average of magnetization of the Gd- and Mn-subsystems, respectively. We have $M_{\mathrm{Mn1}} = M_{\mathrm{Mn2}}$.

The Hamiltonians of Gd and Mn ions have the following form:

$$\begin{aligned} \hat{H}_{\mathrm{Gd}} &= g_J \mu_{\mathrm{B}} \hat{J} \cdot (H_{\mathrm{ex,Gd}} + H), \\ \hat{H}_{\mathrm{Mn}i} &= g_S \mu_{\mathrm{B}} \hat{S}_i \cdot (H_{\mathrm{ex,Mn}i} + H), \quad i = 1,2, \end{aligned} \tag{3}$$

where g_J, g_S represent the g-factors of Gd and Mn, $\hat{J}$ and $\hat{S}$ are the total angular momentum operator of Gd ion and the spin angular momentum operator of Mn ion, respectively.

The free energy of the system can be given as follows:

$$\begin{aligned} F = &- k_{\mathrm{B}} T \ln Z_{\mathrm{Gd}} - k_{\mathrm{B}} T \sum_i^{1,2} \ln Z_{\mathrm{Mn}i} + \frac{1}{2} M_{\mathrm{Gd}} \cdot H_{\mathrm{ex,Gd}} \\ &+ \frac{1}{2} \sum_i^{1,2} M_{\mathrm{Mn}i} \cdot H_{\mathrm{ex,Mn}i} + \sum_i^{1,2} K \sin^2 \theta_i, \end{aligned} \tag{4}$$

where k_{B} is Boltzmann constant, K and θ_i are the magnetic anisotropy constant of Mn ion and the angle between the $M_{\mathrm{Mn}i}$ and c-axis, Z_{Gd} and $Z_{\mathrm{Mn}i}$ are the partition functions of Gd and Mn ions, respectively:

$$\begin{aligned} Z_{\mathrm{Gd}} &= \mathrm{Tr}(-\hat{H}_{\mathrm{Gd}}/k_{\mathrm{B}}T), \\ Z_{\mathrm{Mn}i} &= \mathrm{Tr}(-\hat{H}_{\mathrm{Mn}i}/k_{\mathrm{B}}T), \quad i = 1,2. \end{aligned}$$

From the stable equilibrium condition we can gain the magnitude of thermal average M_{Gd} and $M_{\mathrm{Mn}i}$, as well as their directions with respect to the c-axis; consequently, we can determine the magnetic structure and the temperature and field dependence of magnetization of compounds.

As stated in the introduction, the interlayer Mn–Mn exchange interaction is very sensitive to the intralayer Mn–Mn distance, so it is necessary to take into account the dependence of lattice constant a and, consequently, the temperature dependence of molecular field coefficient $\lambda_{\mathrm{Mn-Mn}}$. Brabers *et al*[16] studied the temperature dependence of interlayer Mn–Mn exchange interaction of compound YMn_2Ge_2. Their studies indicate that the $\lambda_{\mathrm{Mn-Mn}}$ can be expressed as

$$\lambda_{\mathrm{Mn-Mn}} = \alpha - \beta\omega(T), \tag{5}$$

where α is the $\lambda_{\mathrm{Mn-Mn}}$ value at T=0 K and β a parameter. $\omega(T)$ represents the departure of the unit-cell volume variation with temperature from the equilibrium value at T=0 K. In our study we suppose

that the temperature and the lattice parameter dependence of λ_{Mn-Mn} in $GdMn_2Ge_2$ is the same as that in YMn_2Ge_2.

3. Results and discussion

In our calculation we use the values of Gd and Mn magnetic moments equal to $7\mu_B$ and $1.8\mu_B$ respectively at T=4.2 K.[15] The other parameters are evaluated by fitting a series of experimental data of $GdMn_2Ge_2$ single crystals (the experimental data include the magnetic transition temperatures, the magnetization curves along the crystallographic axes at different temperatures and the temperature dependence of magnetization). They are as follows: $\lambda'_{Mn-Mn}=245T/\mu_B$f.u., $\lambda_{Mn-Mn}=10.9T/\mu_B$f.u., $\lambda_{Gd-Mn}=8.4T/\mu_B$f.u., $\lambda_{Gd-Gd}=0.97T/\mu_B$f.u. and $K=6.4T\mu_B$/Mn.

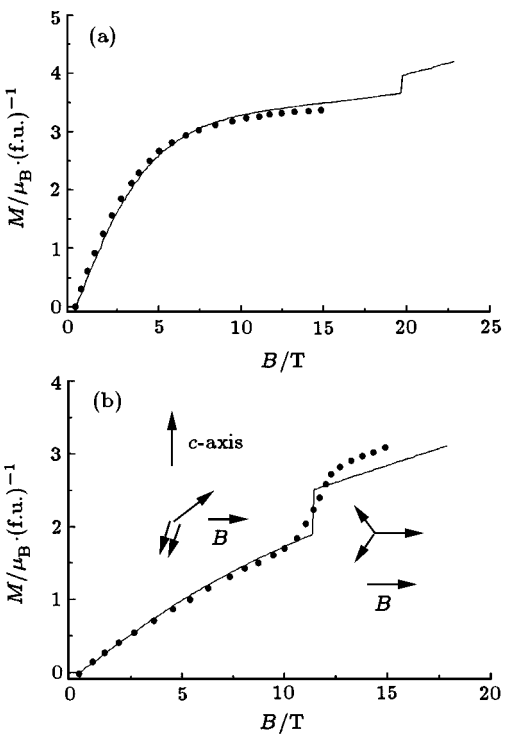


Fig.2. The magnetic curves of $GdMn_2Ge_2$ single crystals under the magnetic field perpendicular to the c-axis at 4.2 K (a) and 77 K (b). Lines are the calculated curves, solid circles represent experimental data (taken from Ref.[10]).

Figure 2 shows the calculated magnetization curves obtained using the above listed parameters and the corresponding experimental data of $GdMn_2Ge_2$ single crystals under the magnetic field perpendicular to the c-axis at T=4.2 and 77 K. It is obvious that a metamagnetic transition occurs in $GdMn_2Ge_2$ when the field exceeds a critical value. In accordance with the magnetic studies, when $T < T_t$=97 K, $GdMn_2Ge_2$ is ferrimagnet (Fi). Gd- and Mn-subsystems have a collinear ferromagnetic structure and antiferromagnetically couple to each other, the magnetizations of the two subsystems are both along the c-axis. When $T > T_t$, the Gd-subsystem becomes magnetically disordered and the Mn-subsystem transforms into a collinear antiferromagnetic state (AF), its magnetic moments being directed to the c-axis. Our theoretical analysis and numerical calculations have shown that, when T=77 K and $H \perp c$, under the action of magnetic field, the ferrimagnetic structure Fi first deforms, but the Mn-subsystem keeps the ferromagnetic structure. As the field increases up to a critical value, $GdMn_2Ge_2$ transforms from Fi into the non-collinear triangular magnetic structure T′ through a first-order magnetic transition; at the same time, the magnetic moments of Gd turn abruptly to the magnetic field direction (as shown in Fig.2(b)). Theory also predicts that this kind of field-induced first-order transition also occurs at higher fields when T=4.2 K.

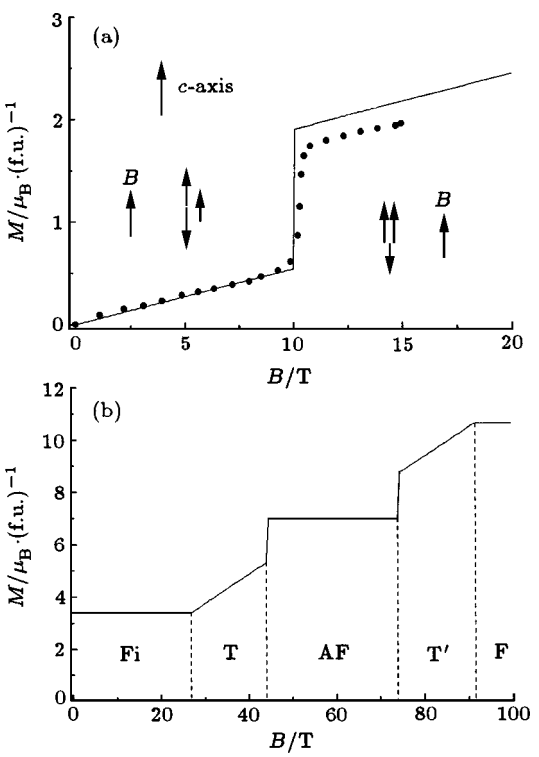


Fig.3. The magnetic curves of $GdMn_2Ge_2$ single crystals under the magnetic field parallel to the c-axis at 290 K (a) and 4.2 K (b). Lines are the calculated curves, solid circles represent experimental data (taken from Ref.[10]).

The calculated field dependence of magnetization and the corresponding experimental curves of $GdMn_2Ge_2$ with the field parallel to the c-axis at T=290 and 4.2 K are presented in Fig.3. When $T > T_t$, as stated above, the Mn-subsystem has a

collinear antiferromagnetic structure, numerical calculation shows that the external field induces the spin-flip transition in the Mn-subsystem from antiferromagnetic to ferromagnetic state (Fig.3(a)); at the same time, the Gd-sublattice becomes ferromagnetically ordered due to the Gd–Mn exchange interaction. When $T < T_t$, with the increment of magnetic field a series of transitions occur in $GdMn_2Ge_2$: at first, $GdMn_2Ge_2$ transforms from Fi to T through a second-order transition. With increasing the field, the angle between the Mn moment and the c-axis increases and the magnetic anisotropy increases gradually, and at a critical field, the T phase becomes unstable and the compound undergoes a first-order transition from T to AF. Further increasing the magnetic field, $GdMn_2Ge_2$ transforms again into the T phase. Finally, when the field is strong enough, all atomic moments are along the direction of the field, signalling the formation of a collinear ferromagnetic structure (F) (Fig.3(b)). The H–T magnetic phase diagram of $GdMn_2Ge_2$ and some available experimental data are depicted in Fig.4.

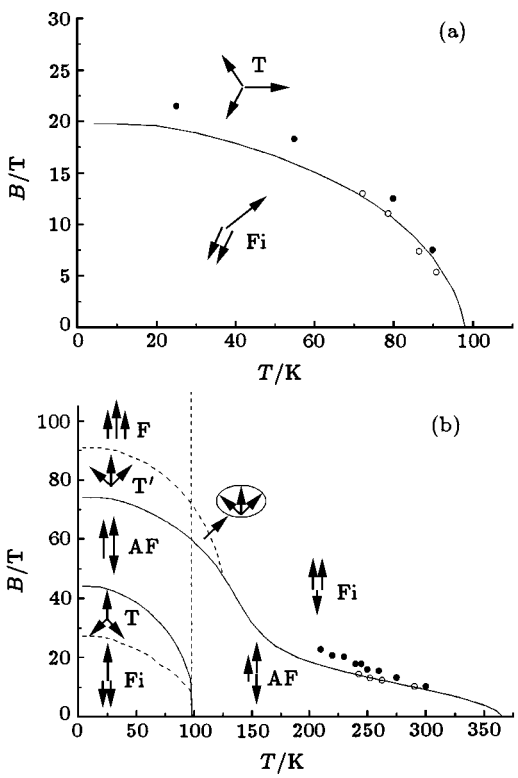


Fig.4. The magnetic phase diagrams of $GdMn_2Ge_2$ under the magnetic fields perpendicular (a) and parallel (b) to the c-axis. Calculated critical fields are given by curves for the second- (dashed) and first-order (solid) phase transitions. Experimental data from magnetic measurement of single crystal[10] and from our magnetostriction of polycrystalline sample[20] are indicated by open and closed circles, respectively.

It is worth to mention that our estimated value of λ_{Mn-Mn} is much lower than the value obtained for YMn_2Ge_2.[16] This is because, as mentioned in the introduction, the interlayer Mn–Mn exchange interaction is very sensitive to the lattice constant a. The lattice constant of the compound $GdMn_2Ge_2$ is larger than that of YMn_2Ge_2, consequently, the antiferromagnetic Mn–Mn interlayer coupling in $GdMn_2Ge_2$ should be weaker than in YMn_2Ge_2. Our evaluated value of intralayer Mn–Mn exchange coupling λ'_{Mn-Mn} is slightly higher, as compared with that reported in Ref.[15]. This disagreement can also be attributed to the necessity of taking into account the lattice constant dependence of interlayer Mn–Mn exchange parameter and, consequently, its temperature dependence. Besides, the value of λ_{Gd-Gd} obtained in our work is positive, which is inconsistent with Ref.[15], in which the Gd–Gd exchange coupling is antiferromagnetic. It is worth noting that, according to the study in Ref.[17], the Dy–Dy exchange coupling is also ferromagnetic. Tils *et al*[18] studied the Gd–Mn exchange coupling in $GdMn_2Ge_2$ by inelastic neutron scattering, the value they derived is $7.6T/\mu_B$f.u., which is in good agreement with our data.

It is necessary to point out that the calculated magnetization after the metamagnetic transition with the field parallel to the c-axis at T=290 K is larger than the experimental data (Fig.3(a)), whereas the calculated value of magnetization with the field perpendicular to the c-axis at T=77 K is smaller than the experimental value (Fig.2(b)). In our model we consider that the transitions are pure magnetic and suppose that the electronic states of the Mn-subsystem of $GdMn_2Ge_2$ are the same before and after the transition. However, it is possible that the electronic state of the Mn-subsystem changes at the transition. Neutron diffraction study of $TbMn_2Ge_2$[19] indicates there is a large change of local Mn magnetic moment ($\Delta M/M \approx 16\%$) at the magnetic transition in Mn-subsystem from ferromagnetic to antiferromagnetic structure. Furthermore, in our model we have neglected the lattice constant and temperature dependence of the intralayer Mn–Mn exchange interaction (for this interaction is not so sensitive to the lattice parameter as the interlayer Mn–Mn exchange coupling).

4. Conclusion

Based on the Yafet–Kittle model, we have investigated the magnetic properties and magnetic structure of the $GdMn_2Ge_2$ compounds in the molecular

field approximation. Theoretical analysis and numerical calculations show that there are five possible magnetic phases, and when magnetic parameters or external conditions (temperature and magnetic field) vary, $GdMn_2Ge_2$ undergoes first- or second-order magnetic transition from one phase to another. We have applied this model to the single crystal $GdMn_2Ge_2$ and obtained a good description of the magnetization curves along the different crystallographic axes and at different temperatures. The coefficients of the molecular fields of interlayer Mn–Mn, intralayer Mn–Mn, Gd–Mn and Gd–Gd exchange interaction are estimated. The H–T magnetic phase diagrams are depicted. It is shown that, in order to describe the magnetic properties of RMn_2Ge_2 compounds, it is necessary to take into account the distance dependence of interlayer Mn–Mn exchange interaction.

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