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Transport and magnetic study of the spin reorientation transition in the $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ magnetocaloric compound

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Abstract

Detailed measurements of the electrical resistivity $\rho(T)$, thermopower $S(T)$ and magnetization of $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ in the vicinity of the spin reorientation transitions observed in this compound are reported. Our results indicate a complex spin reorientation process associated with three different lattice sites occupied by the Tb ions. We identify two critical transition temperatures: one at $T_{\text{SR}_1} = 57$ K, as previously reported, and a new one at $T_{\text{SR}_2} = 40$ K. A simple model based on an approximate magnetic anisotropy energy is presented; it gives a satisfactory qualitative description of the main features of the reorientation processes.

1. Introduction

The $\text{R}_5(\text{Si}_x\text{Ge}_{1-x})_4$ system (the compounds with $\text{R} = \text{Gd}, \text{Tb}, \text{Nd}$ and Er) is under intensive study due to the appearance of unusual first-order magnetostructural transitions, giant magnetoresistance, colossal magnetostriction and particular features of the magnetic and electrical properties [1–4]. These compounds offer great technological promise as regards magnetic refrigeration and magnetostrictive/magnetoresistive sensors [5, 6].

In this paper we will concentrate on the Tb compounds. The corresponding crystallographic and magnetic phase diagrams have been established, as a function of both composition and temperature [7]. The nanoscopic layered structure of the $\text{R}_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds is formed by parallel rigid slabs of atoms (five atomic planes each) with interslab

covalent bonds linking interfacial Si(Ge)–Si(Ge) atoms at low temperatures. At higher temperatures the magnetostructural transition produces total or partial breaking of the covalent bonds (weaker interactions emerge, partly ionic), with drastic changes in the crystal lattice parameters.

At room temperature the $\text{Tb}_5(\text{Si}_x\text{Ge}_{1-x})_4$ compounds can exhibit three structural phases, orthorhombic O(I), O(II) (*Pnma*; Gd_5Si_4 type) and monoclinic (M; *P112₁/a*) according to the composition, all of them paramagnetic (PM) [7].

The compounds with $x > 0.65$ have the O(I) structure at room temperature, which persists down to the lowest temperatures. Under cooling, these compounds become ferromagnetic (FM1) through a second-order phase transition, with a Curie temperature (T_C) increasing linearly with the Si concentration. A spin reorientation transition occurs at a lower temperature (T_{SR}), from FM1 into another ferromagnetic phase (FM2), with T_{SR} almost independent of the Si concentration.

For $x < 0.35$ the Tb compounds are in the O(II) structure at room temperature. Under cooling, they first undergo a second-order magnetic transition from the PM into an AFM1 phase (Néel temperature T_N), and this is followed by a spin reorientation transition at a lower temperature.

The Tb compounds with $0.35 < x < 0.65$, which encompasses the particular composition discussed in this paper, are monoclinic (M; *P112₁/a* group) at room temperature, but upon cooling they have a magnetostructural first-order transition from the [PM, M] into the [FM1, O(I)] phases at a critical temperature T_S [7]. Recent detailed neutron diffraction work [8] shows that such crystallographic/magnetic changes are not simultaneous, as initially considered [7]; i.e. the transitions are not fully coupled [8, 9]: the magnetic transition precedes the structural one by a few degrees (under cooling). At low temperatures a spin reorientation transition was also observed in two Tb compounds with $x = 0.5$ and 0.6 , giving $T_{\text{SR}} \sim 65$ and 73 K respectively [7]. This identification was reached through neutron diffraction (intensity) measurements of the purely magnetic (100) + (020) reflections [7].

A detailed study of the magnetic structure (Rietveld refinements of neutron powder diffraction data) was performed for the sample with $x = 0.5$ [7] at a temperature above T_{SR} ($T = 85$ K) and also at $T = 2$ K. Such study shows that the directions of the Tb moments in $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ depend on the corresponding lattice sites, existing in three distinct kinds of positions in the unit cell, Tb1, Tb2 and Tb3 [7]. This introduces different site magnetic anisotropy constants, and one thus expects more complex spin reorientation transitions in this system than in simple cases where all magnetic sites are equivalent.

In this work we report a comprehensive study of the behaviour of the electrical resistivity $\rho(T)$ and thermopower $S(T)$ and their temperature derivatives $d\rho(T)/dT$ and $dS(T)/dT$ for $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ near the corresponding spin reorientation transitions, as well as the magnetization $M(T)$ and its derivative dM/dT , down to the lowest measured temperatures (~ 10 K). Our results indicate a complex spin reorientation process, revealing two critical transition temperatures: T_{SR_1} , as previously reported [8], and a second one (T_{SR_2}), as discussed below. To our knowledge, this is the first experimental identification of a second spin reorientation transition temperature in the $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ compound. A simple model based on an approximate magnetic anisotropy energy is presented, which gives a satisfactory qualitative description of the main features of the reorientation process.

2. Experimental details

The $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ compound was synthesized by the arc melting of 99.9 wt% pure Tb and 99.9999 wt% pure Si and Ge under a high purity argon atmosphere. Weight losses

during melting were negligible, and therefore the initial composition was assumed unchanged. The quality of the sample was checked by means of scanning electron microscopy and x-ray diffraction at room temperature. The measurements confirmed the presence of a monoclinic ($P112_1/a$) main phase with unit-cell parameters $a = 7.5080(5)$ Å, $b = 14.652(1)$ Å and $c = 7.7117(5)$ Å. A parallelepiped sample with the dimensions cross-section: 0.89×0.76 mm²; length: 1.9 mm was cut from the initial $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ ingot for our transport property measurements. For the thermopower measurements one end of the sample was fixed with indium to a temperature controlled copper block in a 3.7–300 K closed cycle refrigerator. At the other end a miniature heater produced a temperature gradient along the sample, typically ~ 1 K cm⁻¹.

The thermoelectric power (S) was measured between 10 and 300 K with a special four-lead method (two chromel + two Cu wires) joined at their ends so as to make two chromel–Cu junctions [10]. Such junctions were then fixed electrically with silver paint at two different points along the sample (hot and cold junctions), providing two differential thermocouples with the same temperature difference (ΔT): the copper wires provided a ΔV_1 voltage and the chromel wires a ΔV_2 voltage, which were directly measured with two Keithley 181 nanovoltmeters. We then have, at temperature T , $\Delta V_1 = [S(T)_{\text{Cr}} - S(T)]\Delta T$ and $\Delta V_2 = [S(T)_{\text{Cu}} - S(T)]\Delta T$, where S_{Cr} , S_{Cu} and S are the absolute thermopower values for chromel, copper and the sample respectively, at temperature T . Knowing the thermopower of chromel relative to copper, $S_0(T)$, we can eliminate ΔT from these equations, obtaining the required sample thermopower relative to copper at each temperature, $S(T)$. An automatic data acquisition system enabled $S(T)$ measurements every 200 mK, making possible direct (local) measurements of the temperature derivative dS/dT .

The electrical resistivity was measured with a standard four-point potentiometric method using a dc current of 30 mA through the sample, stable to $1:10^6$ [11]. The voltage was measured with a Keithley 182 nanovoltmeter with a resolution of 10 nV during the measurements. An automatic data acquisition system provided $\rho(T)$ data every 80 mK, making possible the direct (local) measurement of the temperature derivative $d\rho/dT$.

Both the magnetization and its temperature dependence $M(T)$ between 4 and 300 K were measured using a superconducting quantum interference device (SQUID) magnetometer.

3. Experimental results and analysis

3.1. Electrical resistivity

Figure 1 displays the temperature dependence of the electrical resistivity of $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ from 10 to 75 K, after subtracting the (large) residual resistivity $\rho_0 = 500$ $\mu\Omega$ cm, together with its temperature derivative $d\rho/dT$. Two clear anomalies are observed in $d\rho/dT$ near $T_{\text{SR}_1} = 57$ K and $T_{\text{SR}_2} = 40$ K. At T_{SR_1} the temperature dependence of $d\rho/dT$ changes abruptly, which we associate with the onset of the spin reorientation transition (see section 3.5). At T_{SR_2} the temperature dependence of $d\rho/dT$ again changes suddenly, marking the end of the spin reorientation process, i.e. completing the transformation of the FM1 phase into the FM2 phase. Between T_{SR_1} and T_{SR_2} , $d\rho/dT$ exhibits an irregular behaviour. To investigate the effects, two further $\rho(T)$ measurements were performed, taking the numerical (local) $d\rho/dT$ derivatives. These experiments confirmed the irregular behaviour of $d\rho/dT$ between T_{SR_1} and T_{SR_2} . The anomalies cannot be attributed to artefacts of differentiation (coupled with finite accuracy of the experimental data), since as soon as T decreases below T_{SR_1} (or increases above T_{SR_2}) a smooth $d\rho/dT$ curve is systematically observed. These results suggest erratic steps in the reorientation of the Tb1, Tb2 and Tb3 magnetic moments during the spin reorientation process.

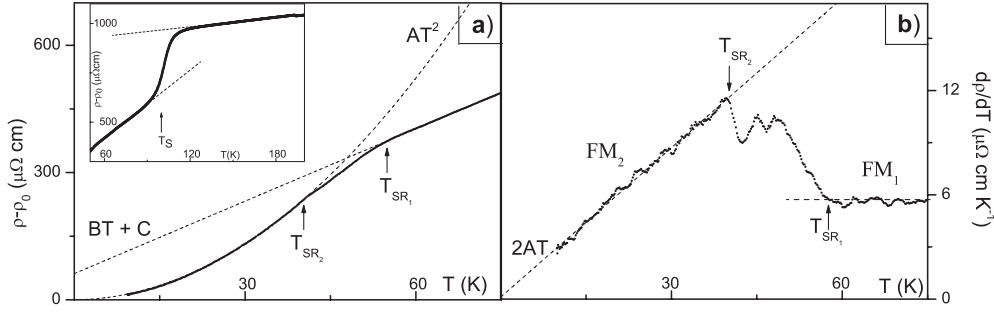


Figure 1. (a) Temperature dependence of the electrical resistivity (ρ) of $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ between 10 and 75 K, after subtraction of the residual resistivity (ρ_0). Inset: temperature dependence $\rho(T)$ between 50 and 180 K. (b) Temperature derivative of the electrical resistivity ($d\rho/dT$) between 10 and 75 K.

Below T_{SR_2} and all the way to the lowest measured temperatures, $d\rho/dT$ exhibits a linear temperature dependence extrapolating through the origin, which means a quadratic resistivity dependence, $\rho(T) = \rho_0 + AT^2$ with $A = 0.14 \mu\Omega \text{ cm K}^{-2}$. We associate this behaviour with spin wave electron scattering in the ferromagnetic FM2 phase, suggesting a quadratic spin wave dispersion relation, $w \propto k^2$ [12].

Remarkably, at T_{SR_1} and above, $\rho(T)$ increases almost linearly with temperature but does not extrapolate through the origin, i.e. $\rho(T) = BT + C$ with $B \simeq 5.73 \mu\Omega \text{ cm K}^{-1}$ and $C = 70 \mu\Omega \text{ cm}$. This linear behaviour is the same for both decreasing and increasing temperatures, being intrinsic to the FM1 phase just above T_{SR_1} . The finite intercept (at the origin) of the linear $\rho(T)$ part above T_{SR_1} indicates an extra constant resistivity in the FM1 phase, besides the residual resistivity ρ_0 . This suggests some temperature-independent spin disorder within the FM1 phase (orthorhombic I), on a scale of the order of or below the electron mean free path. As regards the linear term, one could think of the usual electron-phonon contribution to ρ . Such a term is clearly seen in the PM phase (monoclinic) above $T_S \sim 100 \text{ K}$, but its slope ($0.88 \mu\Omega \text{ cm K}^{-1}$; see the inset of figure 1(a)) is much smaller than that of the B coefficient in $\rho(T)$ just above T_{SR_1} , unless the orthorhombic I structure ($T < T_S$) exhibits an extremely high electron-phonon resistivity. It is thus unlikely that phonon resistivity dominates $\rho(T)$ near T_{SR_1} . One recalls that, according to the available neutron diffraction data [7], below T_{SR_1} the magnetic moments are essentially aligned along the ac bisection direction, whereas above T_{SR_1} they are oriented along the a -axis. Therefore, another possible explanation could be a significant change in the spin wave spectrum, occurring in such a way as to give a resistivity linear contribution, $\rho = BT$, in the FM1 phase.

3.2. Thermoelectric power

The behaviour of the thermoelectric power at low temperatures also confirms the two critical temperatures associated with the reorientation processes in $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$. This is clearly seen in figure 2(a), which displays the $S(T)$ data from 10 to 70 K.

Above $\sim 57 \text{ K}$ the thermopower exhibits a linear temperature dependence, $S(T) = \beta + \alpha T$ with $\beta \simeq -5.73 \mu\text{V K}^{-1}$ and $\alpha \simeq -0.017 \mu\text{V K}^{-2}$, leading to a strictly constant dS/dT behaviour (figure 2(b)).

Upon cooling, the $S(T)$ behaviour changes at $T_{\text{SR}_1} = 57 \text{ K}$ (figure 2(a)), corresponding to the onset of the spin reorientation process, similarly to what was observed for $d\rho/dT$. At $T_{\text{SR}_2} = 40 \text{ K}$ one observes an inflection point in $S(T)$, marking the end of the spin reorientation

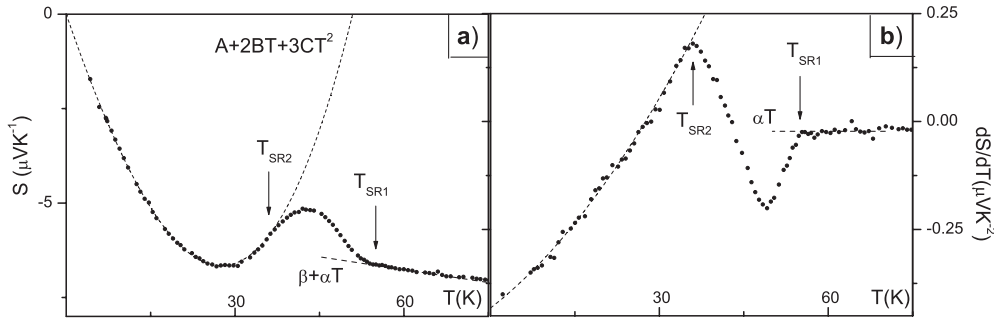


Figure 2. (a) The temperature dependence of the thermoelectric power (S) of $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ between 75 and 10 K. (b) The temperature derivative of the thermoelectric power (dS/dT).

process. From T_{SR_1} to T_{SR_2} , a non-monotonic behaviour is observed in dS/dT (figure 2(b)), suggesting competing effects in the reorientation process: dS/dT first decreases below T_{SR_1} , reaches a minimum at $T \sim 44$ K and then rises as T approaches T_{SR_2} . When we compare the dS/dT and the $d\rho/dT$ curves (figures 2(b) and 1(b)), the latter appears much more irregular between T_{SR_1} and T_{SR_2} . This could be due to the dominant dependence of the electrical resistivity on the electron scattering processes, which are very sensitive to short range order effects. Therefore, ρ is essentially linked to the averaged electron collision time over the Fermi surface. In contrast, $S(T)$ depends strongly on the energy derivatives and density of states of the conduction electrons near the Fermi surface (besides electron scattering), i.e. on the electron band features.

At temperatures below T_{SR_2} the $S(T)$ data are well described by a polynomial fit $S(T) = AT + BT^2 + CT^3$ with $A = -0.432 \mu\text{V K}^{-2}$, $B = 0.005 \mu\text{V K}^{-3}$ and $C = 6.83 \times 10^{-5} \mu\text{V K}^{-4}$. This behaviour is more neatly confirmed by the dS/dT curve (figure 2(b)), described by a quadratic polynomial below T_{SR_2} .

The quadratic term (B), usually characteristic of electron spin wave scattering [12, 13], again suggests the dominance of such excitations below T_{SR_2} . The cubic term coefficient (C), which begins to be important at temperatures around $\sim T_{\text{SR}_2}$, is probably associated with the phonon drag contribution, which at temperatures $T \ll \Theta_{\text{D}}$ should be proportional to $(T/\Theta_{\text{D}})^3$ [14]. At low temperatures (<10 K) the $S(T)$ dependence is practically linear, indicating the dominance of impurity scattering [14].

3.3. Magnetization and neutron diffraction

The temperature dependences of the magnetization $M(T)$ and its derivative dM/dT , obtained both in the zero-field-cooled (ZFC) and field-cooled (FC) regimes for an applied magnetic field of 20.3 Oe, are displayed in figures 3(a) and (b) respectively. The dM/dT curves were obtained from numerical differentiation of high resolution $M(T)$ data (0.5 K steps). The spin reorientation process is hardly observable in the $M(T)$ curve, but small changes are seen in the dM/dT curve between T_{SR_1} and T_{SR_2} . This seems to be reminiscent of the irregular behaviour previously observed for $d\rho/dT$ between $T_{\text{SR}_2} \simeq 40$ K and $T_{\text{SR}_1} \simeq 57$ K. An important anomaly is also observed at $T^* = 67$ K, where dM/dT exhibits a well defined minimum, but such an anomaly is not detected in our transport property measurements (figures 1 and 2). Besides the SQUID magnetization data referred to, we also have information on the basal plane component of the spontaneous magnetization as a function of temperature, from neutron

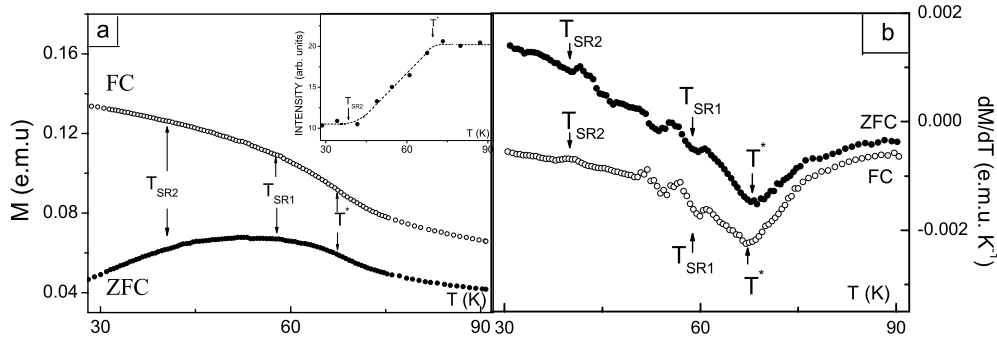


Figure 3. (a) $M(T)$ for $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ as a function of the temperature, in both the zero-field-cooled (ZFC) and the field-cooled (FC) regimes. Inset: temperature dependence of the neutron diffraction intensity of the purely magnetic (100) + (020) reflection, obtained for the $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ sample. (b) dM/dT for $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ as a function of the temperature.

diffraction data [7]. These data (inset of figure 3(a)) show that the reorientation transition actually starts at $T^* = 67$ K, apparently persisting down to $T_{\text{SR}_2} \simeq 40$ K.

3.4. A simplified view of the magnetic structure

Due to differences in local environment of the terbium ions in the unit cell, one has three distinct sites, Tb1 (4c symmetry), Tb2 (8d) and Tb3 (8d). This leads to different magnetic anisotropy site constants, and so different orientational processes as the temperature changes. Also, for each of the 8d positions (Tb2 and Tb3) the magnetic moment can still point in two distinct directions (α and β).

According to the neutron diffraction data [7], at $T = 85$ K the Tb1, Tb2 and Tb3 magnetic moment components in the ac plane make slightly different angles with the a -axis, $\theta_1 = 5^\circ$, $\theta_2 = 13^\circ$ and $\theta_3 = 3^\circ$ respectively, whereas the corresponding angles (with the a -axis) in the ab plane are $\phi_1 = 0^\circ$, $\phi_2 \sim 11^\circ$ and $\phi_3 \sim 4^\circ$. Therefore, at 85 K the spontaneous magnetization (M_S) is quasi-parallel to the a -axis, if one disregards the small ϕ_i angles.

At $T = 2$ K the magnetic moments of each Tb site are split into two subsets with slightly different canting angles, whose average values are $\theta_1 = 33^\circ$, $\theta_2 = 45^\circ$ and $\theta_3 = 34^\circ$.

The above results on the Tb magnetic moments are summarized in figures 4(a) and 5(a) for $T = 85$ K and figures 4(b) and 5(b) for $T = 2$ K. It is very clear that above the spin reorientation transition ($T = 85$ K) all the Tb ions essentially lie in the ac plane, and are mostly oriented along the a -axis. Below the spin reorientation transition (figure 4(b)) most of the Tb moments still lie close to the ac plane, except half of the Tb2 ions, which point in a direction (β) making a large angle ($\sim 33^\circ$) with the ac plane; the other half of the Tb2 moments lie in the ac plane. One notes that the markedly out-of-plane moments at low temperatures represent only 20% of the total number of moments. Therefore, the major reorientational processes occur in the ac plane, as can be seen by comparing figures 5(a) ($T = 85$ K) and (b) ($T = 2$ K), probably influencing the behaviour of different physical properties through the spin reorientation transition.

3.5. Magnetic anisotropy and spin reorientation processes

Considering only a dominant rotation of the Tb moments in the ac plane, from $\theta = 0^\circ$ to 45° , and disregarding the small ϕ_i variations in the ab plane, we consider an approximate

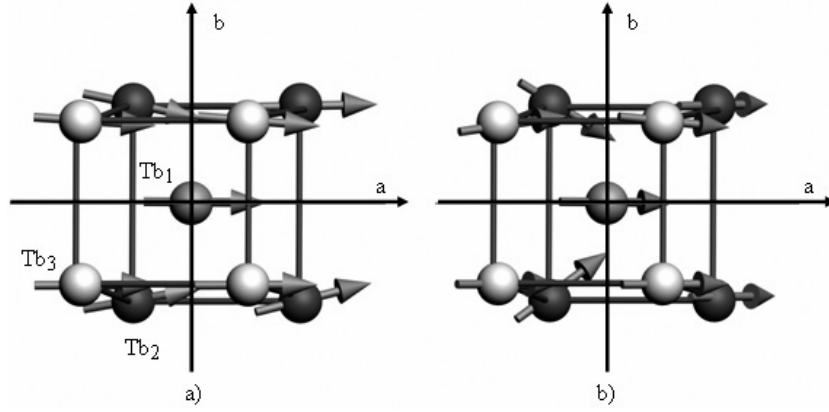


Figure 4. (a) Magnetic structure for $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ projected (orthogonally) into the ab plane at $T = 85$ K (grey, black and white atoms correspond to Tb1, Tb2 and Tb3 atoms respectively). (b) Magnetic structure projected into the ab plane at $T = 2$ K.

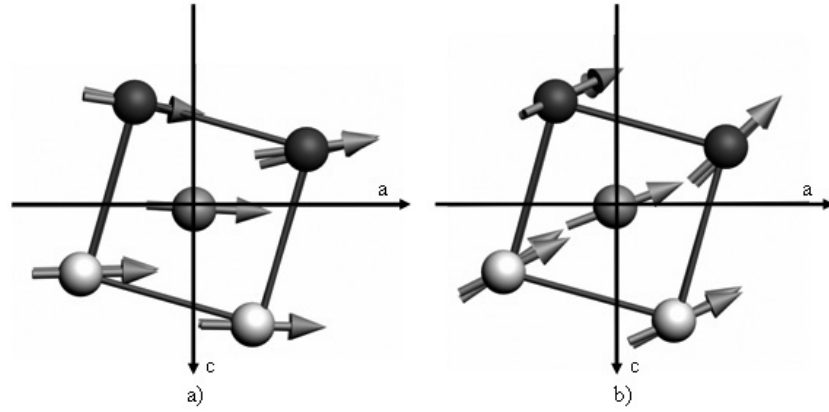


Figure 5. (a) Magnetic structure for $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ projected (orthogonally) into the ac plane at $T = 85$ K (grey, black and white atoms correspond to Tb1, Tb2 and Tb3 atoms respectively). (b) Magnetic structure projected into the ab plane at $T = 2$ K.

anisotropy magnetic energy (E_a) with the required symmetry:

$$E_a = K_1 \sin^2(2\theta) + K_2 \sin^4(2\theta). \quad (1)$$

Near T_{SR_1} we assume that the first anisotropy coefficient has the usual form $K_1(T) \simeq \alpha(T - T_{\text{SR}_1}) + \beta(T - T_{\text{SR}_1})^2$, with α and $\beta > 0$. K_2 is the second anisotropic constant and the positive α and β coefficients ensure $\theta = 0$ for $T > T_{\text{SR}_1}$. Minimizing the energy with respect to θ and considering the above approximation for $K_1(T)$ over the whole reorientation range, one obtains three possible spin structures: $\theta = 0$ for $T > T_{\text{SR}_1}$, $\theta = \pi/4$ for $T \leq T_{\text{SR}_2}$ (see below) and $0 < \theta < \pi/4$ between T_{SR_1} and T_{SR_2} (as follows):

$$\theta = \frac{1}{2} \sin^{-1} \sqrt{-\frac{\alpha(T - T_{\text{SR}_1}) + \beta(T - T_{\text{SR}_1})^2}{2K_2}}. \quad (2)$$

This shows that a spin reorientation transition at $T = T_{\text{SR}_1}$ requires $K_2 > 0$ and $K_1 < 0$, whereas the transition at T_{SR_2} requires $(\alpha(T_{\text{SR}_2} - T_{\text{SR}_1}) + \beta(T_{\text{SR}_2} - T_{\text{SR}_1})^2)/2K_2 = -1$, i.e. $K_1(T_{\text{SR}_2}) = -2K_2(T_{\text{SR}_2})$.

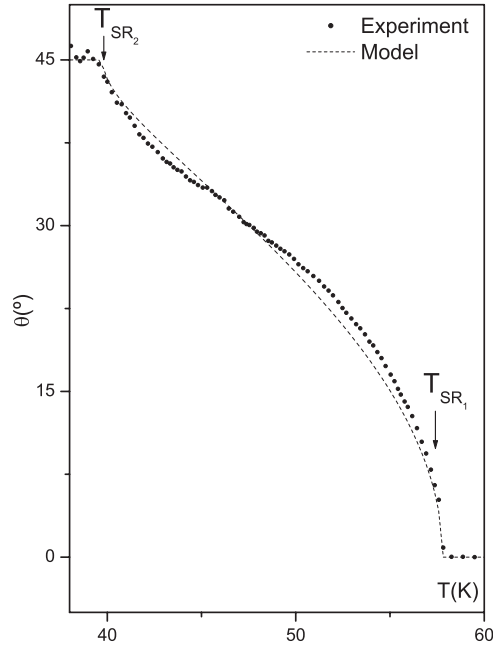


Figure 6. Temperature dependence of the angle θ taken from experimental results for $\rho(T)$ for the $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ compound (point) between 58 and 38 K; the dotted line gives the theoretical results according to equation (2).

One can also calculate the $\theta(T)$ dependence directly from the experimental $\rho(T)$ values. For this one recalls that ρ should satisfy the condition $\rho(\theta) = \rho(\theta \pm \pi/4)$ implicit in equation (1) and a second-order development of ρ (in powers of $\cos(2\theta)$) is generally sufficient for accounting for the mean effect of the reorientation process [15]:

$$\rho(T) = \rho_0 + a(T) + b(T) \cos^2(2\theta). \quad (3)$$

Here ρ_0 is the residual resistivity, $a(T) = AT^2$ is the low temperature resistivity (extrapolated into the reorientation range) and the anisotropy coefficient $b(T) \propto M_S^2(T)$ gives the magnitude of the spin reorientation effects on the electrical resistivity. Assuming that $b \simeq \text{constant}$ in the small temperature range where the spin reorientation occurs, we can estimate b , since at T_{SR_1} one has $\theta = 0^\circ$, so $b = \rho(T_{\text{SR}_1}) - a(T_{\text{SR}_1}) - \rho_0$. We can then extract, from $\rho(T)$, the $\theta(T)$ dependence between T_{SR_2} and T_{SR_1} [15]:

$$\theta(T) = \frac{1}{2} \cos^{-1} \sqrt{\frac{\rho(T) - a(T) - \rho_0}{\rho(T_{\text{SR}_1}) - a(T_{\text{SR}_1}) - \rho_0}}. \quad (4)$$

Such $\theta(T)$ behaviour is shown in figure 6 (heavy points), together with the best fit to the anisotropy model formula (equation (2); dotted line), obtained with $\alpha/2K_2 \sim 0.096 \text{ K}^{-1}$ and $\beta/2K_2 \sim 0.0023 \text{ K}^{-2}$.

It is clear that the agreement is only qualitative, which may be due to the oversimplified model used. In fact we only considered a common θ rotation in the ac plane, disregarding the θ_i differences among the Tb1, Tb2 and Tb3 ions, as well as the ϕ rotations in the ab plane (that occur in Tb2 and Tb3 ions). We also assumed $K_1(T) = \alpha(T - T_{\text{SR}_1}) + \beta(T - T_{\text{SR}_1})^2$ between T_{SR_1} and T_{SR_2} and $b = \text{constant}$ over such a range.

It seems desirable to have refined neutron diffraction data covering in greater detail the temperature range between T_{SR_2} and T_{SR_1} , to provide direct information on the corresponding

$\theta(T)$ dependence, so as to allow comparison with the approximate results obtained in the present work. In addition, such data may help in the (independent) discrimination between T_{SR_1} , T_{SR_2} and T^* .

4. Conclusions

Our transport (ρ , $d\rho/dT$, S , dS/dT) and magnetic measurements (M , dM/dT) on $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ reveal the existence of a complex spin reorientation process within the FM phase at low temperatures. According to our magnetic data and previous neutron diffraction work [7], the spin reorientation process starts at $T^* = 67$ K and ends at $T_{\text{SR}_2} = 40$ K. However, due to the different site anisotropies (Tb1, Tb2, Tb3), corresponding spin reorientation transitions may occur at slightly different temperatures. In particular, the transport properties seem to be insensitive to the spin reorientations between $T^* = 67$ K and $T_{\text{SR}_1} = 57$ K. They are however very sensitive to the reorientation processes between $T_{\text{SR}_1} = 57$ K and $T_{\text{SR}_2} = 40$ K. In particular, they suggest erratic reorientations between T_{SR_1} and T_{SR_2} , indicating local energy barriers preventing smooth spin rotation as T is lowered. One recalls that according to equations (2) and (3) one would expect a linear $d\rho/dT$ dependence between T_{SR_1} and T_{SR_2} . A simple phenomenological treatment, based on an approximate model of the magnetic structure of $\text{Tb}_5(\text{Si}_{0.5}\text{Ge}_{0.5})_4$ at low temperatures, gives a satisfactory description of the main features of the reorientation process and the corresponding effect on the electrical resistivity between T_{SR_1} and T_{SR_2} .

Acknowledgments

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References

- [1] Pecharsky V K and Gshneider K A Jr 1997 *Phys. Rev. Lett.* **78** 4494
- [2] Pecharsky V K and Gshneider K A Jr 1997 *J. Appl. Phys. Lett.* **70** 3299
- [3] Gshneider K A Jr, Pecharsky V K, Pecharsky A O, Ivchenko V V and Levin E M 2000 *J. Alloys Compounds* **303/304** 214
- [4] Pecharsky V K and Gshneider K A Jr 1997 *J. Magn. Magn. Mater.* **167** L179
- [5] Pecharsky V K and Gshneider K A Jr 1999 *J. Appl. Phys.* **85** 5365
- [6] Pecharsky V K, Pecharsky A O and Gshneider K A Jr 2002 *J. Alloys Compounds* **433** 362
- [7] Ritter C, Morellon L, Algarabel P A and Ibarra R 2002 *Phys. Rev. B* **65** 94405
- [8] Morellon L, Ritter C, Magen C, Algarabel P A and Ibarra R 2003 *Phys. Rev. B* **68** 024417
- [9] Morellon L, Arnold Z, Magen C, Ritter C, Prokhnenko O, Skorokhod Y, Algarabel P A, Ibarra M R and Kamarad J 2004 *Phys. Rev. Lett.* **93** 137201
- [10] Sousa J B, Braga M E, Correia F C, Carpinteiro F, Morellon L, Algarabel P A and Ibarra R 2002 *J. Appl. Phys.* **91** 4457
- [11] Sousa J B, Braga M E, Correia F C, Carpinteiro F, Morellon L, Algarabel P A and Ibarra R 2003 *Phys. Rev. B* **67** 9353
- [12] Kasuya T 1959 *Prog. Theor. Phys.* **22** 227
- [13] Pinto R, Sousa J B, Correia F C, Araújo J P, Braga M E, Pereira A M, Morellon L, Algarabel P A, Magen C and Ibarra R 2005 *J. Magn. Magn. Mater.* **290/291** 661
- [14] Barnard R D 1972 *Thermoelectricity in Metals and Alloys* (London: Taylor and Francis)
- [15] Sousa J B, Moreira J M, Del Moral A, Algarabel P and Ibarra R 1990 *J. Phys.: Condens. Matter* **2** 3897