Magnetotransport through the spin-reorientation transition in $Tm_2Fe_{14}B$

2001 J. Phys.: Condens. Matter 13 303

(http://iopscience.iop.org/0953-8984/13/2/307)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 303–310 www.iop.org/Journals/cm PII: S0953-8984(01)17042-6

Magnetotransport through the spin-reorientation transition in $Tm_2Fe_{14}B$

Jolanta Stankiewicz[1,3], **Juan Bartolomé**[1] **and Satoshi Hirosawa**[2]

[1] Instituto de Ciencia de Materiales de Aragón, Consejo Superior de Investigaciones Científicas and Universidad de Zaragoza, 50009-Zaragoza, Spain

[2] Sumitomo Special Metals Company Limited, Egawa, Shimamotocho, Mishimagun, Osaka 618, Japan

E-mail: jolanta@posta.unizar.es (J Stankiewicz)

Received 11 September 2000

Abstract

The electrical resistivity and Hall effect for a single crystal of $Tm_2Fe_{14}B$ have been measured over the range of temperature (T) from 4 to 600 K in magnetic fields of up to 5 T. The resistivity exhibits a small step-like rise at the spin-reorientation temperature T_s, which is 311 K, and a broad minimum at 535 K. In addition, the Hall coefficient shows an anomaly at T_s, and drops sharply as T approaches the Curie temperature (549 K) from below. The lower-temperature anomalies, both in the resistivity and in the Hall coefficient, show that the spin-reorientation transition in $Tm_2Fe_{14}B$ is of first order. The high-temperature Hall anomaly is probably produced by critical spin fluctuations near the Curie point. Dominant scattering mechanisms that underlie the Hall effect and magnetoresistance in $Tm_2Fe_{14}B$ are inferred.

1. Introduction

Spin-reorientation transitions (SRT) in magnetic materials have been extensively studied for several decades. They are still of much interest in compounds with rare-earth ions which have strong magnetic anisotropies [1]. Recently, SRT studies of thin ferromagnetic films are attracting increasing attention because of the fundamental and technological interest in this type of material [2].

Spontaneous spin reorientation in the absence of an external field takes place in some of the $R_2Fe_{14}B$ (R = lanthanide series) compounds, which are important for technological applications as permanent magnets. The $R_2Fe_{14}B$ compounds crystallize in a tetragonal structure. In $Tm_2Fe_{14}B$, the Tm- and Fe-sublattice moments couple ferrimagnetically; the Curie temperature is 549 K [3]. At high temperatures, the Tm- and Fe-sublattice magnetic moments are collinear along the tetragonal c-axis. At low temperatures, the easy-magnetization [100] direction is on the basal plane when the crystalline electric field of Tm is dominant. All

[3] Author to whom any correspondence should be addressed.

magnetic moments lie on this plane but they are not collinear. A five-sublattice model, in which two planar pairs of R sublattices are canted symmetrically away from the direction of the Fe moments, has been proposed for $Tm_2Fe_{14}B$ and confirmed by neutron diffraction experiment [4].

As the temperature increases, a SRT takes place at approximately 311 K. It happens when the uniaxial anisotropy of the Fe sublattice dominates over the planar anisotropy of the R sublattices. Features related to the spin-reorientation transition have been found in nearly all kinds of measurements performed on $Tm_2Fe_{14}B$. These include Mössbauer spectra [5], high-resolution neutron powder diffraction [6], magnetic ac susceptibility [7], ac resistivity [8], and heat capacity [9] ones. In all experiments the observed physical magnitudes change considerably over a very narrow temperature range. It seems therefore that the easy-magnetization direction rotates abruptly from the basal plane towards the *c*-axis, in a manner characteristic of first-order phase transitions. However, an analysis that takes into account crystalline-field effects and exchange fields shows that whether spin reorientation occurs in the Tm compound as a first- or a second-order transition depends on the values of the parameters used [10]. In addition, it was found that spin rotation takes place over a very small temperature range, even for a second-order transition. Thus, the nature of the SRT in $Tm_2Fe_{14}B$ has not yet been clearly established.

Electrical transport coefficients are sensitive to phase transitions in magnetic materials [11]. It has been argued that high-resolution $d\rho/dT$ measurements might provide unambiguous evidence for the first- or second-order nature of the transition [12]. Large variations of the Hall effect as in $Nd_2Fe_{14}B$ can be expected near critical points [13]. These variations are related to strong spin fluctuations. However, the behaviour of the spontaneous Hall effect (SHE) should be quite different through a first-order transition where spin fluctuations are much weaker except at the transition point.

In this paper, we report results of electrical resistivity (ρ), magnetization, and Hall effect measurements on $Tm_2Fe_{14}B$ in the temperature range of 4 to 600 K and in magnetic fields of up to 5 T. It is our aim to study the electrical transport coefficients through the SRT in $Tm_2Fe_{14}B$ to pinpoint the nature of this transition and to determine the dominant mechanisms that underlie the Hall effect and magnetoresistance.

The experimental procedure is described in section 2. Results of magnetoresistance and Hall effect measurements are reported and discussed in section 3. Conclusions are drawn in section 4.

2. Experiment

All measurements were made on a $Tm_2Fe_{14}B$ single crystal that we grew by the floating-zone melting technique [3]. The electrical resistivity and Hall effect measurements were performed with a six-probe method on a bar-shaped sample that we spark cut from the bulk single crystal. Its dimensions were $1 \times 1.2 \times 4$ mm^3. We determined its crystallographic orientation by means of x-ray diffraction. Before measurements, the sample was polished and checked for possible cracks. Contact leads were ultrasonically soldered to the sample.

The Hall resistivity ρ_H was measured as a function of magnetic field up to 0.6 T at all experimental points. In addition, the variation of ρ_H with magnetic field, up to 5 T, was checked at 5, 100, and 280 K. These data were taken simultaneously with the magnetization measurements with a SQUID magnetometer. Usually, the sample was zero-field cooled before performing measurements, although we did not find any significant difference between zero-field-cooled and field-cooled data. The magnetization was measured with a Faraday balance for temperatures higher than 400 K.

We used two different configurations in our measurements. Since the edges of the sample did not come out parallel to the principal crystallographic axes, the latter were not parallel to the external magnetic field. All measurements were performed with the current flowing along the longest dimension of the sample which was at 80° with respect to the c-axis and perpendicular to the external field. In one of the configurations used, the external magnetic field was at an angle of about 20° with the [100] direction. In the second one, the magnetic field was rotated by 90° towards the [010] direction.

3. Results and discussion

How the zero-field resistivity of $Tm_2Fe_{14}B$ varies with temperature, in the range from 4 to 600 K, is exhibited in figure 1. At very low temperatures, the measured resistivity becomes constant. ρ increases rapidly with temperature from 50 K to about 220 K and more slowly thereafter. It shows a small sharp rise at $T = 311$ K and a broad minimum at approximately 535 K. As shown in our earlier papers, the overall behaviours of the electrical resistivities of all $R_2Fe_{14}B$ compounds are alike [11, 14]. What is different in $Tm_2Fe_{14}B$ is the existence of a clear-cut anomaly at T_s, which is shown in the inset of figure 1. Therein, ρ increases seemingly discontinuously within a temperature interval of less than 1 K, and $\mathrm{d}\rho/\mathrm{d}T$ exhibits a sharp symmetric peak, which is very reminiscent of the specific heat behaviour. Since the effect of the magnetic anisotropy on the resistivity depends on the angle θ between the magnetization and the electric current [15], then $\mathrm{d}\rho/\mathrm{d}T = (\partial\rho/\partial T)_\theta + (\partial\rho/\partial\theta)_T\ \mathrm{d}\theta/\mathrm{d}T$. The angle θ is a continuous function of temperature in a second-order SRT. Then, its temperature derivative exhibits a broad, lambda-type anomaly and a corresponding singularity in $\mathrm{d}\rho/\mathrm{d}T$ ensues. On

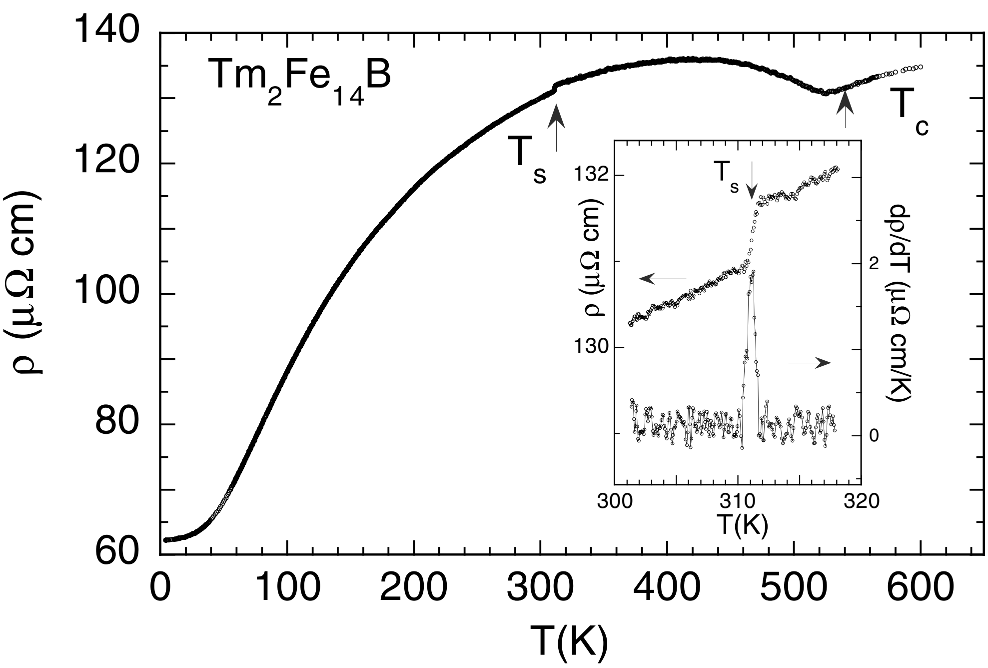

Figure 1. Zero-field resistivity data points versus temperature for $Tm_2Fe_{14}B$ single crystals. The inset shows $\rho(T)$ and its temperature derivative near T_s.

the other hand, in a first-order transition, θ changes discontinuously at T_s, which leads to an abrupt step in the electrical resistivity and to a sharp peak in $\mathrm{d}\rho/\mathrm{d}T$. Thus, inspection of figure 1 suggests that the SRT in $Tm_2Fe_{14}B$ is of first order.

Analysis of the Hall effect data follows. Figure 2 shows how the Hall resistivity of $Tm_2Fe_{14}B$ varies with the magnetization at three different temperatures. Clearly, the Hall resistivity data, which are hole-like, follow the magnetization of the sample. The low-field Hall resistivity and magnetization are shown in figure 3 for the two experimental configurations. ρ_H passes through a small maximum near T_s, where the magnetization drops. It decreases sharply near the Curie temperature.

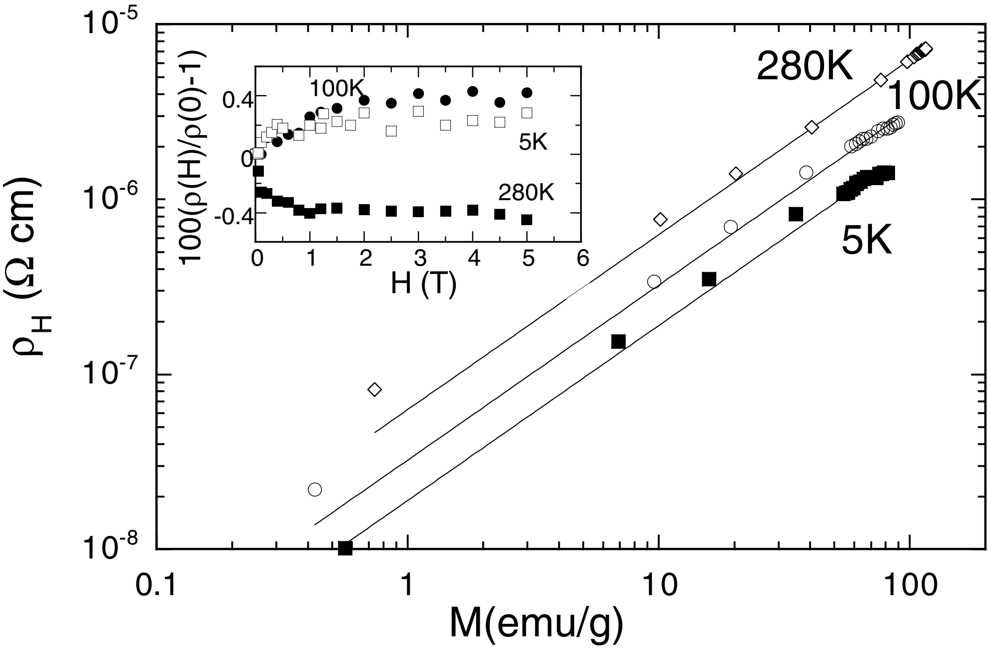

Figure 2. The Hall resistivity as a function of magnetization for $Tm_2Fe_{14}B$ single crystals for three different temperatures. The inset shows the transverse magnetoresistance of $Tm_2Fe_{14}B$ at the same temperatures.

Phenomenologically, the Hall resistivity is defined by $\rho_H = R_o\boldsymbol{B} + R_s 4\pi \boldsymbol{M}$, where R_o is the ordinary Hall coefficient, R_s is the extraordinary Hall effect coefficient (also called the anomalous Hall effect coefficient), $\boldsymbol{B}$ is the applied magnetic induction, $\boldsymbol{M}$ is the spontaneous magnetization. In $Tm_2Fe_{14}B$, R_s is much larger than R_o. Therefore, we obtain the values of the SHE coefficient from $R_s = \rho_H/4\pi M$, where ρ_H is the low-field (0.1 T) Hall resistivity and M is the value of the magnetization at the same field. This is how we avoid effects that are related to the shape and domain structure of the sample.

The temperature variation of R_s is shown in figure 4 for the two measurement configurations. There is a small abrupt drop in R_s at T_s in configuration 1, but no anomaly is observed in this region in configuration 2. R_s exhibits a large drop near the Curie temperature in both configurations. Kondo and Maranzana have shown that the anomalous Hall resistivity, arising from the interaction between localized spins and the orbital momentum of conduction electrons, is proportional to the third moment of the magnetization fluctuations: $\rho_H \propto \langle (M - \langle M \rangle)^3 \rangle$ [16, 17]. We therefore expect no large variations of R_s in the vicinity of a first-order phase transition as magnetic fluctuations are then finite, except at the transition point. This is how R_s

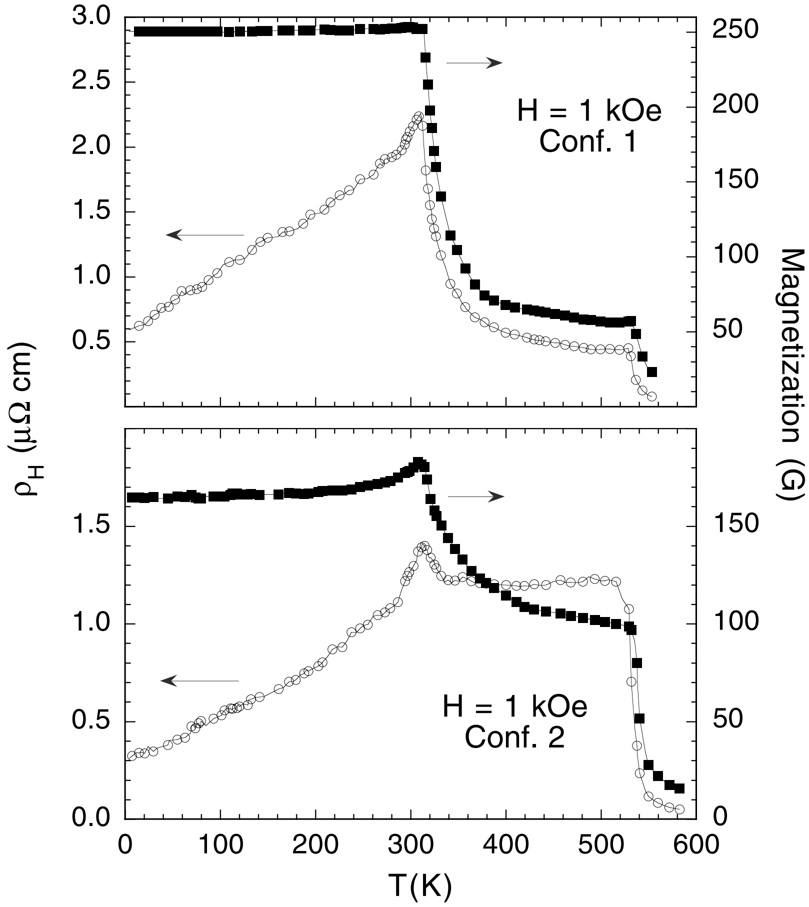

Figure 3. Hall resistivity and magnetization at $H = 0.1$ T, as functions of temperature, for $Tm_2Fe_{14}B$. Data for the two experimental configurations are shown.

behaves in our experiments. On the other hand, in the vicinity of T_c, the longitudinal magnetic fluctuations are critical and give a large contribution to R_s.

We have numerically calculated the magnetization in $Tm_2Fe_{14}B$, following a model outlined in reference [10]. Using values of parameters given therein and a phenomenological mean-field approximation [13], we obtained the correct value of T_s and reproduce the behaviour of R_s through the SRT and at T_c. The calculated third moment of the magnetization fluctuations as a function of temperature is shown in the inset of figure 4 for the two configurations studied. The agreement with the experiment is satisfactory. However, lack of knowledge of the temperature dependence of the thulium anisotropy parameters makes these calculations somewhat unreliable.

Away from the anomalous regions, $R_s \propto \rho$ for 60 K $\leqslant T \leqslant$ 170 K and $R_s \propto \rho^2$ for $T >$ 170 K and $T \leqslant$ 60 K. This is shown in figure 5. A simple relation is commonly satisfied between the SHE and the longitudinal resistivity: $R_s = a\rho + b\rho^2$, where the first term stands for the skew component, and the second term gives the side-jump contribution to the anomalous Hall coefficient [18–20]. Both of these mechanisms are brought about by spin–orbit interactions. Dilute impurities and spin disorder are expected to give $R_s \propto \rho$,

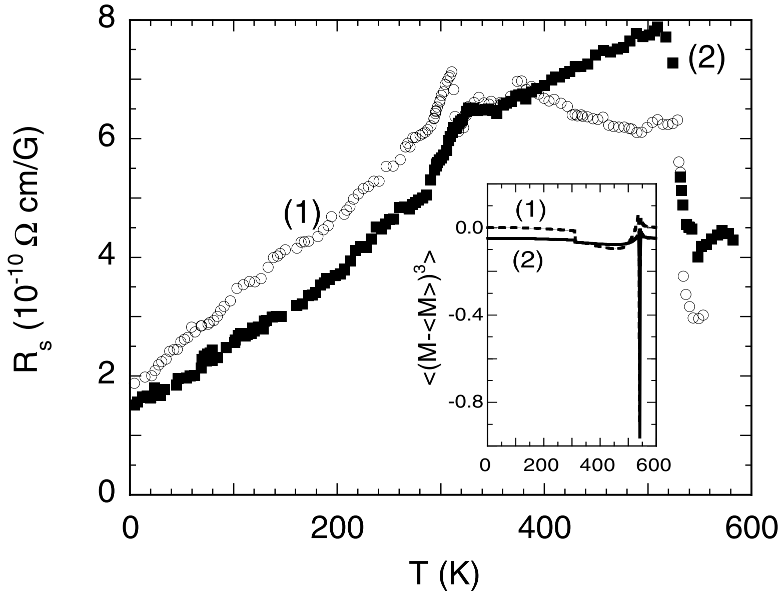

Figure 4. Anomalous Hall coefficient, $R_s = \rho_H/4\pi M$, as a function of temperature for $Tm_2Fe_{14}B$ single crystals in the two experimental configurations. The inset shows the calculated $\langle(M - \langle M\rangle)^3\rangle$ as a function of temperature.

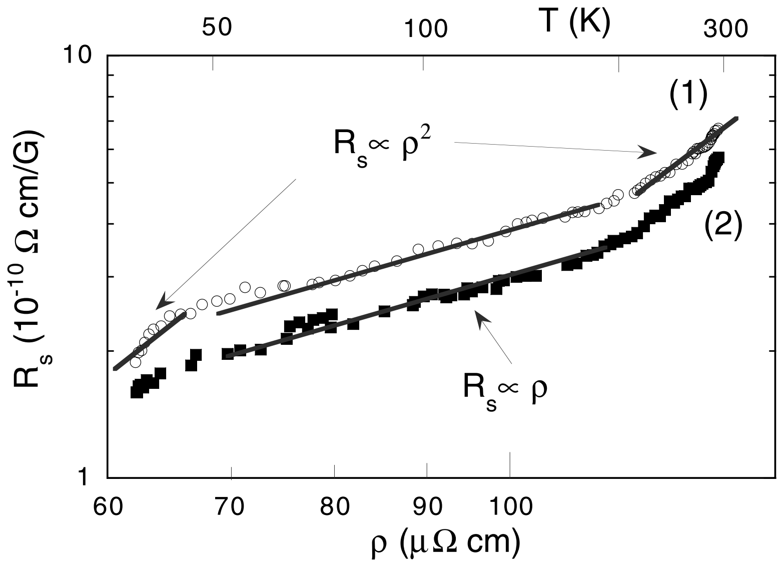

Figure 5. Anomalous Hall coefficient R_s as a function of total longitudinal resistivity for $Tm_2Fe_{14}B$ single crystals in the two experimental configurations. The solid lines are fits to experimental points.

whereas phonons and concentrated spin defects lead to $R_s \propto \rho^2$. As expected, the latter is more important at high temperatures for the sample that we have studied. However, the ρ^2-dependence is also observed at low temperatures, probably arising from a high impurity concentration.

The field dependence of the magnetoresistance (MR) of $Tm_2Fe_{14}B$ is shown in the inset of figure 2 for various temperatures. At 5 and 100 K, a positive transverse magnetoresistance of about 0.4% is observed. The magnitude of this effect is too large to be explained by the classical magnetoresistance mechanisms (assuming an electron mobility of about 5 cm^2 V^{-1} s^{-1}). Furthermore, the observed MR(H) dependence (H is the applied magnetic field) does not agree with the classical H^2-behaviour. Since the observed MR seems to saturate with magnetic field quite rapidly, we attribute most of the variation of ρ with H to scattering mechanisms that depend on the magnetization (such as skew or side-jump scattering). The magnetoresistance of $Tm_2Fe_{14}B$ turns slightly negative at room temperature. This effect can come from the suppression of spin fluctuations by an applied magnetic field, as scattering by spin fluctuations is more important at higher temperatures.

4. Concluding remarks

We have measured the resistivity and Hall effect of a $Tm_2Fe_{14}B$ single crystal as a function of temperature and magnetic field. A jump in resistivity at T_s indicates that the spin-reorientation transition in this compound is of first order. Analysis of the Hall effect data leads to the same conclusion. There is a small difference in the behaviour of the SHE through the SRT in the two configurations studied. A small drop in R_s is observed for the magnetic field almost aligned with the easy-axis direction while no anomaly is evident for the magnetic field almost perpendicular to the easy-axis directions. Unfortunately, the crystallographic orientation of our sample prevents us from observing this effect more clearly.

A combination of skew and side-jump scattering accounts for our SHE data reasonably well. Side-jump scattering dominates at high temperatures while skew scattering is more important at intermediate temperatures. However, the observed sharp drop of the SHE at T_c is produced by a singularity in the skew scattering as longitudinal spin fluctuations become large in this region.

A few remarks about the behaviour of the resistivity follow. ρ is limited by impurity and magnon scattering at low temperatures ($T \leqslant 40$ K). The magnetic resistivity is proportional to T^2 in this temperature range. As the temperature increases, phonon scattering becomes important, in addition to spin disorder. The anomaly in ρ observed just below T_c is probably produced by an Invar-type coupling which leads to the strong lattice softening in this region [9]. Above T_c, all spins are disordered and the magnetic resistivity consequently saturates. However, since electrons also scatter against phonons, the total resistivity increases linearly with temperature.

Acknowledgment

This work was supported by Project MAT 99/1142 of Comisión Interministerial de Ciencia y Tecnología (CICYT).

References

[1] Buschow K H J 1988 *Ferromagnetic Materials, a Handbook on the Properties of Magnetically Ordered Substances* vol 4, ed E P Wohlfarth and K H J Buschow (Amsterdam: North-Holland) p 1
Herbst J F 1991 *Rev. Mod. Phys.* **63** 819 and references therein
Hummler K and Fähnle M 1996 *Phys. Rev.* B **53** 3290

[2] Bland J A C and Heinrich B (ed) 1994 *Ultrathin Magnetic Structures I* (Berlin: Springer)
Gradmann U 1993 *Handbook of Magnetic Materials* vol 7, ed K H J Buschow (Amsterdam: North-Holland) ch 1

[3] Hirosawa S, Matsuura Y, Yamamoto H, Fujimura S, Sagawa M and Yamauchi H 1986 *J. Appl. Phys.* **59** 873
[4] Yamada M, Yamaguchi Y, Kato H, Yamamoto H, Nakagawa Y, Hirosawa S and Sagawa M 1985 *Solid State Commun.* **56** 663
[5] Price D C, Day R K and Dunlop J B 1986 *J. Appl. Phys.* **59** 3585
[6] Davis R L, Day R K and Dunlop J B 1985 *Solid State Commun.* **56** 181
[7] Lazaro F J, Bartolomé J, Navarro R, Rillo C, Lera F, Garcia L M, Chaboy J, Pique C, Burriel R, Fruchart D and Miraglia S 1990 *J. Magn. Magn. Mater.* **83** 289
[8] Luis F, Infante P, Bartolomé J, Burriel R, Piqué C, Ibarra R and Buschow K H J 1995 *J. Magn. Magn. Mater.* **140–144** 1045
[9] Fujii H, Nagata H, Uwatoko Y, Okamoto T, Yamamoto H and Sagawa M 1987 *J. Magn. Magn. Mater.* **70** 331
[10] Yamada M, Kato H, Yamamoto H and Nakagawa Y 1988 *Phys. Rev.* B **38** 620
[11] Sousa J B, Amado M M, Pinto R P, Salgueiro M A, Braga M E and Buschow K H J 1991 *J. Phys.: Condens. Matter* **3** 4119
Stankiewicz J and Bartolomé J 1999 *Phys. Rev.* B **59** 1152
[12] Amado M M, Pinto R P, Braga M E, Rogalski M S and Sousa J B 1997 *J. Appl. Phys.* **81** 5784
[13] Stankiewicz J and Bartolomé J 1999 *Phys. Rev. Lett.* **83** 2026
[14] Stankiewicz J and Bartolomé J 1997 *Phys. Rev.* B **55** 3058
[15] Sousa J B, Montenegro J F D, Moreira J M and Braga M E 1982 *J. Phys. F: Met. Phys.* **12** 351
[16] Kondo J 1962 *Prog. Theor. Phys. (Japan)* **27** 772
[17] Maranzana F E 1967 *Phys. Rev.* **160** 421
[18] Luttinger J M 1958 *Phys. Rev.* **112** 739
[19] Smit J 1955 *Physica* **21** 877
[20] Berger L 1969 *Phys. Rev.* **177** 790