Magnetotransport in Tb_2Fe_{17} single crystals

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Abstract. We have performed measurements of the Hall effect, electrical resistivity, and magnetization on Tb_2Fe_{17} single crystals in the 5 to 300 K temperature range, and in magnetic fields of up to 5 T. We find that the anomalous Hall effect of this ferromagnet depends strongly on the magnetization direction relative to the crystal axes. The AHE resistivity, measured with an applied magnetic field $\mathbf{H}$ perpendicular to the c-axis, is very large and varies linearly with the longitudinal resistivity ρ, whereas the AHE resistivity for $\mathbf{H}$ along the hard magnetization direction is much smaller and increases as ρ^2. For the latter configuration, the electrical resistivity shows a sharp decrease at a field-induced first-order magnetization process (FOMP) which is observed in $H \sim 2.7$ T up to a temperature of 250 K.

1. Introduction

The Tb_2Fe_{17} intermetallic compound crystallizes in a Th_2Ni_{17}-type hexagonal structure [1] in which the Fe layers (perpendicular to the c-axis) are intercalated with dumb-bell Fe pairs along the c-axis. The Fe atoms occupy four nonequivalent crystallographic sites. However, only dumb-bell Fe atoms at $4f$ sites have a non-zero orbital magnetic moment. The Mössbauer effect experiments [2] show that the $3d$ bands coming from the $4f$ ions are distorted giving rise to a larger magnetic polarization and to a lower electron charge density. This agrees with results of band-structure calculations which find a highest orbital moment for Fe atoms at the dumbbell sites [3-4]. The Tb sublattice couples antiferromagnetically to the Fe sublattice in the Tb_2Fe_{17} alloy. A first-order magnetization process (FOMP) is observed at a magnetic field of about 2.5 T applied along the sixfold axis in this compound [5]. The FOMP does not affect the mutual orientation of the sublattices only their relative orientation to the crystallographic axes. Interestingly, the critical field for this transition is nearly independent of temperature up to ≈ 250 K even though the anisotropy constants vary with temperature as expected from existing models [6].

All these properties make Tb_2Fe_{17} compounds attractive for electron–transport studies which are very sensitive to the magnetic nature of the materials. In particular, intermetallic compounds with transition metals may show complex behavior as $3d$ itinerant magnetism and $4f$ localized magnetic moments compete in them. In addition, spin-orbit coupling, either intrinsic or extrinsic, of magnetic electrons gives rise to a magnetization-dependent Hall effect. This, in turn, depends crucially on scattering mechanisms [7, 8] and on the band structure [9]. However, no electronic transport studies have been reported for Tb_2Fe_{17}. Our aim is to fill this gap.

In this paper we report results of electrical resistivity and Hall effect measurements on Tb_2Fe_{17} single crystals in the temperature range of 5–300K and in magnetic fields of up to 6 T. The measurements have been performed for various magnetic field orientations with respect to the easy-magnetization axis. The anomalous Hall effect (AHE), both extrinsic and intrinsic, is very anisotropic in this ferromagnet. We attribute it to the band structure details and to a possible distortion of the $3d$ bands.

2. Experiment

Tb_2Fe_{17} single crystals were grown by inductive melting of a mixture of 99.9%–pure Tb and 99.99%–pure Fe in alundum crucibles under Ar atmosphere. The mixture was rapidly heated up to the melting point (T $\approx$ 1500 C), and subsequently cooled down at a rate of 40-60 C/min. Further heating on to approximately 1200 C, with an annealing time of approximately 20 hours, was performed in order to obtain large crystalline grains. The phase composition was checked by optical metallography. We carefully oriented the samples using x-ray back Laue diffraction. The magnetotransport measurements were performed on Hall bar shaped samples. The typical sample size was $0.5 \times 1 \times 5$ mm^3. We used a six probe method in order to check for samples' homogeneity both in resistivity and Hall coefficient. The Hall resistivity and magnetization were measured in a magnetic field of up to 6 T, in a temperature range from 5 to 300 K. Magnetization measurements were carried out in a superconducting quantum interference device (SQUID) magnetometer on the very same samples that were used in the magnetotransport studies. In this way, we expected to avoid sample–shape and domain dependent effects when comparing results of different experiments.

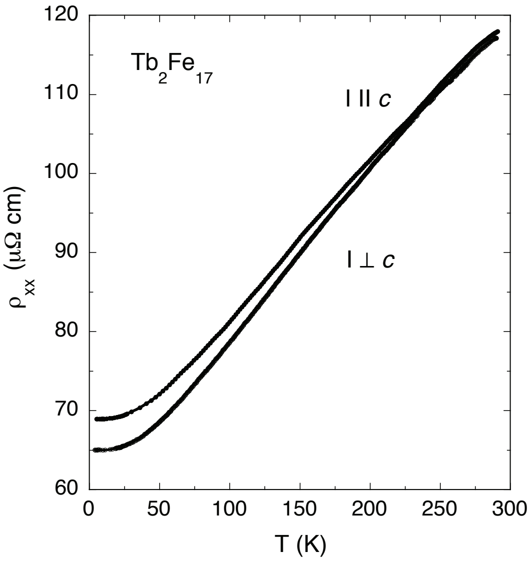

Figure 1. Longitudinal resistivity as a function of temperature for Tb_2Fe_{17} single crystals. The solid lines are guides to the eye.

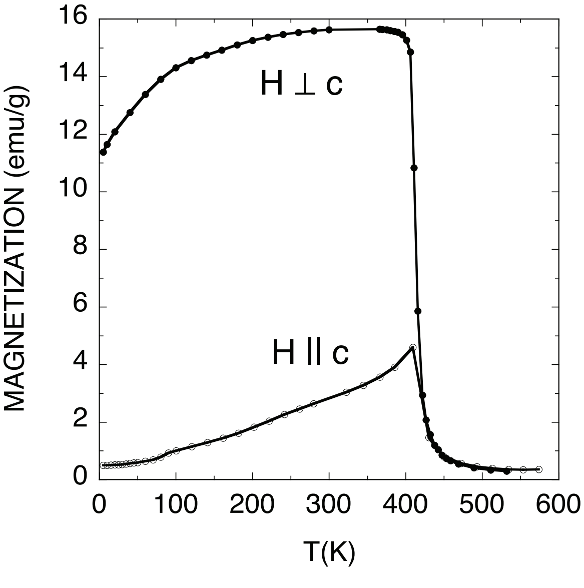

Figure 2. Magnetization measured in the field of 1 kOe as a function of temperature for the same Tb_2Fe_{17} single crystals. The solid lines are guides to the eye.

3. Results and Discussion

Figure 1 shows how the zero-field longitudinal resistivity $\rho_{xx}(T)$ depends on temperature T in Tb_2Fe_{17} single crystals. The resistivity, typical of metallic ferromagnet, is slightly anisotropic at low temperatures: it is approximately 6% lower for the current flowing in the easy plane than for the current along the c–axis. It increases with increasing temperature as spin–disorder and electron–phonon scattering become more important. The low-field magnetization variation with temperature and orientation is shown in Fig. 2. The magnetization M drops sharply at the Curie temperature of 411 K. How the electrical resistivity varies with magnetic field is shown in Figs. 3 and 4. Since an external magnetic field suppresses spin fluctuations, the electrical resistivity decreases when a magnetic field is applied. The observed negative magnetoresistance is very small (less than 3 per mil at $H = 5$ T) for magnetic fields on the easy plane. However, an appreciable jump is seen in $\rho_{xx}(H)$ at a field-induced first-order magnetization process (FOMP) for $\mathbf{H} \parallel c$–axis, as shown in Fig. 3. At 5 K, $\rho(H)$ drops approximately 3% at a field of ~ 2.8 T in this configuration. Such discontinuity is expected at the critical field of a first-order spin-reorientation transition, since the resistivity depends on the angle between $\mathbf{I}$ and the spontaneous magnetization $\mathbf{M}_s$ through spin–orbit coupling. As the temperature increases, the resistivity drop decreases. It becomes negligible for $T \gtrsim 250$ K. The critical field at which the abrupt resistivity change occurs is nearly independent of temperature. This behavior of the critical field is however incidental since the anisotropy constants vary with temperature as expected for intermetallic compounds [6].

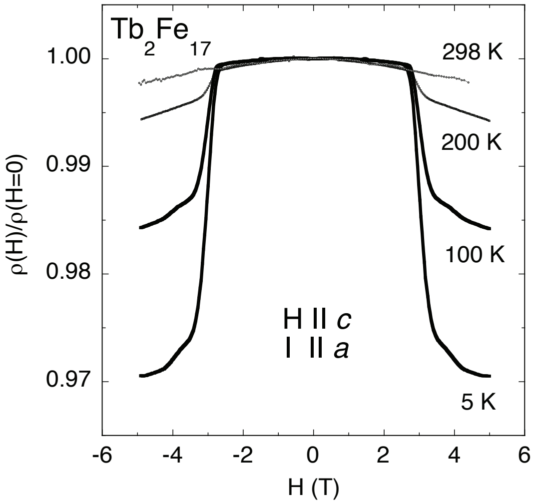

Figure 3. Transverse magnetoresistance in a Tb$_2$Fe$_{17}$ single crystal for $\mathbf{H}$ along the c–axis and $\mathbf{I}$ along the a–axis.

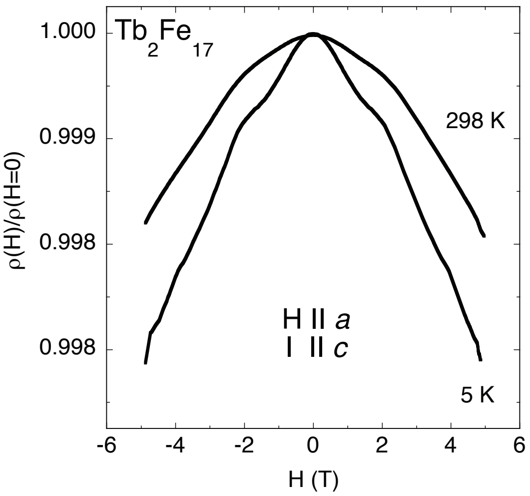

Figure 4. Transverse magnetoresistance in a Tb$_2$Fe$_{17}$ single crystal for $\mathbf{H}$ along the a–axis and $\mathbf{I}$ along the c–axis.

We next discuss the Hall resistivity ρ_{xy} and magnetization data. Their variation with magnetic field are shown in Figs. 5 and 6 for T=5 and 300 K, respectively. Below technical saturation, ρ_{xy} follows the magnetization behavior of the sample. Above saturating field, ρ_{xy} remains almost constant since the negative, ordinary Hall resistivity is very small. The experimental values of ρ_{xy} and M_s are obtained by extrapolating the Hall resistivity and magnetization data from high magnetic fields back to $H = 0$. Since relatively small magnetic fields are sufficient to align magnetic moments, this procedure is quite straightforward. In ferromagnetic metals, the anomalous contribution to ρ_{xy} is usually proportional to the spontaneous magnetization because of spin-orbit interaction. Extrinsic mechanisms such as skew and side–jump scattering contribute terms linear [7] and quadratic [8] in the longitudinal resistivity, respectively. On the other hand, spin-orbit coupling of band electrons brings about intrinsic AHE which is independent of scattering and should give $\rho_{xy} \propto \rho_{xx}^2$ [10].

We plot the Hall resistivity vs the total longitudinal resistivity using our data in Fig. 7. The AHE resistivity varies quite linearly with ρ_{xx} for $\mathbf{M}$ lying in the ab plane. However, it

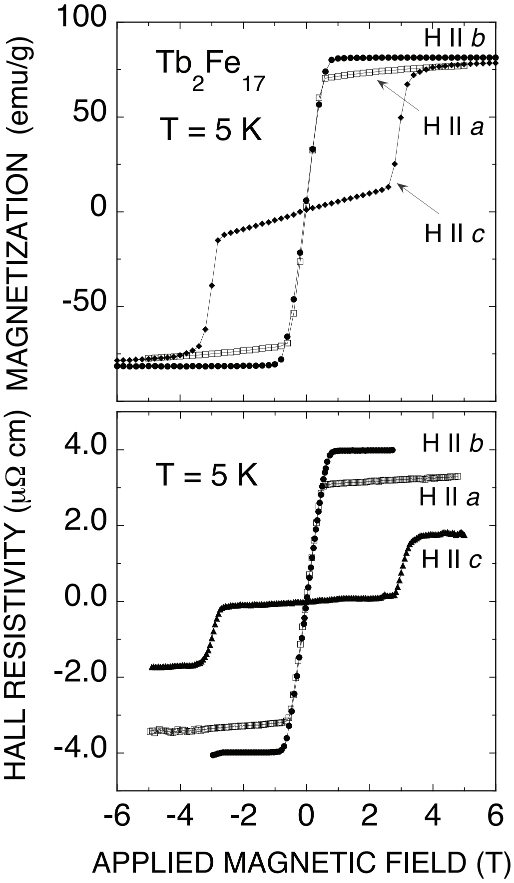

Figure 5. Magnetization and Hall resistivity as a function of magnetic field for Tb_2Fe_{17} single crystals at 5 K. The solid line is a guide to the eye.

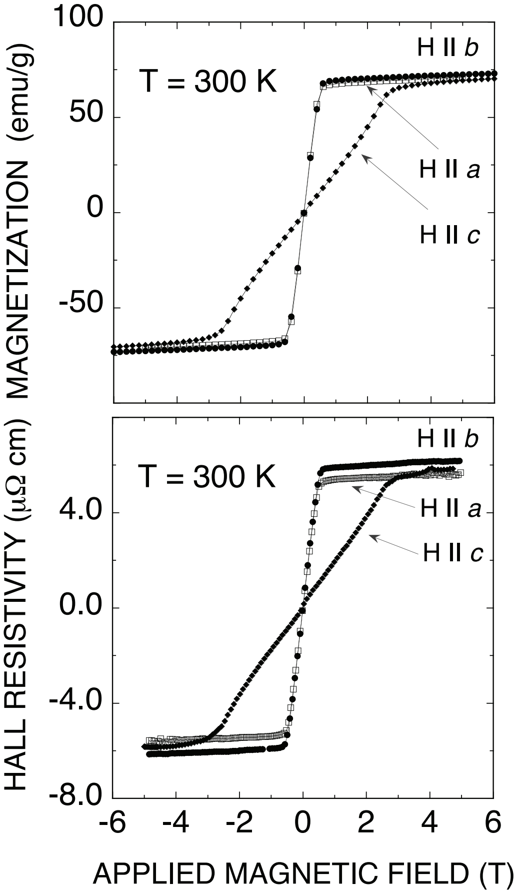

Figure 6. Magnetization and Hall resistivity as a function of magnetic field for Tb_2Fe_{17} single crystals at 300 K. The solid line is a guide to the eye.

varies as a ρ_{xx}^2 for $\mathbf{M}$ along the hard direction. In addition, the AHE depends strongly on orientation: it drops by a factor of ~ 3 as the magnetization is tilted from the easy ab plane to the c–axis at low temperatures. To interpret our data, consider the relation between the AHE and the longitudinal resistivity of the form: $\rho_{xy} = a(M_s)\rho_{xx} + b(M_s)\rho_{xx}^2$ [11-12]. The first term arises from skew scattering contribution that is usually linear in magnetization [7]. Assuming $a(M) \propto M$, and plotting $\rho_{xy}/M_s\rho_{xx}$ versus ρ_{xx} we can estimate a. The second term stands for the intrinsic contribution, related to Berry phase effects on conduction electrons. We consider side-jump scattering, which has identical ρ_{xx} dependence, negligible in Tb_2Fe_{17} on the basis of the observed orientation dependence of the AHE [13]. Such dependence is not expected for extrinsic side-jump mechanism which should lead to an isotropic Hall current [8]. The intrinsic Hall conductivity (AHC) is expressed as $\sigma_{xy}^a = \rho_{xy}/\rho_{xx}^2$, therefore $b(M) = \sigma_{xy}^a$. Since the AHC varies only weakly with temperature [12], a constant low–temperature value of σ_{xy}^a follows. Indeed, the plot of $\rho_{xy}/M_s\rho_{xx}$ vs ρ_{xx} is linear for $\mathbf{H}\|\ c$ configuration at low temperatures (see Fig. 8), hence $\sigma_{xy}^a \propto M$ in Tb_2Fe_{17}.

The variation of $[\rho_{xy}/\rho_{xx}][M_s^o/M_s(T)]$ with ρ_{xx} in Tb_2Fe_{17} single crystals is shown in Fig. 8. Here, $M_s^o = M_s(T = 0)$. For $\mathbf{M}$ on the easy plane, this quantity is constant for $T \lesssim 250$ K. However, when $\mathbf{M}$ is lined up along the hard direction, $\rho_{xy}M_s^o/\rho_{xx}M_s(T)$ varies linearly with

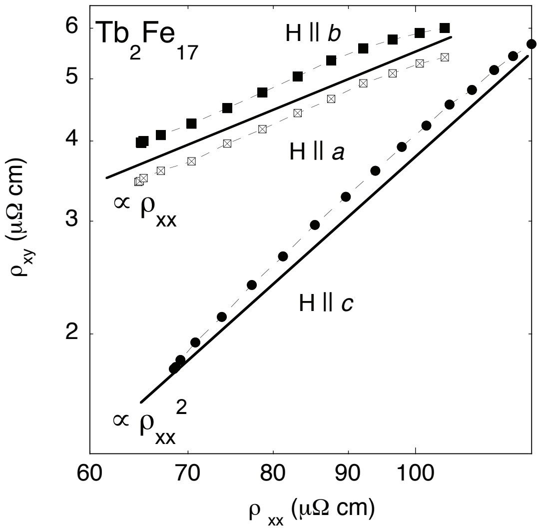

Figure 7. Hall resistivity of Tb$_2$Fe$_{17}$ single crystals as a function of the longitudinal resistivity for various configurations. The solid lines show a linear and a quadratic in ρ_{xx} variation.

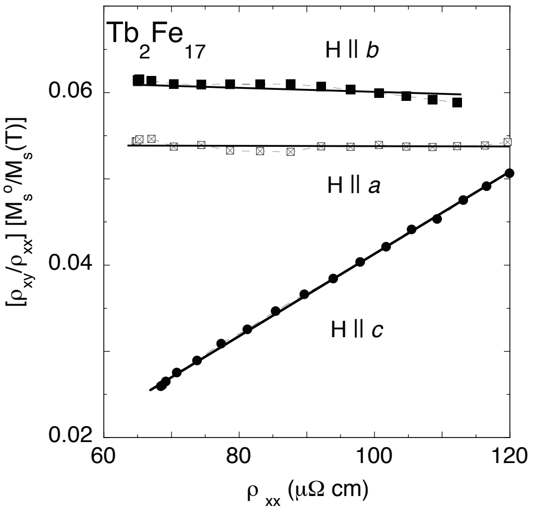

Figure 8. Plots of $[\rho_{xy}/\rho_{xx}][M_s^o/M_s(T)]$ as a function of the longitudinal resistivity for $\mathbf{H}\perp$ c-axis and $\mathbf{H}\|$ c-axis in Tb$_2$Fe$_{17}$ single crystals. The solid lines are linear fittings to the data points.

ρ_{xx}. This clearly suggests that large skew scattering entirely dominates the AHE resistivity for magnetic fields on the easy plane, but it has almost no effect otherwise. We find a value of approximately 0.055 for the skew–scattering coefficient a in Tb$_2$Fe$_{17}$ when $\mathbf{M}$ lies on the easy plane. It is one order of magnitude larger than skew–scattering contributions to the AHE found for metallic ferromagnets [11]. This might be related to the distortion of d–bands corresponding to the dumb-bell Fe sites. However, a detailed knowledge of the impurity potential would be needed for a quantitative estimation of skew-scattering contributions. On the other hand, we find a value of 420 $\Omega^{-1}\text{cm}^{-1}$ for σ_{xy}^{a} which is very close to theoretical estimations for the intrinsic Hall conductivity [14,15]. This agrees with the recent observations that the intrinsic mechanism is important for the AHE in moderately conductive ferromagnets [9]. In particular, variations of the total Berry curvature, coming from avoided crossings of nearly degenerate d–levels of Fe ions states near the Fermi energy, may resonantly enhance the AHC amplitude. Such scenario also explains the orientation dependence of the intrinsic AHC [13]. Band structure calculations for Y$_2$Fe$_{17}$ [16], which should be not very different from those for Tb$_2$Fe$_{17}$, seem to give ground for the above conclusions. However, we cannot quite rule out that the large intrinsic AHC, observed in the system we study, follows from inter-band hopping between nearly degenerate d orbitals [15].

Acknowledgments

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References

[1] Givord D, Lemaire R, Moreau J M and Roudaut E 1972 *J. Less Common Met.* **29** 361
[2] Gubbens P C M, van Loef J J and Buschow K H J 1974 *J. Phys. (Paris) Colloq.* **35** C6-617
[3] Cochoorn R 1989 *Phys. Rev.* B **39**, 13072

[4] Sabiryanov R F and Jaswal S S 1998 *Phys. Rev.* B **57** 7767
[5] Verhoef R, Quang P H, Radwański R J, Marquina C and Franse J J M 1992 *J. Magn. Magn. Mater.* **104-107** 1473
[6] Kuz'min M D, Skourski Y, Skokov K P, Müller K -H and Gutfleisch O 2008 *Phys. Rev.* B **77** 132411
[7] Smit J 1955 *Physica (Amsterdam)* **21** 877
[8] Berger L 1970 *Phys. Rev.* B **2** 4559
[9] Nagaosa N, Sinova J, Onoda S, MacDonald A H and Ong N P 2010 *Rev. Mod. Phys.* **82** 1539
[10] Yao Y, Kleinman L, MacDonald A H, J. Sinova J, Jungwirth T, Wang D-S, Wang E and Niu Q 2004 *Phys. Rev. Lett.* **92** 037204
[11] Sales B C, Jin R and Mandrus D 2008 *Phys. Rev* B **77** 024409
[12] Zeng C, Yao Y, Niu Q and Weitering H 2006 *Phys. Rev. Lett.* **96** 037204
[13] Roman E, Mokrousov Y and Souza I 2009 *Phys. Rev. Lett.* **103** 097203
[14] Miyasato T, Abe N, Fujii T, Asamitsu A, Onoda S, Onose Y, Nagaosa N and Tokura Y 2007 *Phys. Rev. Lett* **99** 086602
[15] Kontani H, Tanaka T and Yamada K 2007 *Phys. Rev.* B **75** 184416
[16] Kulakowski K and del Moral A 1994 *Phys. Rev.* B **50** 234