Anomalous Hall effect in $Y_2Fe_{17-x}Co_x$ single crystals

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We study experimentally the Hall resistivity of $Y_2Fe_{17-x}Co_x$ single crystals with $x \leq 4$ for wide temperature and applied magnetic field ranges and for various magnetic field orientations with respect to the easy-magnetization axis. For small x, the anomalous Hall effect (AHE) is very anisotropic in these naturally layered compounds. For $x \leq 2$, the AHE resistivity, measured with an applied magnetic field $H \perp c$ axis, is nearly 1 order of magnitude larger than the AHE resistivity for H along the hard-magnetization direction ($H \| c$ axis). Furthermore, the former is very large and varies linearly with the longitudinal resistivity ρ, whereas the latter increases as ρ^2. The behavior of the AHE for $H \| c$ axis comes quite likely from the intrinsic effect related to the hopping between different Fe d orbitals. Such hopping is allowed for high-symmetry points at the crystallographic dumbbell sites in this configuration. On the other hand, interorbital hopping is not allowed for $H \perp c$ axis. However, a huge amplitude of the AHE scattering for this configuration, which follows from skew scattering, is puzzling. Both the AHE anisotropy and the large skew scattering vanish for sufficiently high Co content. We attribute this to variations in the electronic structure of the $Y_2Fe_{17-x}Co_x$ system when Co atoms start to occupy the dumbbell crystallographic sites.

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I. INTRODUCTION

Electronic transport studies can reveal interesting features in magnetic materials. In particular, the Hall effect contributes an anomalous term, proportional to the magnetization of the material, in addition to the ordinary term which arises from the Lorentz force. The spontaneous (or anomalous) contribution can be brought about by asymmetric scattering of current carriers which are subject to spin-orbit interactions.[1] It is generally accepted that this extrinsic mechanism for the anomalous Hall effect (AHE) involves skew scattering[2] and side-jump scattering.[3] In the past decade, much attention has been paid to the intrinsic AHE, arising from the "anomalous velocity" acquired by current carriers in a ferromagnet.[4] The intrinsic mechanism is usually interpreted in terms of Berry-phase effects on conduction electrons. Such models have been used to calculate the intrinsic anomalous Hall conductivity (AHC) in ferromagnetic semiconductors,[5] transition metals,[6] and oxides.[7] Recently, it has been proposed that band crossing, close to the Fermi level, can resonantly enhance the Berry-phase curvature and the AHC.[8,9] On the other hand, as has been shown in Ref. 10, large intrinsic AHC may follow from interband hopping between nearly degenerate d orbitals in ferromagnetic transition metals, with no need for a special model of the band structure. Therefore, despite intense theoretical efforts made for well over a decade, the physical mechanisms behind the AHE remain imperfectly understood and their importance has not been clearly established.

The number of experimental reports on the intrinsic AHE in magnetic semiconductors,[11] ferromagnetic thin films,[12,13] chalcogenide spinels,[14] oxides,[7] and other ferromagnetic compounds[15] keeps growing. However, the separation between the intrinsic and extrinsic AHEs from experiment is frequently ambiguous. On the other hand, the skew-scattering contribution to the AHE resistivity ρ_{xy} is proportional to the longitudinal resistivity ρ; the extrinsic side-jump scattering and the intrinsic mechanism lead to the same relation: $\rho_{xy} \propto \rho^2$. In addition, the ordinary Hall effect, which is linear in the applied magnetic field, can just as well be an appreciable part of the measured signal. Consequently, measurements of the Hall resistivity in a wide range of temperatures and magnetic fields are required for a reliable interpretation.

In order to elucidate the origin of the AHE, its studies should preferably be carried out on magnetic materials whose band structure is relatively well known. The $Y_2Fe_{17-x}Co_x$ system seems to be a good candidate as its structural and magnetic properties have been the subject of many studies. Y_2Fe_{17} crystallizes in a Th_2Ni_{17}-type hexagonal structure.[16] This compound is a natural multilayer system in which the Fe layers (perpendicular to the c axis) are intercalated with dumbbell Fe pairs. Fe atoms occupy four nonequivalent sites in the unit cell. Only the dumbbell atoms have a sufficiently high symmetry to contribute to the orbital magnetic moment. Y_2Fe_{17} is a weak ferromagnet below ≈ 320 K. When Co is substituted for Fe in $Y_2Fe_{17-x}Co_x$ alloys, the easy-plane anisotropy prevails up to $x \cong 8$; for larger x it changes to an easy axis anisotropy. There is experimental evidence for preferential site substitution of Fe by Co: Co atoms hardly substitute Fe at the dumbbell sites up to a Co content of 40%.[17] The Curie temperature increases nonlinearly with x.[18] Results of self-consistent *ab initio* band-structure calculations for Y_2Fe_{17} have been reported in several papers.[19]

In this paper we report results of Hall-effect and magnetization measurements in $Y_2Fe_{17-x}Co_x$ single crystals for wide temperature and applied magnetic field ranges and for

various magnetic field orientations with respect to the easy-magnetization axis. We find a large AHE anisotropy in this system for $x \leq 2$. The low-field AHE resistivity, measured with an applied magnetic field $H \perp c$ axis, is nearly 1 order of magnitude larger than the one for H along the hard-magnetization direction ($H \| c$ axis). Furthermore, the former is very large and linear in ρ whereas the latter follows ρ^2 for $T \lesssim 150$ K. We tentatively interpret the behavior of ρ_{xy} for $H \| c$ axis in terms of the intrinsic effect related to the interorbital hopping between degenerate d orbitals. Such hopping is allowed for high-symmetry points at the dumbbell sites in this configuration. On the other hand, interorbital hopping is not allowed for $H \perp c$ axis. However, a huge amplitude of the AHE resistivity for this configuration, which follows skew-scattering behavior, is puzzling. Such gigantic skew scattering may well arise from resonant scattering of itinerant electrons through the virtual bound states formed by the hybridization of Fe d states with s-p conduction states.

II. EXPERIMENT

$Y_2Fe_{17-x}Co_x$ single crystals were grown using inductive melting of the initial pure components in alundum crucibles under Ar atmosphere. The initial mixture was rapidly heated up to the melting point, cooled down at a rate of 50–70 K/min, and subsequently heated further to approximately 1180 °C and held at this temperature for 20 h in order to obtain large crystalline grains. We carefully oriented samples using x-ray back Laue diffraction. The electrical resistivity and Hall-effect measurements were performed with a six-probe method on bar-shaped samples with typical size of $0.5 \times 2 \times 7$ mm^3. We measured the Hall resistivity as a function of magnetic field up to 9 T in a temperature range of 5–300 K and up to 1 T for $300 < T < 800$ K. Magnetization measurements were performed with a superconducting quantum interference device (SQUID) magnetometer on the same samples that were used in magnetotransport studies. In this way, we expect to avoid domain and sample-shape related effects when comparing results of different experiments. These measurements were performed in magnetic fields of up to 9 T and in the 5–800 K temperature range.

III. RESULTS AND DISCUSSION

We first discuss briefly the results from electrical resistivity measurements. Figure 1 shows how the electrical resistivity depends on temperature T in $Y_2Fe_{17-x}Co_x$ single crystals ($x \leq 4$). The observed behavior is typical of a ferromagnet in which phonon and spin-disorder scatterings are important. At low temperatures and for $x \leq 2$, ρ is approximately 8% higher along the c axis than in the plane perpendicular to the c axis; this asymmetry disappears in samples with higher Co content. The resistivity decreases slightly in an external magnetic field. At 5 K, the negative magnetoresistance is less than 3% for $H \perp c$ axis and decreases with increasing temperature.

We next turn to the Hall-effect results. Figure 2(a) shows low-field Hall resistivity data as a function of temperature for $x \leq 4$. ρ_{xy} is much larger for the easy-magnetization direction

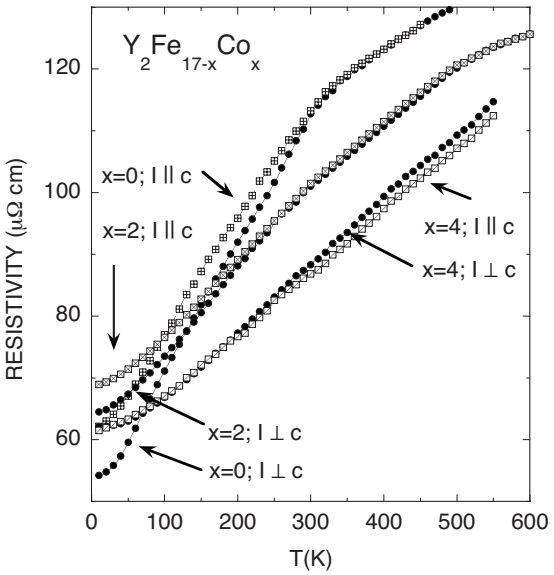

FIG. 1. Temperature dependence of the electrical resistivity in $Y_2Fe_{17-x}Co_x$ single crystals with $x \leq 4$ for two orientations of the electrical current **I** with respect to the c axis. The solid lines are guides for the eyes.

($H \perp c$ axis) than for the hard direction as reported recently for Y_2Fe_{17}.[20] It peaks close to T_c for both orientations. The Hall resistivity follows quite closely the magnetization M of the samples as is shown in Fig. 2(b). Variations in the Hall resistivity and in the magnetization versus magnetic field are shown in Fig. 3 for an $x=0$ alloy. Since the first anisotropy constant is much larger than the second one in $Y_2Fe_{17-x}Co_x$ alloys,[17] the magnetization varies nearly linearly with the magnetic field below reaching saturation. Below the technical saturation, Hall resistivity data, which are holelike, fol-

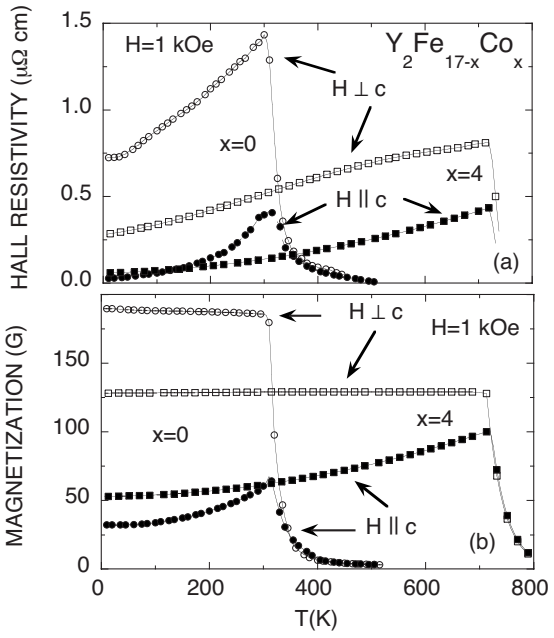

FIG. 2. (a) Low-field Hall resistivity and (b) magnetization as a function of temperature for single crystals of $Y_2Fe_{17-x}Co_x$ for fields applied along and perpendicular to the c axis.

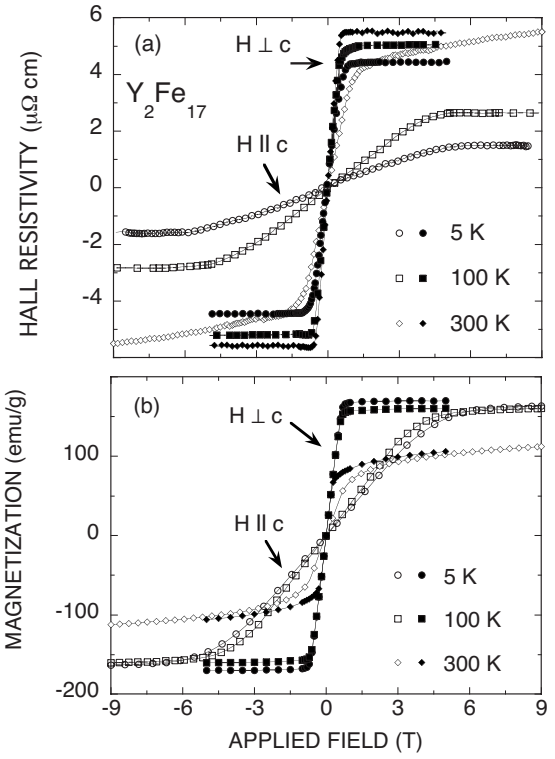

FIG. 3. (a) Hall resistivity and (b) magnetization as a function of magnetic field for a Y_2Fe_{17} single crystal at 5, 100, and 300 K. The solid line is a guide for the eyes.

low the magnetization of the sample: ρ_{xy} increases linearly with the applied magnetic field as M does. Above the saturating field, the Hall resistivity remains nearly constant since the negative ordinary Hall resistivity is very small.

The maximum magnetic field we apply in our experiments (9 T) is too small to saturate the magnetization in the hard direction of the $Y_2Fe_{17-x}Co_x$ system for $x \geq 2$. Therefore, in the discussion which follows below, we consistently calculate the relevant quantities making use of the linear part of the $M(H)$ and $\rho_{xy}(H)$ curves from all the samples. This might raise some concern since the magnetization and the Hall resistivity are sample-shape and domain dependent in this region. In addition, comparison of the results obtained for parallel and perpendicular (to the c axis) applied fields is not straightforward for nonsaturating fields because the magnetization state might differ in both cases. We avoid these effects by relating the values of $\rho_{xy}(H)$ and $M(H)$ measured on the same sample under the same field. Furthermore, the most important quantity we calculate ($\rho_{xy}/M\rho$) depends on $\rho_{xy}(H)$ and $M(H)$ only through their ratio. Thus, our procedure should yield correct results since ρ_{xy} follows the magnetization of the sample. We checked this by using data we obtained for a Y_2Fe_{17} single crystal (Fig. 3) for which we could reach magnetization saturation in both configurations. Figure 4 shows a plot of $\rho_{xy}/M\rho$ vs ρ using both (i) data points from the linear part of $\rho_{xy}(H)$ and $M(H)$ curves and (ii) data points obtained for $\rho_{xy}(H)$ and $M(H)$ above the saturating magnetic field. Clearly, both procedures yield nearly the same results.

Let us assume that the ordinary contribution to the Hall effect is negligible in our samples. ρ_{xy} would then arise entirely from the AHE. Consider the relation between the AHE and the longitudinal resistivity of the form[12,15] $\rho_{xy}=a(M)\rho+b(M)\rho^2$. Coefficients $a(M)$ and $b(M)$ are some function of the magnetization. The first term stands for the skew-scattering contribution that is usually linear in magnetization.[12] Accordingly, by plotting $\rho_{xy}/M\rho$ vs ρ we can obtain $a(M)$. The second term represents the intrinsic contribution; in particular, the anomalous Hall conductivity, $\sigma_{xy}^a=\rho_{xy}/\rho^2$, is given by $b(M)$. How $\rho_{xy}/M\rho$ varies with ρ in the $Y_2Fe_{17-x}Co_x$ single crystals is shown in Fig. 5 for two orientations of the applied magnetic field with respect to the c axis. For H along the easy-magnetization direction ($H \perp c$ axis), $\rho_{xy}/M\rho$ is constant for $\rho \lesssim 95$ $\mu\Omega$ cm (or $T \lesssim 150$ K) in alloys with $x \leq 2$. This implies that skew scattering dominates the AHE for this configuration. Curiously, the magnitude of the AHE is very large: it is almost 1 order of magnitude larger than for other metallic ferromagnets.[15,22]

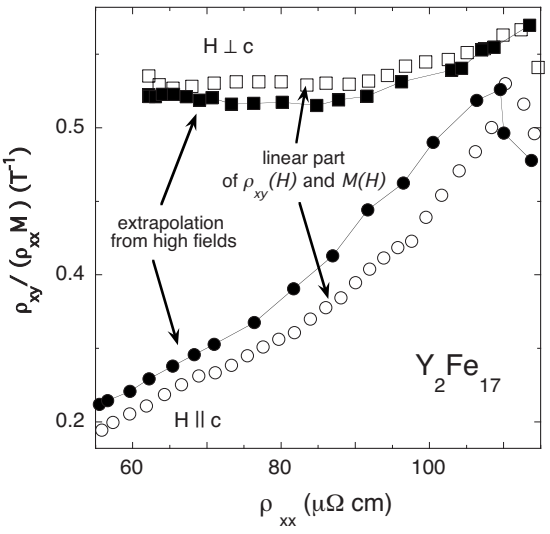

FIG. 4. Plot of $\rho_{xy}/M\rho$ vs ρ for Y_2Fe_{17} single crystals: open points are for data from the linear part of $\rho_{xy}(H)$ and $M(H)$ and closed points for data obtained above the technical saturation. The solid line is a guide for the eyes.

It is difficult to quantify skew-scattering resistivity as details of the scattering potential are needed. In general, asymmetric scattering can come from two mechanisms: (a) the intrinsic $\mathbf{s}\cdot\mathbf{L}$ coupling, while scattering from impurities takes place, between an itinerant electron's intrinsic spin $\mathbf{s}$ and its angular momentum $\mathbf{L}$, and (b) the extrinsic coupling $\mathbf{M}\cdot\mathbf{L}$ between the scattering ion's total magnetic moment $\mathbf{M}$, proportional to the total spin of all the d electrons (partially localized), and the scattered electron's orbital momentum $\mathbf{L}$.[1,21] The former requires unequal spin population for the itinerant electrons to produce asymmetry and is proportional to the impurity concentration. However, we find that the sample with $x=2$, which is less conductive than the $x=0$ sample, shows smaller ρ_{xy}. Then, the extrinsic mechanism is the more likely origin of the AHE in the $H \perp c$ axis configuration because it may occur through extra channels, involving resonant virtual bound states of the Fe ion. We do not know of any other mechanism which could yield such a huge AHE skew resistivity as found for $Y_2Fe_{17-x}Co_x$ alloys.

We now discuss data obtained for H in the hard direction. The plot of $\rho_{xy}/M\rho$ vs ρ, shown in Fig. 5(b), is linear for

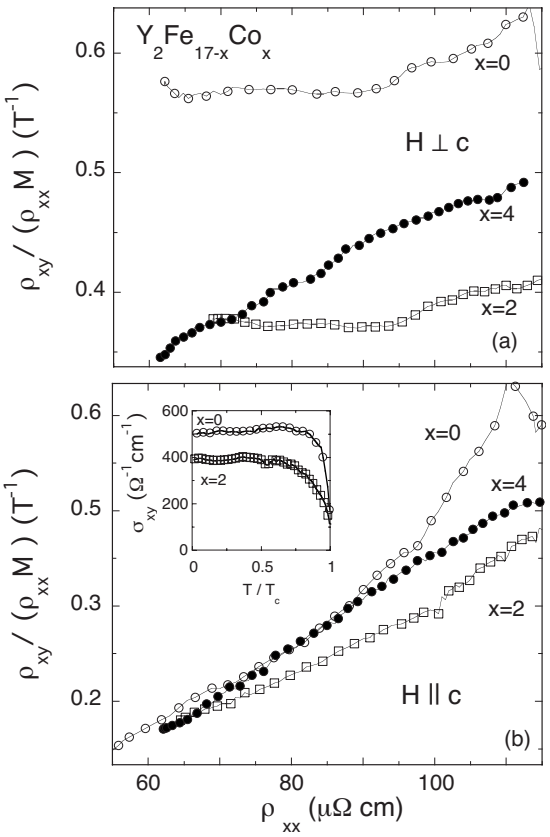

FIG. 5. Plot of $\rho_{xy}/M\rho$ vs ρ for $Y_2Fe_{17-x}Co_x$ single crystals: (a) $H \perp c$ axis and (b) $H \| c$ axis. The inset in (b) shows the temperature variation in the intrinsic AHC.

$\rho \lesssim 100$ $\mu\Omega$ cm for all alloys studied. This suggests that the intrinsic contribution, related to Berry-phase effects on conduction electrons, dominates the AHE. The negative skew-scattering contribution, obtained from a linear fit of $\rho_{xy}/M\rho$ vs ρ, is much smaller than the one found for H applied in the easy plane. In addition, the side-jump scattering contribution, which also gives the quadratic term in the AHE resistivity, is too small to account for the observed effect. We estimate, using expressions for side-jump conductivity derived from several papers,[3,23] that this mechanism accounts for less than 5% of the total value of ρ_{xy} in this configuration. The approximately constant slopes of the curves in Fig. 5(b) show that the intrinsic AHC is proportional to M in a broad temperature interval. We plot the AHC as a function of temperature in the inset of Fig. 5(b). σ_{xy}^a extrapolates to 502 and 395 Ω^{-1} cm^{-1} at 0 K for $x=0$ and 2, respectively. This is very close to theoretical predictions which give for Y_2Fe_{17} $\sigma_{xy}^a \approx e^2/(ha_0)=490$ Ω^{-1} cm^{-1}, where a_0 is the lattice constant.[10,13] It is interesting to note that the side-jump scattering contribution has been shown to be on the order of $(e^2/ha_0)(E_{so}/E_F)$, where E_{so} and E_F are the spin-orbit interaction and Fermi energy, respectively.[24] Since $E_{so}/E_F \lesssim 10^{-2}$ for metallic ferromagnets, it shows that the extrinsic quadratic contribution is quite small as discussed above.

The question arises as to why we observe an intrinsic AHE only in one configuration and extrinsic skew scattering in another one. The main contribution to the intrinsic AHE comes from electronic states corresponding to a few high-symmetry points in the Brillouin zone for which the orbital moment is not quenched. Band-structure calculations for Y_2Fe_{17}, within the tight-binding model with d orbitals, show that d_{xz} and d_{yz} orbitals corresponding to dumbbell Fe ions are coupled through off-diagonal elements of $\mathbf{L}$ for an effective magnetic field parallel to the c axis.[25] In addition, these states are nearly degenerate and lie close enough to the Fermi level so that electrons may hop between them. It has recently been reported that interorbital $(d_{xz}$-$d_{yz})$ hopping can give rise to the large intrinsic AHC in metallic ferromagnets.[10] This would explain the observation of the intrinsic Hall effect for H applied in the hard direction when we force the spontaneous magnetization (or the effective field proportional to it) to align with the c axis, and lack of the intrinsic AHE for H in the easy plane as interorbital hopping is not allowed in such a configuration. On the other hand, skew scattering seems to arise from resonant scattering of s electrons. Its effect will be negligible for electrical current in the easy plane ($H \| c$ axis) since the AHE is proportional to the product $\lambda \mathbf{E} \times \mathbf{M}$. Here, λ is the enhanced spin-orbit parameter and $\mathbf{E}$ is the electrical field. However, a large Hall resistivity can ensue in the perpendicular configuration.

Further support for our explanation comes from the composition dependence of the observed effects. Upon substitution of Co for Fe in $Y_2Fe_{17-x}Co_x$ alloys, we find that the behavior of the AHE does not vary significantly for $x \leq 2$. Preferential substitution which leaves the dumbbell sites unchanged (where Fe ions with nonzero orbital moment reside) can account for this. However, the large anisotropy in the AHE resistivity as well as huge skew scattering vanishes in alloys with $x > 2$. We attribute this to variations in the electronic structure of the $Y_2Fe_{17-x}Co_x$ system with increasing Co content.

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