Temperature dependence of the anomalous Hall conductivity in the Heusler alloy Co_2CrAl

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The Heusler alloy, Co_2CrAl, is an itinerant magnet that orders at $T=333$ K. We have measured its temperature-dependent anomalous Hall effect and magnetoresistance from room temperature down to low temperatures. The magnetoresistance is positive and very small. The anomalous Hall conductivity follows the magnetization with the ratio of the two quantities being constant over the whole temperature range, suggesting that the origin of the anomalous Hall effect is intrinsic.

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In 1879, Edwin Herbert Hall discovered a voltage perpendicular to the current direction in the presence of a perpendicular magnetic field, the now well-understood Hall effect. Within a year, Hall[1] measured ferromagnetic iron with a Hall coefficient about ten times larger than that of silver and gold (which he had studied before) and of opposite sign. Over the years, this extraordinary or anomalous Hall effect (AHE) in magnetic materials has led to considerable experimental and theoretical work, with a peak of activities in the 1950s and 1960s.[2,3] More recently, the sharp rise of interest in magnetic materials for spin manipulation in semiconductor devices has revived efforts to investigate this phenomenon and its cousin, the spin Hall effect,[4] further.

The current density in a ferromagnetic metal depends linearly, to the first order (neglecting any higher-order terms), on the electric field **E**, internal magnetic field **B**, and magnetization **M** as follows:[2]

$$\mathbf{J} = \sigma_0 \mathbf{E} + \sigma_H \mathbf{B} \times \mathbf{E} + \sigma_H{}^1 \mathbf{E} \times \mathbf{M}. \quad (1)$$

The first term is the normal electrical conductivity, the second shows the ordinary Hall effect, while the dependence on the magnetization leads to the AHE. Origins for the AHE are multiple but can be divided into two classes. The first class entails all mechanisms that are so-called extrinsic. The existence of the spin-orbit coupling term in the Boltzmann transport equation leads to spin-dependent scattering events, such as skew scattering[5] by impurities, thermal spin disorder, and phonons, and side jumps of quantum mechanical origin;[6] for an overview see Ref. 2. The second class entails all mechanisms that are so-called intrinsic. Karplus and Luttinger[7] in 1954 looked at the contributions to the AHE from the anomalous-velocity term. In modern language, the intrinsic effects are geometric due to Berry phase connections. This is a band structure property[8–10] or, as in the manganites such as $La_{0.7}Ca_{0.3}MnO_3$,[11] due to a topologically nontrivial spin background.

Earlier experimental studies had focused on distinguishing between skew scattering and the side-jump mechanism.[12] It is difficult to calculate, theoretically, the exact size of the AHE from scattering effects; however, comparison between experiments and calculations show discrepancies by orders of magnitude.[13]

Recently, more evidence is pointing toward the importance of the intrinsic effects. Initially, real-space effects in the manganites led to theoretical calculations[11] of the AHE from geometric effects. These calculations confirmed the experimentally measured temperature dependence of the AHE in the manganite $La_{0.7}Ca_{0.3}MnO_3$.[14] More recently, comparison between theoretical calculations of the intrinsic effects from the Bloch wave approach and experiments in the magnetic semiconductors (III,Mn)V,[8] in $SrRuO_3$,[9] and in Fe[10] have shown good agreement. Lee *et al.*[15] experimentally argued a strong case that the Hall current in the ferromagnetic spinel system, $CuCr_2Se_{4-x}Br_x$, is intrinsic.

In this paper, we report on an experimental study of the temperature dependence of the anomalous Hall conductivity in the ferromagnetic metal Co_2CrAl. Co_2CrAl is a full Heusler alloy that orders into the $L2_1$ structure. It has been predicted that this compound has a fully spin-polarized conduction band,[16,17] making it a suitable candidate for spintronic devices. Our samples are polycrystalline with a grain size of about 50 μm. Cobalt, chromium, and aluminum of at least 99.99% purity were vacuum arc melted, until all components, combined, held at melting temperature to allow mixing of the components, and then cooled. The ingot was turned over and remelted. Rapid cooling took place in a water-cooled copper crucible.

The composition of the samples was determined using energy dispersive x-ray analysis in a scanning electron microscope with a relative precision of 1.5%. The samples were slightly deficient in Al, the stoichiometry was $Co_{2.06}Cr_{1.04}Al_{0.90}$, with small deviations in composition of $<4\%$ throughout the sample. The samples were mechanically polished to be optically flat on both sides down to 337 μm thickness. Electrical contacts were done by wire-bonding thin Au wires to silver epoxy pads on a rectangular sample of dimensions 1 mm$\times$0.55 mm$\times$0.34 mm. Contact resistances of a few ohms were achieved. Six probe-resistivity and Hall measurements were done with a low-frequency ac lock-in technique in the dc limit. The absolute error bars on the resistivity are 20% due to the finite size of the contacts; the relative errors are 0.02%. Magnetization data were taken in a superconducting quantum interference device (SQUID) with fields of up to 50 kOe.

Figure 1 shows the resistivity as a function of magnetic field at $T=36$ K and 278 K and as a function of temperature on cooling in zero field (see inset). Interestingly, the longitudinal resistivity increases almost linearly with decreasing temperature. Similar behavior has been seen in

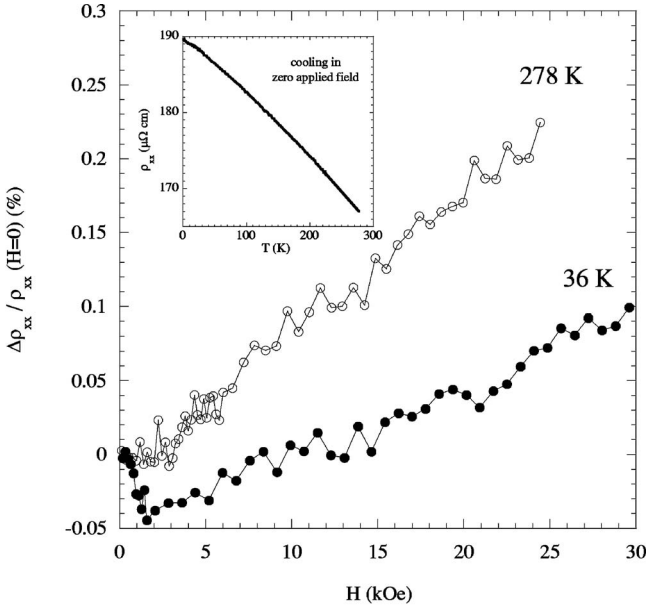

FIG. 1. Magnetoresistance at $T=36$ and 278 K. Inset: longitudinal resistivity, ρ_{xx}, as a function of temperature from room temperature down to 1.5 K.

$Co_2Cr_{0.6}Fe_{0.4}Al$.[18] The origin for this temperature dependence remains unclear. Single crystals of good quality are needed to determine to what extent this temperature dependence is intrinsic to the material and to what extent it is due to scattering of impurities and grain boundaries. We observe only a very small, unsaturated positive magnetoresistance of 0.1% at a magnetic field $H_{\mathrm{appl}}=30$ kOe at 36 K. The slope roughly doubles at 278 K (see Fig. 1). Semiclassical theory of magnetotransport in metals[19] predicts a magnetoresistance of this order that increases with decreasing zero field resistivity ρ_{xx}. There is a small negative magnetoresistance at low temperatures up to $H_{\mathrm{appl}}=2$ kOe. At this applied field, the Hall data (see Fig. 2) shows that the spins have ordered, and the initial negative magnetoresistance is due to reduced scattering from spin disorder as the domains align.

The magnetization as a function of temperature in a magnetic field $H_{\mathrm{appl}}=20$ kOe can be fitted well to $M=M_S\,[1-(T/T_C)^2]^{0.5}$. This fit gives a ferromagnetic transition temperature $T_C=333$ K, as shown in Fig. 3. The saturation magnetization is 311 Gauss/4π, which translates to a saturated moment per formula unit of $1.65\pm0.05\ \mu_B$. This is significantly smaller than the theoretical predicted value of 3 μ_B.[16] However, it is in agreement with other experimental results reported by Refs. 20 (1.55 μ_B) and 21 (1.5–3 μ_B). Co_2CrAl is a soft magnet: the coercive fields are of the order of 100 Oe at low temperatures, decreasing to around 10 Oe at room temperature. More of the $M(H,T)$ phase space has been measured and will be published elsewhere.[22]

In our polycrystals with cubic symmetry, all conductivity tensors in Eq. (1) are scalars. For ferromagnetic metals in applied magnetic fields up to field strength of a few kOersted, $\sigma_0 > \sigma_H^1|M| > \sigma_H|B|$. The magnetization vector **M** is aligned with the applied magnetic field in the z direction (M_x and M_y are zero) in our bulk samples. By inverting the above vector equation, we can write ρ_{xy} as: $\rho_{xy}=R_0\,B_z$

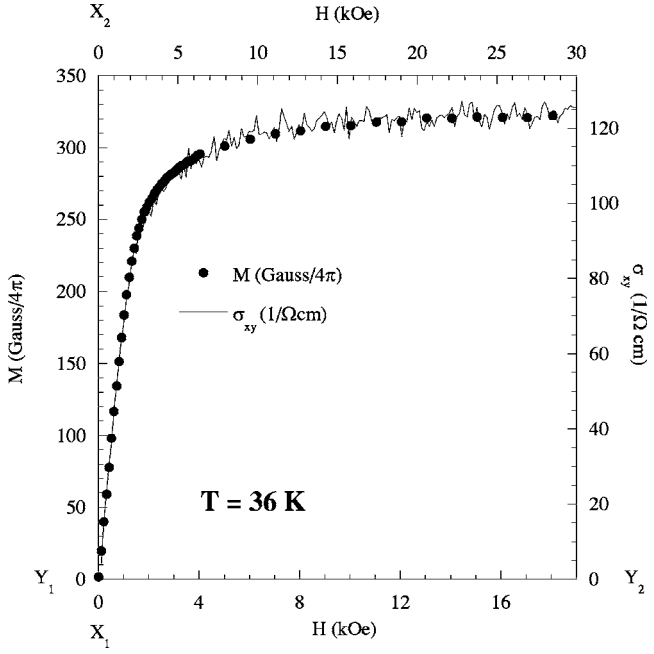

FIG. 2. Y_1 shows the magnetization M as a function of magnetic field at $T=36$ K, Y_2 the Hall conductivity σ_{xy} at the same temperature. The two data sets were taken on two different samples with different demagnetization corrections, hence, the different magnetic field scales.

$+R_S\,M_z$. This is the more familiar version of Eq. (1). To first order, $R_0=\sigma_H/(\sigma_0)^2$, $R_S=\sigma_H^1/(\sigma_0)^2$, and $\sigma_{xy}=\rho_{xy}/(\rho_{xx})^2$. The maximum Hall angle φ (defined as $\tan\varphi=\rho_{xy}/\rho_{xx}=\omega_c\tau$, ω_c is the cyclotron frequency and τ the scattering time) in our experiments is only 1.2° ($\tan\varphi=0.021$), which justifies the approximations in inverting Eq. (1) that neglect any higher-order terms in φ.

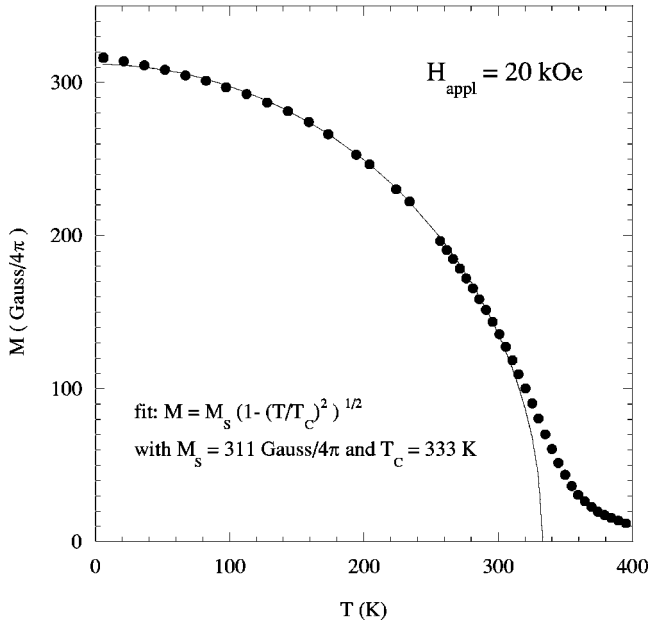

FIG. 3. Temperature-dependent magnetization in applied magnetic field $H_{\mathrm{appl}}=20$ kOe on cooling. The fit $M(T)=M_S\,[1-(T/T_C)^2]^{0.5}$ gives $M_S=311$ Gauss/4π and $T_C=333$ K.

We show in Fig. 2 magnetization data on the Y_1 axis and Hall conductivity $\sigma_{xy}=\rho_{xy}/(\rho_{xx})^2$ on the Y_2 axis at $T=36$ K. Both are shown as a function of applied magnetic field. The two data sets are taken on two samples with different demagnetization corrections, hence, the different field axes. By rescaling one of the field axes, both data sets overlap. This scaling factor is constant over the whole temperature range of the experiment.

As can be seen from the conductivity data, we cannot make a good estimate for the carrier concentration. The slope up to 30 kOe is 6.1×10^{-12} Ω cm/Oe (both at $T=36$ K and at $T=278$ K) due to the long tail in magnetization. From that, we can estimate that the carrier concentration is larger than 3×10^{20} cm^{-3}, which is more than 1.5×10^{-3} carriers per unit cell. The exact carrier concentration should be measured well above the Curie temperature, where the contributions from the AHE are expected to be negligible.

In Fig. 4, we show the combined data for all temperatures. We have plotted Hall conductivity versus magnetization by implicitly taking out the dependence on the externally applied magnetic field. All the data fall on one straight line. In the inset, we show the fits at each temperature to σ_H^1; see Eq. (1) for definition. This collapse of all data implies that the anomalous Hall conductivity coefficient σ_H^1 is a constant over the range of temperatures probed in this experiment, $\sigma_H^1=0.383\pm0.003$ Gauss/4 $\pi\Omega$ cm (±0.14 Gauss/4 $\pi\Omega$ cm from systematic errors in absolute resistivity and magnetization). For this itinerant magnet, the global magnetization has replaced the local magnetization in Eq. (1). This simple description of the temperature dependence of the AHE suggests that, just as in Fe and dilute semiconductors (GaMn)As as well as the spinel ferromagnet $CuCr_2Se_{4-x}Br_x$, the measured anomalous Hall effect in Co_2CrAl is largely due to intrinsic effects.

Figure 5 in Ref. 23 compares σ_{xy} versus M for a large number of different itinerant ferromagnets. Our data stretches from the group of itinerant magnets, such as dilute Ni alloys,[24] the Half Heuslers CoMnSb and NiMnSb,[25] and $\alpha Co_{0.72}Gd_{0.15}Mo_{0.11}$ (Ref. 26) films down to $(Fe_{1-y}Co_y)Si$.[23] It is interesting to note that $\sigma_H^1(=\sigma_{xy}/M)$ is very similar in all these different material systems.

We hope that our result will encourage more theoretical calculations of the intrinsic contributions to the AHE from first principles even in more complex compounds, such as these Heusler alloys. There are practical considerations for why it is important to know whether the AHE is intrinsic or extrinsic as has been pointed out by Lee *et al.*[15] For the extrinsic case, the scattering direction depends on the angle between the spin of the conduction electron and the scattering center. This leads to a spin-Hall current with the same spin polarization as the conduction electrons. However, the spin-Hall current is predicted to be fully spin polarized for all itinerant ferromagnets in the intrinsic case, even for magnetic metals that are not half metallic.

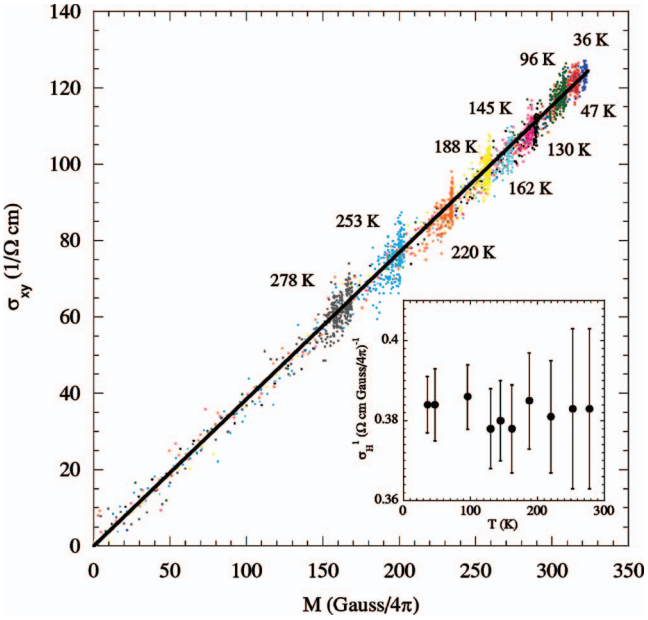

FIG. 4. (Color) Combined data σ_{xy} as a function of M at temperatures $T=36$, 47, 96, 130, 145, 162, 188, 220, 253, and 278 K. The line through the data is a guide to the eye. Insert: fits to σ_H^1 (see text for definition) at each temperature.

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