Half-Metallic Ferromagnetism with Unexpectedly Small Spin Splitting in the Heusler Compound Co_2FeSi

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Half-metallic ferromagnetism stands for the technologically sought-after metallicity with 100% spin polarization. Electrical transport should, in principle, sensitively probe half-metallic ferromagnetism, since electron-magnon scattering processes are expected to be absent, with clear-cut consequences for the resistivity and the magnetoresistance. Here we present electrical transport data for single-crystalline Co_2FeSi, a candidate half-metallic ferromagnet Heusler compound. The data reveal a textbooklike exponential suppression of the electron-magnon scattering rate with decreasing temperature which provides strong evidence that this material indeed possesses perfect spin polarization at low temperature. However, the energy scale for thermally activated spin-flip scattering is relatively low (activation gap $\Delta \approx 100$ K) which has decisive influence on the magnetoresistance and the anomalous Hall effect, which exhibit strong qualitative changes when crossing $T \approx 100$ K.

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A crucial ingredient of any kind of spintronics, i.e., the exploitation of spin-dependent electron transport phenomena [1,2], is the realization of highly spin-polarized materials. Half-metallic ferromagnets (HMFs) [3] are considered ideal candidates [4], because they are predicted to possess a 100% spin polarization at the Fermi level E_F. The complete absence of minority spin states at E_F implies the existence of a gap Δ for only one spin direction [see Fig. 1(a)] and, thus, clear-cut consequences in various physical properties [4,5]. For the electrical resistivity ρ, one expects for $T \lesssim \Delta$ an exponential suppression of the characteristic $\sim T^2$ dependence that arises in ordinary ferromagnets from electron-magnon (i.e., spin-flip) scattering [6–8], because in a HMF material the presence of the gap Δ suppresses spin flips of conduction electrons [5,9]. Despite its simplicity, clean experimental evidence for such a fundamental behavior is lacking, because experimental realizations of true HMF materials are still scarce. Apart from pioneering transport studies on HMF candidate materials [10,11], an exponentially suppressed spin-flip scattering has been observed only in granular CrO_2 thin films [8].

HMF physics has originally been predicted [3] and is being discussed for some Heusler compounds [10–16]. Most prominent among these is the material Co_2FeSi, since this ferromagnetic metal possesses the highest Curie temperature ever found in Heusler compounds ($T_C = 1100$ K) together with a large magnetic moment of $m = 6\mu_B$/f.u. ("formula units" are denoted by f.u.). Based on these experimental facts, the size of the minority spin state splitting has been theoretically estimated as $\Delta \gtrsim 0.1$ eV [14]. In this Letter, we present the electronic transport properties, viz., resistivity, magnetoresistance, and Hall effect, of high-quality Co_2FeSi single crystals [15]. Our data provide fresh evidence of Co_2FeSi being a true HMF, because we indeed observe an exponentially suppressed $\sim T^2$ behavior of the resistivity at $T \lesssim 100$ K, consistent with an unexpectedly small $\Delta \sim 100$ K, as is inferred from careful analysis. Our results represents therefore a textbook example of the expected transport behavior for a HMF. This crucial finding is further corroborated by a sign change of magnetoresistance from negative at high temperatures ($T \gtrsim 110$ K) to

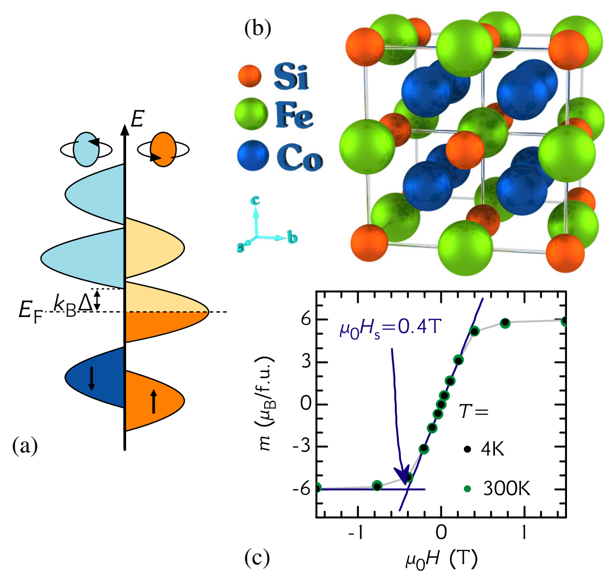

FIG. 1 (color online). (a) $L2_1$ crystal structure of Co_2FeSi. (b) Sketch of the density of states of a HMF for minority and majority electrons with an energy gap at Fermi level E_F for minority electrons. (c) Magnetic moment of a Co_2FeSi single crystal.

positive at lower temperatures. Furthermore, we observe that the thermal activation of spin-flip scattering across Δ has a profound impact on the anomalous Hall effect (AHE) [17]. While the anomalous Hall coefficient is practically temperature independent at $T \lesssim 100$ K, consistent with dominating extrinsic skew scattering or the ordinary Lorentz-force induced Hall effect, it strongly increases at higher T, where it follows $\sim\rho^2$, suggestive of dominating Berry phase and/or side-jump contributions caused by ferromagnetic magnons.

Experimental details.—Large single crystals of Co_2FeSi, which crystallizes in the $L2_1$ structure with a unit cell size of $a = 5.64$ Å [16] [Fig. 1(b)], were prepared by using the floating zone technique [15]. Magnetization measurements confirm the predicted magnetization of $m = 6\mu_B$/f.u. and yield a saturation field of $\mu_0 H_s = 0.4$ T [see Fig. 1(c)]. The susceptibility below H_s is $\chi = 5.4$. Because of the high Curie temperature, the temperature dependences of the saturation field and susceptibility are negligible up to room temperature. For resistivity measurements in a magnetic field, a single crystal with dimensions of $a = b = c = 3$ mm was used. Resistivity was measured by using a standard four-probe alternating current dc technique. Hall measurements were done simultaneously by using two additional Hall voltage contacts.

Resistivity.—Figure 2 shows the resistivity ρ as a function of temperature T for zero magnetic field and for $\mu_0 H = 15$ T. In zero field the resistivity shows a clear metallic temperature dependence with a monotonic increase with increasing temperature. A residual resistivity of $\rho_R \approx 4\ \mu\Omega$ cm and a residual resistivity ratio of $\rho(300\ \text{K})/\rho_R = 6.5$ show the extraordinary quality of our samples.

The resistivity of metals is usually dominated by electron-phonon scattering. This typically leads to a linear temperature dependence at elevated temperatures ($T \gtrsim 50$ K) and a saturation towards residual resistivity at lower temperatures. Such a linear part is completely absent in Co_2FeSi. However, for ferromagnetic materials like the compound under scrutiny, another scattering process may be found, i.e., electron-magnon scattering, which often leads to a quadratic temperature dependence of the resistivity (see, e.g., [7,11,18–22]).

However, the attempt to fit $\rho(T)$ of Co_2FeSi with a quadratic and a quadratic plus linear temperature dependence (for pure magnon and magnon plus phonon scattering of the electrons, respectively) fails completely (see Fig. 3). Thus, the conventional electron-magnon scattering of band ferromagnets cannot account for the observed temperature dependence of the resistivity. It can, however, be rationalized much better if the predicted half-metallic character of the material is taken into account. In HMF, the minority spin electronic states are completely gapped at the Fermi level, where $k_B\Delta$ is the minimum excitation energy of majority charge carriers to occupy empty minority states involving a spin flip [see Fig. 1(b)]. As a consequence, at low temperatures ($T \lesssim \Delta$) electron-magnon scattering (which involves a spin flip) must be exponentially suppressed. At higher temperatures ($T \gtrsim \Delta$) magnon scattering would become possible when the Fermi distribution smears out the occupation of energy states around the Fermi level.

In order to test against this scenario, we fitted the resistivity with combined scattering of a residual (ρ_R), magnonic (ρ_M), and phononic (ρ_P) contribution,

$$\rho(T) = \rho_R + \rho_M(T) + \rho_P(T). \qquad (1)$$

The residual resistivity ρ_R is temperature independent and caused by defects of the ideal crystal lattice. The usually

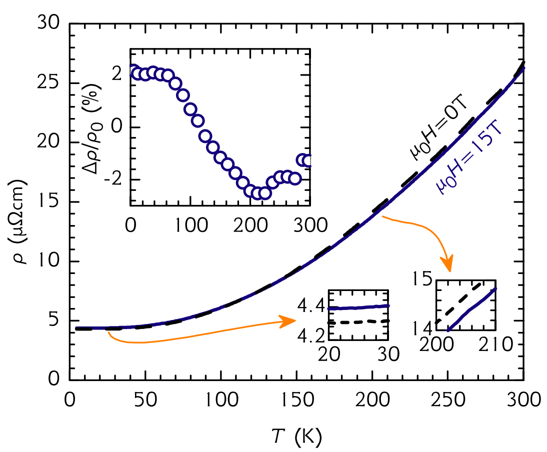

FIG. 2 (color online). Temperature dependence of the resistivity in zero and 15 T magnetic field. Upper left inset: difference [$\Delta\rho = \rho(15\ \text{T}) - \rho(0\ \text{T})$, $\rho_0 = \rho(0\ \text{T})$] of these measurements. Lower right insets: close-up to low and high temperatures.

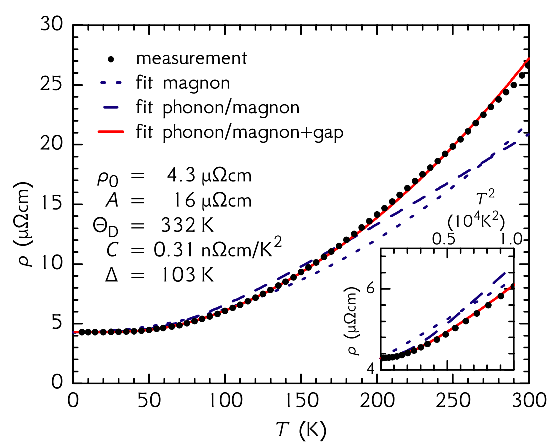

FIG. 3 (color online). Resistivity as a function of the temperature. Measured values are displayed as black dots. Blue dashed lines show fits with magnonic ($\sim T^2$) and magnonic-phononic temperature dependence. The red continuous line shows a fit where an additional Boltzmann factor was added to the magnonic scattering; its fitting parameters are displayed (see the text for formulas).

quadratic magnonic term (ρ_M) is expected to be exponentially suppressed due to the gapped spin-flip scattering [6–8]. Since a rigorous theory for the description of this suppression is still lacking up to present, we empirically weight the usual electron-magnon scattering term by a Boltzmann factor [8],

$$\rho_M(T) = CT^2 e^{-\Delta/T}. \quad (2)$$

The parameter C is a measure of the strength of the magnon scattering process. The phononic part of the scattering process (ρ_P) is described by the Bloch-Grüneisen formula,

$$\rho_P(T) = A\left(\frac{T}{\Theta_D}\right)^5 \int_0^{\Theta_D/T} \frac{x^5}{(e^x - 1)(1 - e^{-x})} dx. \quad (3)$$

With the Debye temperature $\Theta_D = 332$ K, determined from specific heat measurements, the resistivity fit [24] yields an energy gap [25] of

$$\Delta = 103 \text{ K} \quad (k_B\Delta = 8.9 \text{ meV}). \quad (4)$$

This energy gap measures the difference of the Fermi energy and the nearest band edge of unoccupied minority spins; see Fig. 1(a). It is worthwhile to point out that the extracted gap size is much smaller as compared to theoretical calculations which predict a more than 1 order of magnitude larger separation of the minority states from the Fermi level. Nevertheless, the observed exponential suppression of spin-flip scattering confirms clear HMF behavior. However, the unexpectedly small extracted value of Δ implies that perfect spin polarization of the electrons at the Fermi level is present only for $T < \Delta$.

Magnetoresistance.—An independent verification of the afore-derived scenario of gapped-out magnon scattering can be obtained from the corresponding magnetoresistance of Co_2FeSi, because the influence of a magnetic field on the magnons should be reflected by the resistivity. As can be seen in Fig. 2, a magnetic field of 15 T yields a relatively small ($\sim 2\%$) but well resolvable change of the resistivity as compared to the zero-field data. The inset highlights the sign and the temperature dependence of $\Delta\rho = \rho(15 \text{ T}) - \rho(0)$. Apparently, $\Delta\rho > 0$ ($\Delta\rho < 0$) for $T \lesssim 110$ K ($T \gtrsim 110$ K).

To get a more detailed view of this dependence on the magnetic field we measured the resistivity while sweeping the magnetic field and maintaining the temperatures (Fig. 4). The inset in Fig. 4 shows the slope of the resistivity for fields higher than the saturation field. Up to the saturation field of $\mu_0 H_s = 0.4$ T, a small decrease of the resistivity can be seen, which can naturally be explained by alignment and joining of magnetic domains which decrease the probability of domain wall scattering. For high fields $H > H_s$, the resistivity decreases with increasing field at high temperatures; at low temperatures it increases with increasing field.

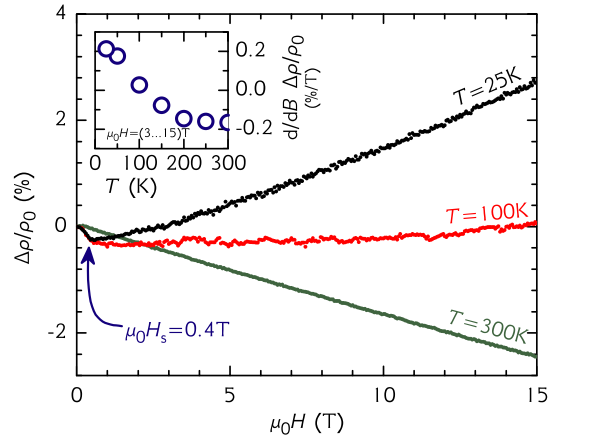

FIG. 4 (color online). Magnetic field dependence of resistivity at different temperatures. Inset: slope of $\rho(B)$ in the applied field range of $\mu_0 H = (3–15)$ T.

This temperature dependence of the magnetoresistance can perfectly be explained by the afore-presented scenario of the resistivity being dominated by gapped spin-flip scattering of spin-polarized electrons. The negative magnetoresistance at high temperatures signals a decrease of the electronic scattering with increasing field. This is consistent with ferromagnetic magnons being the main scattering centers for electrons, since a magnetic field enlarges the magnon energy and thus lowers the number of magnons. Upon lowering the temperature, electron magnon scattering in a HMF is expected to freeze out exponentially, since only one spin state is present at the Fermi level. Hence, electron-defect scattering must become important at lower temperatures, and one expects a more conventional positive magnetoresistance. Indeed, such a behavior is observed at $T \lesssim 110$ K: $\Delta\rho > 0$ and $\Delta\rho(B)$ approaches a positive quadratic field dependence, which is characteristic for conventional multiband metals.

Hall effect.—Having established Co_2FeSi being a HMF and activated spin-flip scattering for $T \gtrsim 100$ K, we now move on to the AHE of Co_2FeSi. The Hall resistivity of this compound is shown in Fig. 5 along with its corresponding Hall coefficients in the inset. The Hall resistivity $\rho_{xy}(\mu_0 H)$ is characteristic for the AHE in ferromagnetic materials: It has a linear dependence on the applied magnetic field but with a strong kink and a changing slope just at the saturation field of $\mu_0 H_s = 0.4$ T. Since the Hall effect depends on the magnetic field H as well as on its magnetization M, one considers two Hall coefficients, viz. the ordinary Hall coefficient R and the so-called anomalous Hall coefficient R_A, which connects the applied magnetic field H and the magnetization M of the sample, respectively, with the Hall resistivity [11,17],

$$\rho_{xy} = \mu_0(RH + R_A M). \quad (5)$$

Thus, from our Hall data in high magnetic fields the ordinary Hall coefficient R can be obtained, and, with the

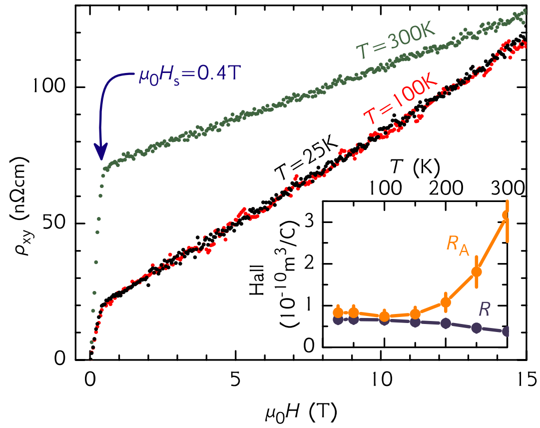

FIG. 5 (color online). Hall resistivity as a function of the applied magnetic field for different temperatures. Inset: ordinary Hall coefficient R and anomalous Hall coefficient R_A as a function of the temperature.

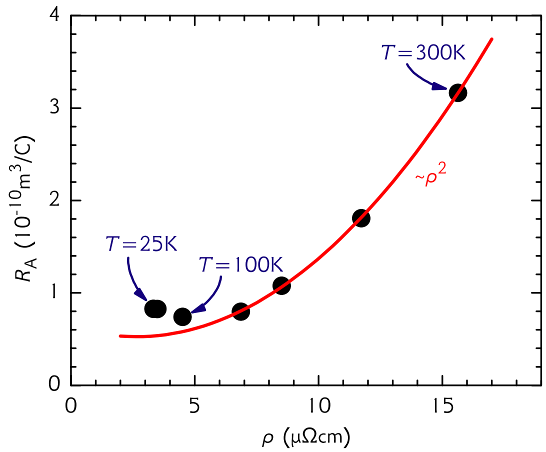

FIG. 6 (color online). Anomalous Hall coefficient R_A as a function of the zero-field resistivity ρ (black circles). Line: quadratic fit for temperatures above 100 K.

knowledge of this coefficient and $\chi = 5.4$, the anomalous Hall coefficient R_A can be extracted from the low-field data, where $M = \chi H$. The results are shown in the inset in Fig. 5. The ordinary Hall coefficient R is small and does not change much with temperature as expected for a metal. It slightly decreases with increasing temperature which indicates a change of the mobility of the charge carriers consistent with the multiband nature of this material [14]. At low temperatures the anomalous Hall coefficient R_A is comparable to the ordinary one, while at $T \gtrsim 100$ K it strongly increases with T. The huge increase of R_A cannot be explained by Lorentz force deflection and implies the relevance of fundamental electronic deflection mechanisms known in ferromagnetic materials. More specifically, following the classification given by Nagaosa *et al.* [17], one expects on the one hand $R_A \sim \rho^2$ for the *intrinsic* Berry phase-related contribution to the AHE as well for the so-called *side-jump* scattering contribution. On the other hand, for the extrinsic *skew scattering*, $R_A \sim \rho$ is expected. In order to test the AHE in Co_2FeSi against one of these scenarios, we plot R_A as a function of the measured $\rho(T)$ (Fig. 6). Indeed, for ρ larger than the residual resistivity ρ_R, the data very well follow $R_A \sim (\rho - \rho_R)^2 + \text{const}$, which provides clear evidence that the AHE is dominated by the intrinsic Berry phase and/or the side-jump scattering contributions. Interestingly, this resistivity regime corresponds to temperatures $T \gtrsim 100$ K, i.e., to high temperatures where spin-flip scattering becomes possible, which may affect both the Berry phase as well as the side-jump scattering. The latter requires magnetic scattering centers, which can be realized by the magnon-related spin-flip scattering. At lower resistivity values, which correspond to the low-temperature regime where magnon scattering apparently freezes out, the observed $\sim\rho^2$ behavior breaks down and R_A becomes independent of ρ, implying that a qualitatively different deflection mechanism becomes dominant. Indeed, the comparable magnitude of the anomalous (R_A) and ordinary (R) Hall coefficients for $T \lesssim 100$ K suggests a contribution of the Lorentz-force induced Hall effect (see the inset in Fig. 5), but also skew scattering contributions cannot be excluded. Note that the temperature-dependent change of ρ in this regime is very small, and thus R_A cannot be tested against the expected linear dependence of ρ. Impurity-dependent investigations of R_A should shed more light on this issue but are beyond the scope of this study. It is, however, worthwhile to mention that Imort *et al.*, in agreement with the conjectured skew scattering, reported $R_A \sim \rho$ for Co_2FeSi thin films, which naturally contain more defects than our high-quality crystal [23]. Finally, it is worthwhile to point out that the easily accessible and by temperature well distinct regimes of the AHE suggest Co_2FeSi as an ideal test bed for theoretical treatments of the AHE.

In conclusion, for Co_2FeSi our electrical transport and magnetotransport provide compelling evidence that this material indeed is a half-metallic ferromagnet. However, the extracted separation of the minority spin state from the Fermi level $\Delta \approx 100$ K is much smaller than is expected from theory. Thus, the technological exploitation of perfect spin polarization in Co_2FeSi at room temperature and above seems out of reach. However, Co_2FeSi remains of fundamental interest, because this material is a perfect material to study the consequences of half-metallic ferromagnetism over a large temperature regime. In particular, it provides access to the crossover regime between half-metallic ferromagnetism ($T \ll \Delta$) to more conventional ferromagnetism ($T \gtrsim \Delta$) in an experimentally convenient temperature range. The observed properties are indeed intriguing: First, spin-flip scattering becomes important in the resistivity in a thermally activated fashion which allows one to quantitatively extract Δ. Second, the magnetoresistance changes sign from positive to negative upon thermally smearing out the Fermi edge across Δ, and finally, we observe that the activation of spin-flip scattering at high temperatures $T \gtrsim \Delta$ has a strong impact on the anomalous Hall effect. In this high-temperature regime it is strongly temperature dependent and governed by Berry phase and side-jump deflection, whereas at $T \lesssim \Delta$ it becomes temperature independent and is consistent with dominating skew scattering or intrinsic Lorentz-force induced Hall effect.

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