Berry-phase contribution to the anomalous Hall effect in gadolinium

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(Received 20 March 2004; revised manuscript received 27 October 2004; published 14 March 2005)

When conduction electrons are forced to follow the local spin texture, the resulting Berry phase can induce an anomalous Hall effect (AHE). In gadolinium, as in double-exchange magnets, the exchange interaction is mediated by the conduction electrons and the AHE may therefore resemble that of CrO_2 and other metallic double-exchange ferromagnets. The Hall resistivity, magnetoresistance, and magnetization of single crystal gadolinium were measured in fields up to 30 T. Measurements between 2 K and 400 K are consistent with previously reported data. A scaling analysis for the Hall resistivity as a function of the magnetization suggests the presence of a Berry-phase contribution to the anomalous Hall effect.

DOI: 10.1103/PhysRevB.71.104407 PACS number(s): 75.47.Np, 72.15.Gd

I. INTRODUCTION

While many theories account for an anomalous Hall effect (AHE), proportional to the magnetization of a material, these theories often predict effects significantly smaller than those found in ferromagnetic materials.[1–7] An even more significant deficiency of the conventional theories is that most predict an anomalous Hall resistivity that is proportional to a power of the longitudinal resistivity, and in the absence of a metal-insulator transition cannot account for an AHE that peaks near the Curie temperature T_C. Kondo's *s-f* (*s-d*) Hamiltonian model may give the correct temperature dependence, but still does not predict an effect of sufficient magnitude.[6,8] Recent models based on a geometric, or Berry, phase have had great success in describing the AHE in double-exchange systems (e.g., manganites and chromium dioxide) and pyrochlores.[7,9–16]

The anomalous Hall effect in chromium dioxide, a metallic double-exchange ferromagnet,[17] was shown[11] to agree well with the description based on geometric phase first suggested by Ye *et al.*[7] In gadolinium, as in double-exchange magnets, the exchange interaction among localized ($4f$) core spins is mediated by the conduction electrons. The anomalous Hall effect may therefore resemble that of CrO_2 and other metallic double-exchange ferromagnets. Monte Carlo simulations predict that the same spin-texture excitations that cause the anomalous Hall effect in double-exchange systems are also intrinsic to Heisenberg ferromagnets.[18] Thus it is reasonable to seek to explain the anomalous Hall effect in other systems using the same theory.

Gadolinium has an unexpectedly large anomalous Hall effect.[19] In particular, when the applied magnetic field is parallel to the *c* axis the anomalous Hall resistivity peaks at $\rho_{xy} \approx -6\ \mu\Omega$ cm just below T_C.[20] This makes it a good candidate for showing a maximum near $2/3$ of its saturation magnetization as chromium dioxide does. Since gadolinium is metallic even above T_C, conventional theories cannot explain a maximum in the Hall effect near the transition temperature. In order to test for the presence of Berry-phase contributions to the anomalous Hall effect, it is necessary to measure the Hall resistivity when the magnetization is greater than (or at least close to) $2/3$ of its saturation value at temperatures near T_C. A maximal anomalous Hall effect in this regime is the signature of Berry-phase contributions. Previous measurements of gadolinium have been at significantly lower magnetization values, except at the lowest temperatures, due to the relatively low applied magnetic fields used. At T_C we have just reached $2/3$ of the saturation magnetization in an applied field of 30 T.

II. SAMPLE PREPARATION

A *c*-axis oriented gadolinium (99.99% purity) single crystal was purchased from MaTecK GmbH. Two cuts were made parallel to an in-plane axis direction, the sides were polished lightly to clean up rough edges from the saw cuts, and the *c*-axis plane was thinned as much as possible. The resulting shape is a rectangular prism with an approximately square cross section and irregular ends. Gold contact pads were sputtered onto the sides of the sample.

III. EXPERIMENTAL RESULTS

Data were taken using a Quantum Design Physical Property Measurement System (PPMS) in fields up to 7 T. The zero-field resistivity for the gadolinium crystal is shown in Fig. 1. An alternating current (37 Hz) is applied along the *a* axis. An abrupt change in slope occurs at the ferromagnetic transition temperature. The residual resistivity ratio ($R_{300\ \mathrm{K}}/R_{4.2\ \mathrm{K}}$) is 31. For Hall effect and magnetization measurements, the field was applied along the *c* axis. The demagnetizing factor $N=0.5$, and the saturation magnetization is 7.7 μ_B/Gd. The large values of the Hall resistivity and the magnetization allowed for very precise measurements (Figs. 2 and 3). Figure 4 shows the Hall resistivity plotted vs reduced magnetization ($m=M/M_{saturation}$); these data were collected in fields up to 7 T. The Hall resistivity increases rapidly with magnetization below the Curie temperature as domains are swept out. There is some indication that the data maximize at $|\rho_{xy}| \approx 7\ \mu\Omega$ cm when $|m| \approx 0.7$. It is conventional to separate the Hall resistivity into ordinary [ordinary Hall effect (OHE)] and anomalous (AHE) contributions: $\rho_{xy}=R_oB_{in}+R_s\mu_0M$, where $B_{in}=\mu_0H_{applied}+\mu_0(1-N)M$. R_o and R_s are the ordinary and anomalous (or spontaneous) Hall

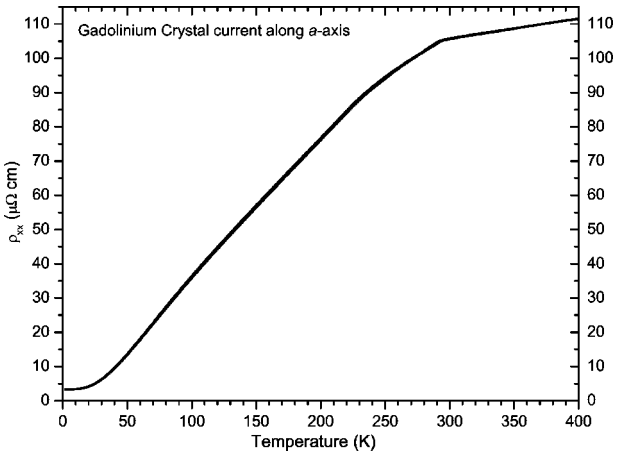

FIG. 1. Gadolinium resistivity vs temperature.

coefficients, respectively. The upturns at large values of m are from the OHE, which is small but not completely negligible.

It is difficult to make a reliable separation of the OHE and AHE contributions. To obtain the values shown in Fig. 5, we first choose the anomalous Hall coefficient R_s. Next, the corresponding term (linearly proportional to magnetization) is subtracted from the dataset. Then, a linear least-squares fit of Hall resistivity vs internal field is made. The value chosen for the anomalous Hall coefficient is adjusted until the fitting error is minimized. The best-fit anomalous Hall coefficients are shown in Fig. 6. This method works even slightly above T_C because of the large demagnetizing correction; at temperatures significantly above T_C the magnetization curves become linear in field, and this method fails. The other disadvantage to this method is that the Berry-phase theories predict that the anomalous Hall resistivity is linear in magnetization near T_C only for low values of m.

The low-temperature ordinary Hall coefficient agrees with previously reported values (see Fig. 5).[20] The qualitative behavior is also similar. Lee and Legvold report that the ordinary Hall coefficient of gadolinium has temperature dependence which differs dramatically from those of lutetium and yttrium and cannot be explained by a two-band model.[20]

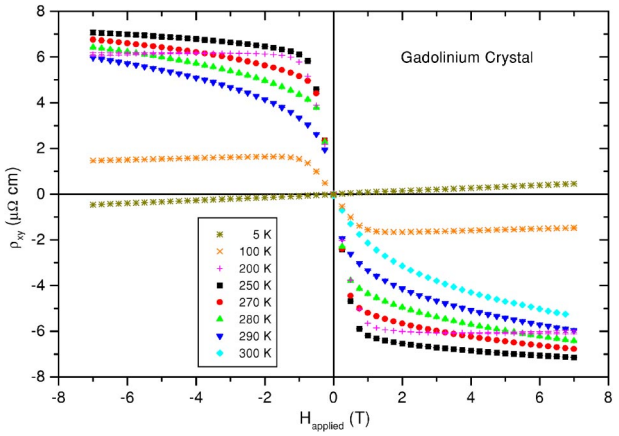

FIG. 2. (Color online) Gadolinium Hall resistivity vs applied magnetic field.

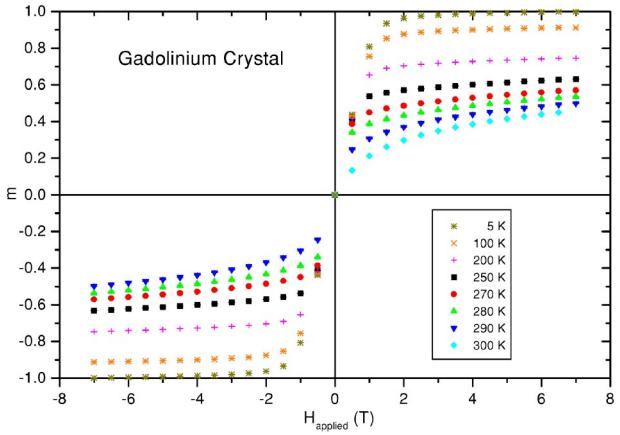

FIG. 3. (Color online) Gadolinium reduced magnetization vs applied magnetic field.

They obtained a Hall coefficient which changes sign near 130 K (instead of 260 K, as seen in Fig. 5) and decreases even more rapidly as T_C is approached. The most likely cause of these discrepancies is a problem with the separation of OHE and AHE. Lee and Legvold only applied 3 T, whereas the values reported here include data up to 7 T. Indeed, when a subtraction was attempted using the noisier 30 T data (see Figs. 7 and 8, and discussion below), the ordinary Hall coefficient did not appear to change sign until T_C. There are two possible explanations for this behavior. The simplest is that the AHE is underestimated, and the residual gives an apparent contribution to the OHE. The other possibility is that the sign change and the sharp increase in the magnitude of the Hall coefficient are real effects (possibly due to exchange splitting of the conduction band). In this case, the decreasing magnitude that we observe at higher fields and higher magnetization may be the result of an anomalous Hall effect that is not strictly linear in magnetization at high fields. This nonlinearity, if real, would support the hypothesis that Berry-phase effects contribute to the anomalous Hall effect in gadolinium. This contribution would decrease as the magnetization increases, thus giving rise to the apparent field dependence of the ordinary Hall coefficient. The ordinary Hall resistivity will not be sub-

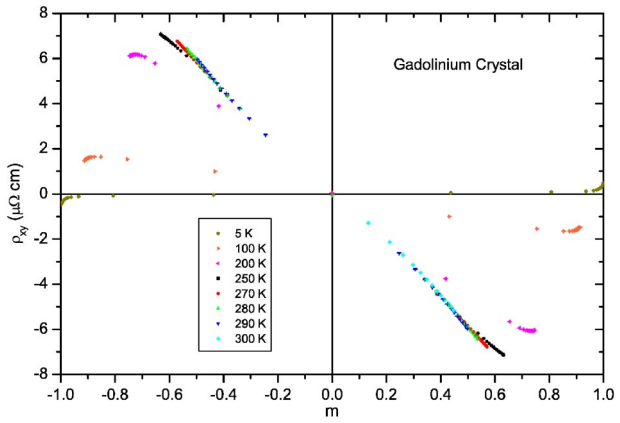

FIG. 4. (Color online) Gadolinium Hall resistivity vs reduced magnetization.

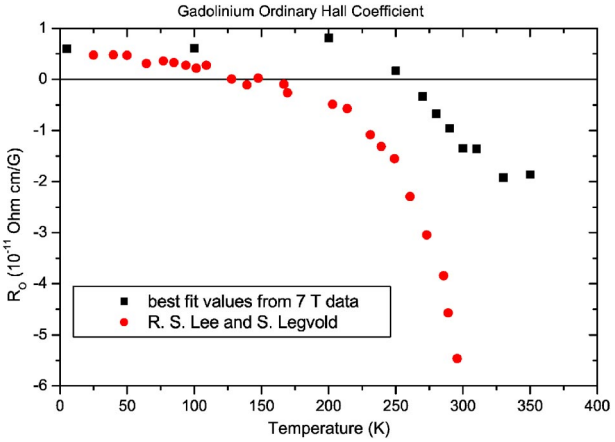

FIG. 5. (Color online) Gadolinium ordinary Hall coefficient vs temperature. This represents the best fit to the data in fields below 7 T. Lee and Legvold's data are shown for comparison. (Ref. 20).

tracted from plots of the data because of this dilemma.

If the anomalous Hall effect results from the thermal excitation of topological excitations, it is possible to use scaling relations for the magnetization and expected Skyrmion density to obtain[11]

$$\rho_{xy}^{A}T = \rho_{xy}^{0}T_C m[1 - D(x)m^{(1-\alpha)/\beta}], \tag{1}$$

where $D(x)$ is a scaling function of the scaling variable $x = t/h^{1/(\beta\delta)}$, and t and h are the reduced temperature and magnetic field, respectively. Along the critical isotherm $t \equiv 1 - T/T_C = 0$, making ρ_{xy} a function of m only.

In an effort to extend the results in the vicinity of the Curie temperature to larger values of m, we measured both the Hall resistivity and magnetization at the National High Magnetic Field Laboratory in fields up to 30 T. The high-field data are consistent with those taken in the PPMS, but are noisier due to problems both with the vibrating sample magnetometer and with pickup from ripple in the Bitter magnets. Nonetheless, there is a clear tendency for the Hall resisitivity to reach an extremal value close to $m = 2/3$. This is shown in Fig. 9, where the closed symbols in the legend are

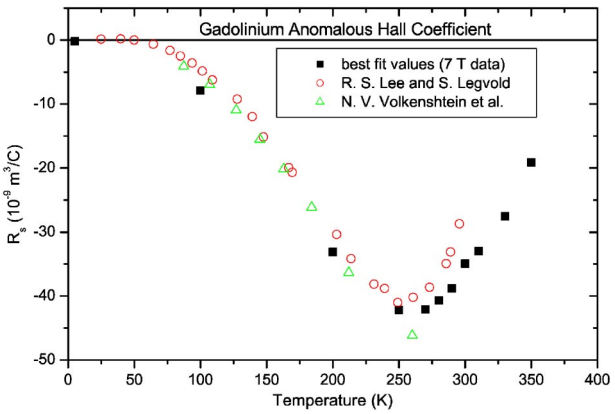

FIG. 6. (Color online) Gadolinium anomalous Hall coefficient vs temperature. Previously reported data are shown for comparison (Refs. 20 and 27).

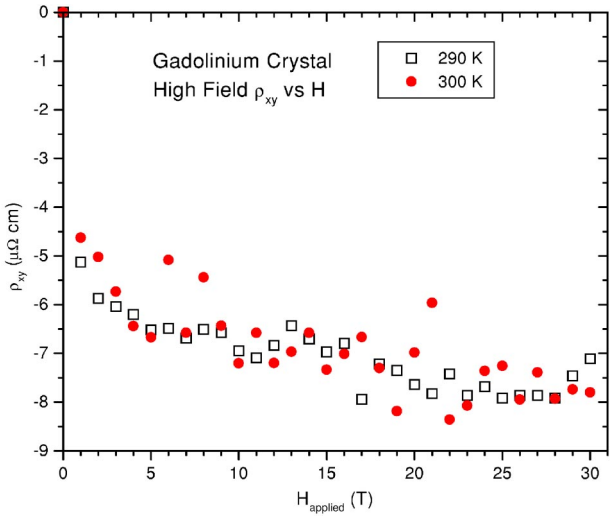

FIG. 7. (Color online) Gadolinium Hall resistivity vs applied magnetic field.

from the PPMS measurements and the remainder from the 30 T experiment. The solid line is the Skyrmion expression [Eq. (1)] using the critical exponents for gadolinium[21] and $D(0) = 1$. The data are expected to fall along this line near T_C, and within the space enclosed by the line and the x axis away from T_C.[11–13]

IV. DISCUSSION

Clearly the data in Fig. 9 do not collapse well, yet suggest a tendency to fit the Skyrmion picture. The initial slope at T_C, $\rho_{xy}^{0} = -15\ \mu\Omega$ cm, depends on the Skyrmion density and spin-orbit constant through

$$\rho_{xy}^{0} = -\frac{1}{ne}\frac{\Phi_0}{\pi}\frac{\lambda_{so} n_e a S}{k_B T_C}\langle n \rangle. \tag{2}$$

Assuming $n_e = 1$ carrier per Gd atom, $S = 7/2$, and a Skyrmion density $\langle n \rangle \approx 0.05$ near T_c, we estimate a spin-orbit

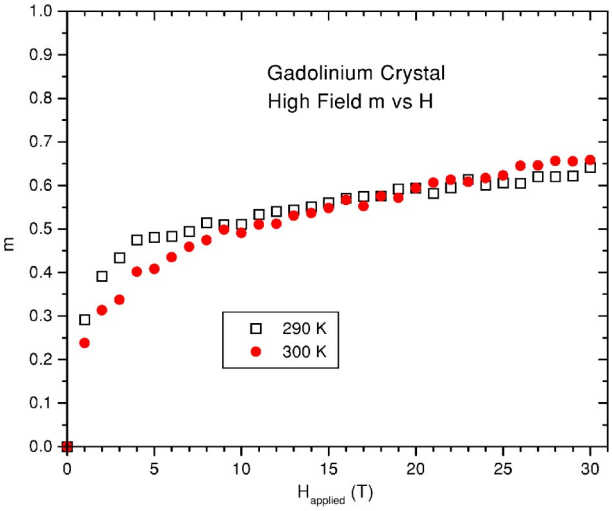

FIG. 8. (Color online) Gadolinium reduced magnetization vs applied magnetic field.

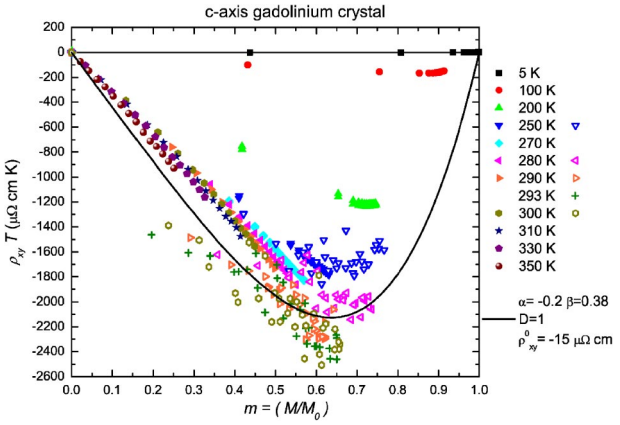

FIG. 9. (Color online) Temperature scaled Hall resistivity vs reduced magnetization.

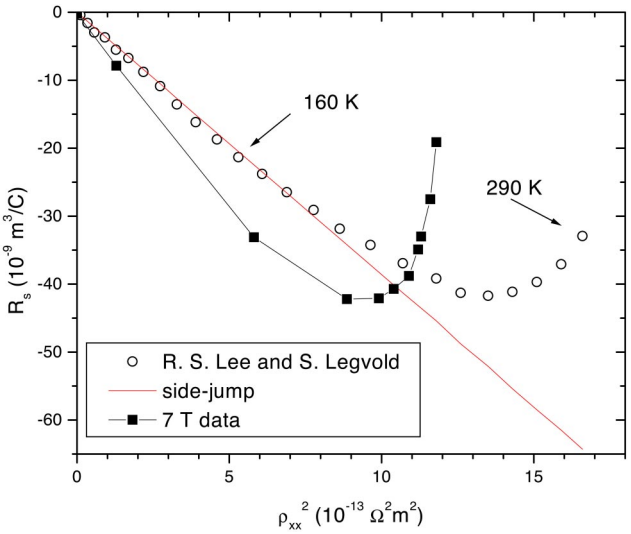

FIG. 10. Anomalous Hall coefficient vs resistivity squared. The residual resistivity has been subtracted. The ordinary Hall coefficient has been neglected when converting Lee and Legvold's data from R_1 to R_s (Ref. 20). The discrepancy in the plots is either due to an error in estimating the length between voltage contacts, or a systematic error in reading Lee and Legvold's data from their log-scale plot. The line is the side-jump prediction using the experimental coefficient for iron (Refs. 1, 3, and 5).

coupling constant of $\lambda_{so} \approx 12$ K. Although the fit is consistent with the data, the data collapse is not so good. The spin-orbit coupling constant also seems rather large. We can make a rough estimate of the spin-orbit coupling energy, as Ye *et al.*[7] have done for manganite, from the Hamiltonian

$$H_{so} = -\frac{\vec{S}\cdot\vec{L}}{2m^2c^2}\frac{1}{r}\frac{\partial V}{\partial r}. \tag{3}$$

Next we approximate the gradient of the potential using

$$\frac{\partial}{\partial r}V = -\frac{\partial}{\partial r}\frac{Ze^2}{r} \approx \frac{Ze^2}{r_d a}, \tag{4}$$

where r_d is the orbital radius and a is the lattice constant. Then an approximation of the spin-orbit coupling λ_{so} is given by

$$\lambda_{so} = \frac{Ze^2\hbar^2 k_{F_z}}{4m^2c^2 r_d a}. \tag{5}$$

In the free-electron model

$$k_{F_z} = \frac{\sqrt[3]{3\pi^2}}{a\sqrt{3}}, \tag{6}$$

so

$$\lambda_{so} \approx 1.8\left(\frac{Ze^2}{2mc^2 r_d}\right)\left(\frac{\hbar^2}{2ma^2}\right). \tag{7}$$

Ye *et al.* called the middle term the "dimensionless coupling constant appropriate for *d* orbitals," and the final term the "band kinetic energy."[7] This rough estimate of the spin-orbit coupling constant works out to be about 9 K for gadolinium.

Unlike CrO_2, where only those electrons that participate in the double-exchange contribute to the conductivity, Gd has both *s*- and *d*-electron contributions. It is not surprising, therefore, that the temperature dependence (below 160 K) appears to be dominated by side-jump processes ($R_s \propto \rho_{xx}^2$),[20] as seen in a plot of R_s vs ρ_{xx}^2 in Fig. 10. A side-jump contribution, presumably from those portions of the Fermi surface that are not strongly spin-polarized, should be distinguishable from the Skyrmion contributions, for which $R_s \propto e^{-E_c/(k_B T)}/(k_B T)$.[7,11] As a further complication, Lee and Legvold's data show a low-temperature sign change of the anomalous Hall coefficient at a temperature different from the temperature at which the ordinary Hall coefficient changes sign; neither side-jump nor Skyrmion models can account for this. Extrapolation of the contribution proportional to the square of the resistivity predicts a much larger Hall effect above 200 K than is observed. The ordinary Hall effect has been neglected when converting Lee and Legvold's data.[20] Berger's prediction for the side-jump contribution is independent of the potential, so it should be essentially material independent, except for the enhancement due to band effects.[4] Using the rough estimate calculated for iron (see the Appendix) gives a slope that is an order of magnitude too small for both iron and gadolinium.[5] The straight line in Fig. 10 has a coefficient that is one order of magnitude larger than this estimate. This coefficient is consistent in magnitude with experimental values for iron between 80 K and 267 K.[1,3,5] While this term fits the lower temperature data, it is clearly too large near T_C.

We next explore whether the anomalous Hall effect may be due to a combination of side-jump and Berry-phase processes. If we assume that the spin-orbit coupling constant is 9 K (our rough estimate), then Berry-phase effects can only account for five-sixths of the Hall effect at T_C. The remaining sixth can be accounted for by a small side-jump process contribution, i.e.,

$$R_s^{sj} = \frac{-254\ \Omega^{-1}\,\mathrm{cm}^{-1} \times \rho_{xx}^2}{\mu_0 M_0}. \tag{8}$$

The subtracted term is strictly linear in the magnetization, with a temperature dependence that depends on the square of

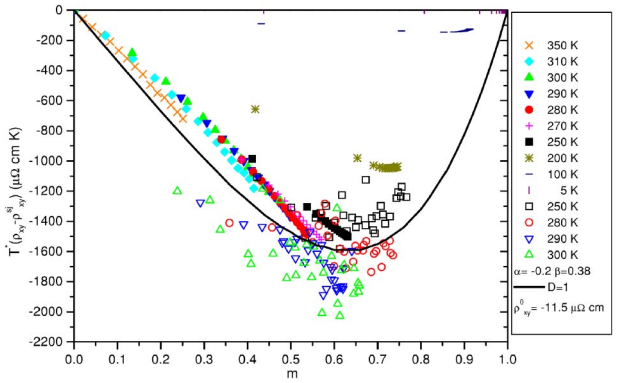

FIG. 11. (Color online) Possible Berry-phase contribution vs reduced magnetization. The estimated side-jump contribution accounts for one-sixth of the Hall effect at T_C.

the zero-field resistivity (excluding the residual resistivity). This side-jump contribution is of the same order of magnitude as expected theoretically, as shown in the Appendix. Figure 11 is a plot of the difference vs reduced magnetization, showing a reasonable collapse of the data at both low and high fields, with an extremum in the vicinity of $m=0.6$. Indeed, subtraction of a side-jump contribution of up to twice this size cannot be distinguished from Berry-phase only. Thus, we estimate the spin-orbit coupling constant to be between 8 K and 12 K depending on the relative contributions of Berry-phase and side-jump processes. The evidence for a decrease in the anomalous Hall effect at high fields is even more convincing after subtracting the conventional term. The line shown in Fig. 11 is the same as in Fig. 9, except the initial slope is reduced, and $D(0)=1$ has been chosen. This same value for $D(x)$ also provided a good fit for CrO_2.[11]

V. CONCLUSION

The data provide evidence for a Berry-phase contribution to the AHE. This contribution in combination with a side-jump term can account for the magnitude, magnetic-field dependence, and temperature dependence of the AHE near T_C. Unfortunately, fits to the temperature dependence of the low-temperature data cannot be used to resolve the exact magnitude of the Berry-phase term (as was possible for CrO_2),[11] because of the difficulty in separating the ordinary and anomalous Hall effects. Although the presence of the predicted decrease of the AHE at high magnetic fields is strongly suggested by these measurements of Gd and Yanagihara's measurements of CrO_2,[11] higher field measurements near the Curie temperatures should be able to remove any remaining doubts about the shape of the curve as the effect decreases.

ACKNOWLEDGMENTS

This paper is based upon work supported by the U.S. Department of Energy, Division of Materials Sciences under Grant No. DEFG02-91ER45439, through the Frederick Seitz Materials Research Laboratory at the University of Illinois at Urbana-Champaign. The authors would like to thank the National High Magnetic Field Laboratory in Tallahassee for the use of their facilities. Scott Bailey acknowledges support from the National Research Council.

APPENDIX: SKEW SCATTERING AND SIDE JUMP

More conventional explanations for the anomalous Hall effect include side-jump and skew scattering.[22] Side-jump scattering is when carriers scatter off impurities asymmetrically. Skew scattering is a process caused by interference between spin-orbit coupling and second-order spin-flip scattering.[7] In conventional ferromagnets, this theory yields values of R_s two orders of magnitude smaller than experimental data (according to some authors).[6,7] Since the carrier-electron spins must align with the localized core spins in double-exchange systems, spin-flip scattering cannot occur, and therefore skew scattering cannot explain the Hall effect in manganites and other systems with strong double exchange.

Karplus and Luttinger developed an early model for the anomalous Hall effect resulting from the spin-orbit interaction of spin-polarized conduction electrons.[23] Their model gave $R_s \propto \rho_{xx}^2$, but Smit criticized their model arguing that a periodic potential could not cause scattering and produce the anomalous Hall effect.[24] Smit's theory, known as skew scattering, is based on anisotropic scattering caused by the spin-orbit interaction.[25] After scattering off of an impurity, the momentum of the charge carriers is changed. Spin-orbit coupling makes scattering to one side more likely; this gives rise to the Hall effect. Skew scattering is generally distinguished by $R_s \propto \rho_{xx}$,[5] but can also give terms proportional to the square of the resistivity. The quadratic term occurs at high impurity concentrations (simultaneous scattering from multiple impurities) and from phonon scattering (at least above the Debye temperature).[25] Leribaux[2,5] estimates the phonon scattering contribution in iron as

$$R_s = \frac{20.9\ \Omega^{-1}\,\mathrm{cm}^{-1}}{\mu_0 M_s(T)} \rho_{xx}^2 [1 + T^2 \times 1.12 \times 10^{-8}\ \mathrm{K}^{-2}]. \tag{A1}$$

Somewhat later, Berger proposed the side-jump mechanism that yields $R_s \propto \rho_{xx}^2$.[4] The side-jump mechanism occurs when the center of mass of a carrier's wave packet is translated to the side while inside the scattering potential. The effect can be envisioned by picturing light striking a window at an angle. The refractive index of the window results in a displacement of the light's path but no change in direction because both glass/air interfaces are parallel. In general, this translation can be in any direction, but only asymmetric (due to the spin-orbit interaction) sideways jumps will directly contribute to the Hall effect. Klaffky and Coleman[3,5] estimate the side-jump scattering contribution in iron to be five times larger than the skew-scattering contribution [Eq. (A1)], and given by

$$R_s^{sj} = \frac{100\ \Omega^{-1}\,\mathrm{cm}^{-1}}{\mu_0 M_s} \rho_{xx}^2. \tag{A2}$$

Recently, the skew and side-jump mechanisms have been treated simultaneously using a model based on the Kubo formalism and the Dirac equation.[26] Experimental results for single-crystal iron show that the anomalous Hall coefficient is proportional to the square of the resistivity between 75 K and room temperature.[1,3,5] The experimental coefficients, which range from $9.3\times10^{2}\ \Omega^{-1}\,\mathrm{cm}^{-1}$ to $1.44\times10^{3}\ \Omega^{-1}\,\mathrm{cm}^{-1}$, are much larger than either estimate.[1,3,5] These results do not conclusively eliminate these mechanisms as the major source of the anomalous Hall effect, because the estimates are only valid to about one order of magnitude.

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