First Principles Calculation of Anomalous Hall Conductivity in Ferromagnetic bcc Fe

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We perform a first principles calculation of the anomalous Hall effect in ferromagnetic bcc Fe. Our theory identifies an intrinsic contribution to the anomalous Hall conductivity and relates it to the k-space Berry phase of occupied Bloch states. This dc conductivity has the same origin as the well-known magneto-optical effect, and our result accounts for experimental measurement on Fe crystals with no adjustable parameters.

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In ferromagnets the Hall resistivity, ρ_H, exhibits an anomalous contribution proportional to the magnetization of the material, in addition to the ordinary contribution proportional to the applied magnetic field, $\rho_H = R_0 B + R_s 4\pi M$ [1–3]. The anomalous Hall effect (AHE) has played an important role in the investigation and characterization of itinerant electron ferromagnets because R_s is usually at least one order of magnitude larger than the ordinary constant R_0. Although the effect has been recognized for more than a century [2], it is still somewhat poorly understood, a circumstance reflected by the controversial and sometimes confusing literature on the subject. Previous theoretical work has failed to explain the magnitude of the observed effect even in well understood materials like Fe [4].

Karplus and Luttinger [5] pioneered the theoretical investigation of this effect, by showing how spin-orbit coupling in Bloch bands can give rise to an anomalous Hall conductivity (AHC) in a perfect ferromagnetic crystal. This conclusion was questioned by Smit [6], who argued that R_s must vanish in a periodic lattice. Smit proposed an alternative mechanism, skew scattering, in which spin-orbit coupling causes spin polarized electrons to be scattered preferentially to one side by impurities. The skew-scattering mechanism predicts an anomalous Hall resistivity linearly proportional to the longitudinal resistivity; this is in accord with experiment in some cases, but an approximately quadratic proportionality is more common. Later, Berger [7] proposed yet another mechanism, the side jump, in which the trajectories of scattered electrons shift to one side at impurity sites because of spin-orbit coupling. The side jump mechanism does predict a quadratic dependence of the AHC on the longitudinal resistivity. However, because of inevitable difficulties in modeling impurity scattering in real materials, it has not been possible to compare quantitatively with experiment. It appears to us that the AHE has generally been regarded as an extrinsic effect due solely to impurity scattering, even though this notion has never been critically tested, and that the intrinsic contribution initially proposed by Karplus and Luttinger has been discounted.

Several years ago, the scattering free contribution of Karplus and Luttinger was rederived in a semiclassical framework of wave packet motion in Bloch bands by taking into account Berry phase effects [8,9]. According to this work, there is an AHC purely from the equilibrium distribution given by the sum of Berry curvatures [see Eqs. (2) and (6) below] of the occupied Bloch states [10]. Recently, Jungwirth *et al.* [11,12] applied this picture of the AHE to (III, Mn)V ferromagnetic semiconductors and found very good agreement with experiment. (III, Mn)V ferromagnets are unusual, however, because they are strongly disordered and have extremely strong spin-orbit interactions. In this Letter, we report on an evaluation of the intrinsic AHC in a classic transition metal ferromagnet, bcc Fe. Our calculation is based on spin-density functional theory and the linearized augmented plane wave method. The close agreement between theory and experiment that we find leads us to conclude that the AHC in transition metal ferromagnets is intrinsic in origin, except possibly at low temperature in highly conductive samples.

We begin our discussion by briefly reviewing the semiclassical transport theory. By including the Berry phase correction to the group velocity [8,9], one can derive the following equations of motion:

$$\dot{\mathbf{x}} = \frac{1}{\hbar}\frac{\partial \varepsilon_n(\mathbf{k})}{\partial \mathbf{k}} - \dot{\mathbf{k}} \times \mathbf{\Omega}_n(\mathbf{k}), \qquad (1)$$

$$\hbar\dot{\mathbf{k}} = -e\mathbf{E} - e\dot{\mathbf{x}} \times \mathbf{B},$$

where $\mathbf{\Omega}_n$ is the Berry curvature of the Bloch state defined by

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$$\mathbf{\Omega}_n(\mathbf{k}) = -\mathrm{Im}\langle\nabla_{\mathbf{k}} u_{n\mathbf{k}}| \times |\nabla_{\mathbf{k}} u_{n\mathbf{k}}\rangle, \tag{2}$$

with $u_{n\mathbf{k}}$ being the periodic part of the Bloch wave in the nth band. We are interested in the case of $\mathbf{B} = 0$, for which $\varepsilon_n(\mathbf{k})$ is just the band energy. The distribution function satisfies the Boltzmann equation with the usual drift and scattering terms, and can be written as $f_n(\mathbf{k}) + \delta f_n(\mathbf{k})$, where f_n is the equilibrium Fermi-Dirac distribution function and δf_n is a shift proportional to the electric field and relaxation time. The electric current is given by the average of the velocity over the distribution function, yielding to first order in the electric field [13]

$$-\frac{e^2}{\hbar}\mathbf{E} \times \int d^3k \sum_n f_n \mathbf{\Omega}_n(\mathbf{k}) - \frac{e}{\hbar}\int d^3k \sum_n \delta f_n(\mathbf{k}) \frac{\partial \varepsilon_n}{\partial \mathbf{k}}. \tag{3}$$

The first term is the anomalous Hall current originally derived by Karplus and Luttinger [5], and can lead to quantized Hall conductivity for full magnetic bands [9,14]. In the second term, apart from the longitudinal current, there can also be a Hall current in the presence of skew scattering because the distribution function can acquire a shift in the transverse direction. This shift will be small when normal scattering processes are strong which keep the distribution very close to equilibrium. This is the case for pure crystals at temperature ranges where phonon scattering dominates, because phonon scattering is known to have very little skew part [1]. At low temperatures, where impurity scattering dominates, there can be a significant skew part that shifts the distribution in the transverse direction. In general, because the shift is proportional to the relaxation time, the skew-scattering contribution to the Hall conductivity goes up linearly with the longitudinal conductivity. This contribution can thus be identified, when it is dominant, by the traditional test, $\rho_{xy} \propto \rho_{xx}$. On the other hand, the Berry curvature contribution to the Hall conductivity is independent of scattering, and should lead to a quadratic dependence, $\rho_{xy} \propto \rho_{xx}^2$.

We now discuss our scheme for calculating the Berry curvature and the AHC. For a cubic material with magnetization aligned along [001], only the z component $\Omega^z(\mathbf{k}) \neq 0$. In our calculation, we find it convenient to use a different but equivalent expression for the Berry curvature that arises naturally from the Kubo-formula derivation [14],

$$\Omega_n^z(\mathbf{k}) = -\sum_{n' \neq n} \frac{2\mathrm{Im}\langle\psi_{n\mathbf{k}}|v_x|\psi_{n'\mathbf{k}}\rangle\langle\psi_{n'\mathbf{k}}|v_y|\psi_{n\mathbf{k}}\rangle}{(\omega_{n'} - \omega_n)^2}, \tag{4}$$

where $E_n = \hbar\omega_n$, and v's are velocity operators. It is also instructive to introduce the sum (for each $\mathbf{k}$) of Berry curvatures over the occupied bands:

$$\Omega^z(\mathbf{k}) = \sum_n f_n \Omega_n^z(\mathbf{k}). \tag{5}$$

Then the intrinsic AHC is an integration over the Brillouin zone (BZ):

$$\sigma_{xy} = -\frac{e^2}{\hbar}\int_{\mathrm{BZ}} \frac{d^3k}{(2\pi)^3}\Omega^z(\mathbf{k}). \tag{6}$$

The recent development of highly accurate *ab initio* electronic structure calculation methods enables us to complete the work of Karplus and Luttinger by evaluating their intrinsic Hall conductivity and comparing it with experiment. We employ the full-potential linearized augmented plane-wave method [15] with the generalized gradient approximation (GGA) for the exchange-correlation potential [16]. Fully relativistic band calculations were performed using the program package WIEN2K [17]. A converged ground state with magnetization in the [001] direction was obtained using 20 000 $\mathbf{k}$ points in the first Brillouin zone and $K_{\max}R_{MT} = 10$, where R_{MT} represents the muffin-tin radius and $K_{\max}$ the maximum size of the reciprocal-lattice vectors. Wave functions and potentials inside the atomic sphere are expanded in spherical harmonics up to $l = 10$ and 4, respectively, and $3s$ and $3p$ semicore local orbitals are included in the basis set. The calculations were performed using the experimental lattice constant of 2.87 Å. The spin magneton number was found to be 2.226, compared to the experimental value of 2.12 as deduced from measurements of the magnetization [18] and of the g (=2.09) factor. The calculated energy bands are very similar to those obtained in Ref. [19]. If the spin-orbit interaction is parametrized as $\xi\,\mathbf{l}\cdot\mathbf{s}$, its strength ξ is found to be approximately 5.1 mRy from the band splitting near the H point and the Fermi energy.

After obtaining the self-consistent potential with 20 000 $\mathbf{k}$ points, we calculated the Berry curvature with several larger sets of $\mathbf{k}$ points in order to achieve the convergence for σ_{xy} shown in Fig. 1. The Monkhorst-Park special-point method [20] was used for the integration in Eq. (6). To go beyond 2×10^6 points, we adopted a method of adaptive mesh refinement; i.e., when $\Omega^z(\mathbf{k})$ is large at a certain $\mathbf{k}$ point, we construct a finer mesh by adding 26 additional points around it. This procedure yields a converged value of $\sigma_{xy} = 751\ (\Omega\,\mathrm{cm})^{-1}$ at zero temperature (using a step function for the Fermi-Dirac distribution) and a slightly smaller value of $\sigma_{xy} = 734\ (\Omega\,\mathrm{cm})^{-1}$ at room temperature (300 K). Our result is in fair agreement with the value $1032\ (\Omega\,\mathrm{cm})^{-1}$ extracted from Dheer's data on iron whiskers [21] at room temperature.

The slow convergence is caused by the appearance of large contributions of both signs to Ω^z which occur in very small regions of $\mathbf{k}$ space. Spin-orbit effects are small except when they mix states that are degenerate or nearly degenerate. A pair of mixed states contributes negligibly if both are occupied or are unoccupied. Therefore, only when the Fermi surface lies in a spin-orbit induced gap is

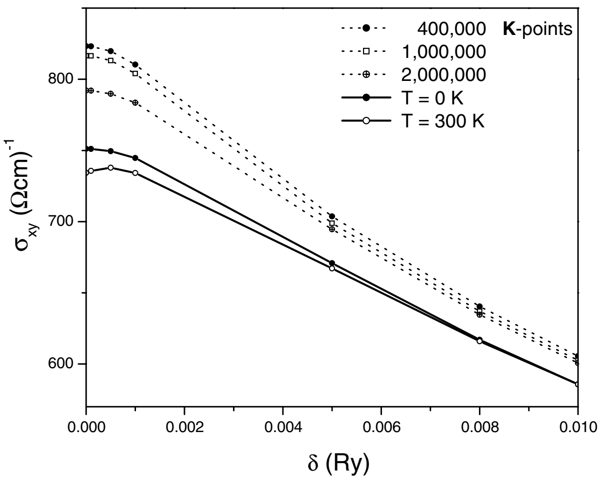

FIG. 1. Anomalous Hall effect vs δ with different numbers of **k** points in full Brillouin zone. Here δ is introduced by adding δ^2 to the denominator in Eq. (4). The dotted lines are obtained (for zero temperature) using a different number of **k** points. The solid lines are obtained by an adaptive mesh refinement method.

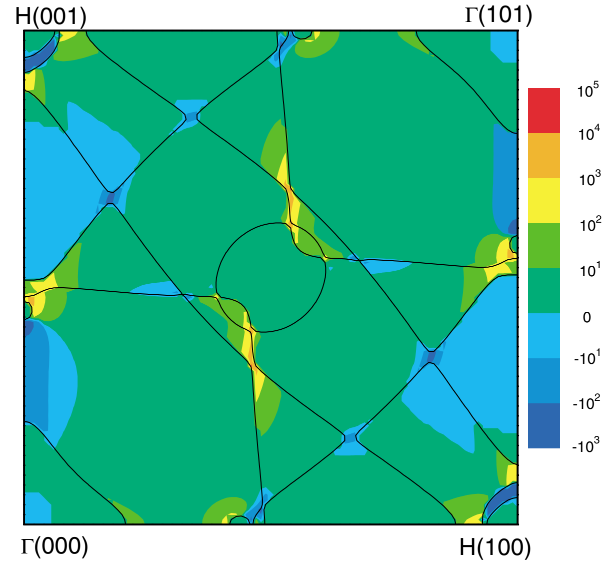

FIG. 3 (color). Fermi surface in (010) plane (solid lines) and Berry curvature $-\Omega^z(\mathbf{k})$ in atomic units (color map).

there a large contribution. For example, as shown in Fig. 2, the large spike near $H(1,0,0)$ in the direction of $P(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ is due to a pair of spin-orbit coupled bands, one occupied and one unoccupied in a small **k** interval. The small energy gap gives rise to a small energy denominator, making the contribution to the Berry curvature very large in this small interval. The largest peaks and valleys in the distribution of the total Berry curvature are, however, located off the **k**-space symmetry lines. For example, as can be seen in Fig. 3, the Berry curvature shows sharp peaks and valleys of several orders of magnitude in height and depth at general **k** points of the (010) plane.

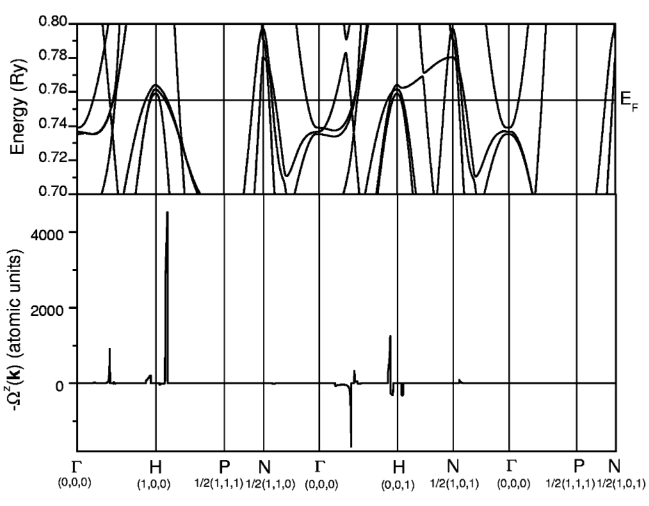

FIG. 2. Band structure near Fermi energy (upper panel) and Berry curvature $\Omega^z(\mathbf{k})$ (lower panel) along symmetry lines.

In order to further understand the role of spin-orbit coupling in the AHE, we artificially varied the speed of light, thereby changing the spin-orbit coupling strength $\xi \propto c^{-2}$. As shown in Fig. 4, σ_{xy} is linear in ξ for small coupling, but not for large coupling. For iron, nonlinearity becomes significant for $\xi/\xi_0 > 1/2$, which means that the spin-orbit interaction in iron cannot be accurately treated in a perturbative manner.

It is straightforward to extend our calculation to the ac Hall case by using the Kubo-formula [22] approach:

$$\sigma(\omega)_{xy} = \frac{e^2}{\hbar}\int_{V_G}\frac{d^3k}{(2\pi)^3}\sum_{n\neq n'}(f_{n,\mathbf{k}} - f_{n',\mathbf{k}}) \times \frac{\mathrm{Im}\langle\psi_{n\mathbf{k}}|v_x|\psi_{n'\mathbf{k}}\rangle\langle\psi_{n'\mathbf{k}}|v_y|\psi_{n\mathbf{k}}\rangle}{(\omega_{n'}-\omega_n)^2 - (\omega + i\delta)^2}, \tag{7}$$

where δ is a positive infinitesimal. In the upper panel in

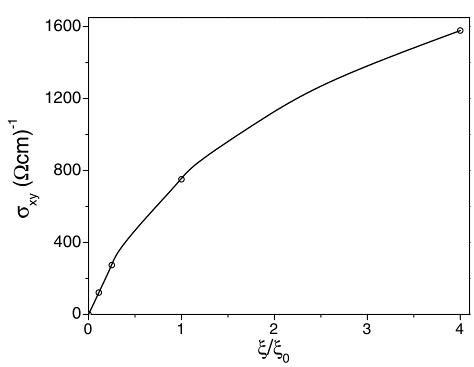

FIG. 4. Calculated anomalous Hall conductivity (open circles) vs the effective spin-orbit coupling strength relative to its value for iron. The line is a guide to the eye.

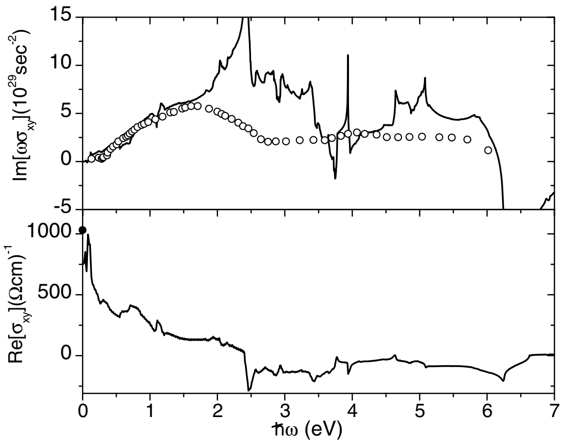

FIG. 5. Frequency dependence of the Hall conductivity at zero temperature. In the upper panel, the calculated imaginary part of $\omega\sigma_{xy}$ (solid curve) is compared with experimental results ○; see Ref. [23]. In the lower panel, the real part of σ_{xy} is shown together with the dc experiment value ● extracted from Ref. [21].

Fig. 5, we show results for the imaginary part of $\omega\sigma_{xy}$ as a function of frequency that are in agreement with earlier calculations [24]. Experimental results [23] are in excellent agreement below 1.7 eV but become smaller at higher energies. In the lower panel of the figure, the real part of the Hall conductivity, obtained from the imaginary part by a Kramers-Kronig relation, is shown as a function of frequency. The dc limit result, $\sigma(\omega = 0)_{xy} = 750.8\ (\Omega\,\mathrm{cm})^{-1}$, is essentially identical to that obtained from Eq. (6). Despite the small discrepancy with theory in the dc limit, the experimental point ● [21] seems to agree rather well with the overall trend of the frequency dependence of the calculated AHC.

In conclusion, we have shown that the AHC of bcc Fe, and presumably all other transition metal ferromagnets, is primarily intrinsic. [The only previous evaluation of the AHC of which we are aware [4] found that $\sigma_{xy} = 20.9\ (\Omega\,\mathrm{cm})^{-1}$ for Fe.] The remaining discrepancy between theory and experiment is likely due to shortcomings of the GGA, neglect of scattering effects, and experimental uncertainties.

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Note added.—After submission we became aware of two recent works on Kubo-formula evaluation of the anomalous Hall conductivity in ferromagnetic crystals with similar conclusions [25].

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