Intrinsic anomalous Hall effect in nickel: An GGA+U study

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The electronic structure and intrinsic anomalous Hall conductivity of nickel have been calculated based on the generalized gradient approximation (GGA) plus on-site Coulomb interaction (GGA+U) scheme. The highly accurate all-electron full-potential linearized augmented plane wave method is used. It is found that the intrinsic anomalous Hall conductivity (σ_{xy}^{H}) obtained from the GGA+U calculations with U = 1.9 eV and J = 1.2 eV, is in nearly perfect agreement with that measured recently at low temperatures while, in contrast, the σ_{xy}^{H} from the GGA calculations is about 100 % larger than the measured one. This indicates that, as for the other spin-orbit interaction (SOI)-induced phenomena in 3d itinerant magnets such as the orbital magnetic magnetization and magnetocrystalline anisotropy, the on-site electron-electron correlation, though moderate only, should be taken into account properly in order to get the correct anomalous Hall conductivity. The intrinsic σ_{xy}^{H} and the number of valence electrons (N_v) have also been calculated as a function of the Fermi energy (E_F). A sign change is predicted at E_F = -0.38 eV (N_v = 9.57), and this explain qualitatively why the theoretical and experimental σ_{xy}^{H} values for Fe and Co are positive. It is also predicted that fcc Ni_{1-x}Co(Fe,Cu)_x alloys with x being small, would also have the negative σ_{xy}^{H} with the magnitude being in the range of 500 ~ 1400 Ω^{-1} cm^{-1}. The most pronounced effect of including the on-site Coulomb interaction is that all the d-dominant bands are lowered in energy relative to the E_F by about 0.3 eV, and consequently, the small minority spin X₂ hole pocket disappears. The presence of the small X₂ hole pocket in the GGA calculations is attributed to be responsible for the large discrepancy in the σ_{xy}^{H} between theory and experiment.

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I. INTRODUCTION

Anomalous Hall effect (AHE) refers to the transverse charge current generation in solids in a ferromagnetic phase by the electric field and has received intensive renewed interest in recent years mainly because of its close connection with spin transport phenomena\textsuperscript{1}. There are several competing mechanisms proposed for the AHE. Extrinsic mechanisms of skew scattering\textsuperscript{2} and side jump\textsuperscript{3} refer to the modified impurity scattering caused the spin-orbit interaction (SOI). Another mechanism arises from the transverse velocity of the Bloch electrons induced by the SOI, discovered by Karplus and Luttinger\textsuperscript{4}, and thus is of intrinsic nature. This intrinsic AHE has recently been reinterpreted in terms of the Berry curvature of the occupied Bloch states.\textsuperscript{5,8} Furthermore, recent quantitative first-principles studies based on the Berry phase formalism showed that the intrinsic AHE is important in various materials.\textsuperscript{9} In particular, in itinerant ferromagnets such as Fe and Co, the intrinsic anomalous Hall conductivity given by first-principles calculations\textsuperscript{7,10} has been found to agree with the experimental anomalous Hall conductivity\textsuperscript{11,13} within 30 %, thereby demonstrating the dominance of the intrinsic mechanism. Nonetheless, the physical origin of the AHE in nickel is still not fully understood. Recent first-principles density functional calculations with the generalized gradient approximation (GGA) predicted a large intrinsic anomalous Hall conductivity of -2203 Ω^{-1} cm^{-1} in Ni\textsuperscript{10}, which is more than three times larger than the corresponding experimental value of -646 Ω^{-1} cm^{-1}\textsuperscript{14} at room temperature. In the latest experiment\textsuperscript{15}, the intrinsic Hall conductivity was found to be -1100 Ω^{-1} cm^{-1} at low temperatures. Though this value is significantly larger than that of the much earlier experiment\textsuperscript{14}, it is still only half of the calculated intrinsic Hall conductivity\textsuperscript{10}.

First-principles GGA calculations have been rather successful in describing many physical properties such as crystal structure, elastic constant and spin magnetic moment, of itinerant ferromagnets Fe, Co and Ni (see, e.g., Ref. 16 and references therein). However, GGA calculations fall in describing some relativistic SOI-induced phenomena in these itinerant magnets. For example, the theoretical values of orbital magnetic moment account for only about 50 % of the measured ones in Fe and Co\textsuperscript{17} and the calculated magnetocrystalline anisotropy energy of Ni is even wrong in sign\textsuperscript{18}. This failure of the GGA is generally attributed to its incorrect treatment of the moderate 3d electron-electron correlation in these systems. Several theoretical methods that go beyond the density functional theory (DFT) with the local density approximation (LDA) or GGA, such as the orbital-polarization correction\textsuperscript{19} and LDA/GGA plus on-site Coulomb interaction U (LDA/GGA+U)\textsuperscript{20,22} schemes, have been developed for better description of...
the SOI-induced phenomena in magnetic solids. Indeed, the orbital-polarization correction has been found to bring the calculated orbital moments in many itinerant magnets such as Fe and Co in good agreement with experiments. Furthermore, it has been demonstrated that the correct easy axes and the magnitudes of the magnetocrystalline anisotropy energy of Fe and Ni can be obtained within the LDA+U scheme.

Therefore, in this work, we perform GGA+U calculations for nickel to better understand the mechanism of its anomalous Hall effect. We use highly accurate all-electron full-potential linearized augmented plane wave (FLAPW) method. We find that including on-site electron-electron correlation in nickel has significant effect on its anomalous Hall effect as well as its electronic structure near the Fermi level. In particular, the calculated anomalous Hall conductivity of a solid comes from the static limit (can be evaluated by using the Kubo formula). The intrinsic Hall effect on its anomalous Hall effect as well as its electronic

**II. THEORY AND COMPUTATIONAL DETAILS**

The intrinsic anomalous Hall conductivity of a solid is calculated within the linear-response Kubo formalism as well as the numerical method and computational details used in the present work. In Sec. III, we first present the calculated anomalous Hall conductivity, magnetic moments and also relativistic band structure. We then compare our results with available experiments and also previous calculations. Finally, we make some predictions about the intrinsic anomalous Hall conductivity for fcc Ni by taking the variation of the exchange correlation potential. To further take d-electron correlation into account, we include on-site Coulomb interaction U in the GGA+U approach. The so-called around-mean-field (AMF) scheme for double counting correction is adopted here. U = 1.9 eV and J = 1.2 eV, which were found to give the correct sign and magnitude of magnetocrystalline anisotropy energy for fcc Ni, are used. Furthermore, the GGA+U calculations using the double counting correction scheme designed for approximate self-interaction correction (SIC) for strongly correlated systems such as transition metal oxides, have also been performed. Nevertheless, the theoretical anomalous Hall conductivities from the GGA+U calculations using both double counting correction schemes are almost identical (see Table I below). Since the AMF scheme is believed to be more suitable for metallic systems, here we will concentrate on the results of the AMF calculations. Moreover, the GGA+U calculations with a larger U value of 2.5 eV are also performed to see how the variation of U may affect the calculated anomalous Hall conductivity.

The experimental lattice constant a = 3.52 Å is used here. The muffin-tin sphere radius (Rmt) used is 2.2 a.u. The wave function, charge density, and potential were expanded in terms of the spherical harmonics inside the muffin-tin spheres, and the cutoff angular momentum (Lmax) used is 10, 6, and 6, respectively. The wave function outside the muffin-tin spheres was expanded in terms of the augmented plane waves, and a large number of augmented plane waves (about 70 APWs per atom, i.e., the maximum size of the crystal momentum Kmax = 8/Rmt) were included in the present calculations. The improved tetrahedron method is used for the Brillouin-zone integration. To obtain accurate ground state charge density as well as spin and orbital magnetic moments, a fine 56 × 56 × 56 grid of 185193 k-points in the first Brillouin zone was used.

Then obtain the real part from the imaginary part by a Kramers-Kronig transformation

\[
\sigma_{xy}^{(1)}(\omega) = -\frac{2}{\pi} \int_0^{\infty} d\omega' \omega' \sigma_{xy}^{(2)}(\omega') \left( \frac{\omega}{\omega^2 - \omega'^2} \right)
\]

where P denotes the principal value of the integral. The intrinsic anomalous Hall conductivity \( \sigma_{xy}^{(1)} \) is the static limit of the off-diagonal element of the optical conductivity \( \sigma_{xy}^{(2)}(\omega=0) \). We notice that the anomalous Hall conductivity of bcc Fe calculated in this way is in quantitative agreement with that calculated directly by accounting for the Berry phase correction to the group velocity.
In the present work, the magnetization is assumed to be along the [001] direction, i.e. the easy axis. Therefore, the energy bands near the Z point are slightly different from that near the X and Y points, as shown in Fig. 2 and Fig. 3. The relativistic band structure may be regarded as the result of a superposition of the corresponding scalar-relativistic spin-up and spin-down band structures with many accidental band-crossings (degeneracies) lifted by the SOI. In nickel, nevertheless, these SOI-induced splittings are generally much smaller than the experimental values by Ye et al. [15] and the experimental value by Ye et al. [16].

To help understand how the on-site electron-electron correlation affects the electronic band structure and AHE in nickel, we plot in Fig. 2 and Fig. 3 the relativistic energy bands along the high symmetry lines in the Brillouin zone calculated both without and with on-site Coulomb interaction U. The relativistic band structure may be regarded as the result of a superposition of the corresponding scalar-relativistic spin-up and spin-down band structures with many accidental band-crossings (degeneracies) lifted by the SOI. In nickel, nevertheless, these SOI-induced splittings are generally much smaller than the experimental values by Ye et al. [15] and the experimental value by Ye et al. [16].

Theoretical calculations of the magnetocrystalline anisotropy energy of bulk magnets [18], a very fine k-point mesh is needed for the anomalous Hall conductivity calculation. Therefore, we perform the present calculations using several extremely fine k-point meshes with the finest k-point mesh being 106 × 106 × 106. The calculated σ_{xy}^{H} is plotted as a function of the inverse of the number (N_k) of k-points in the first Brillouin zone in Fig. 1. The calculated values of σ_{xy}^{H} are fitted to a polynomial to get the extrapolated value of σ_{xy}^{H} at N_k = 0. For comparison, the theoretical value by Wang et al. [11] and the experimental value by Ye et al. [15] are also shown as the horizontal dotted and dot-dashed lines, respectively.

III. RESULTS AND DISCUSSION

Like the calculation of the magnetocrystalline anisotropy energy of bulk magnets [18], the calculated anomalous Hall conductivity σ_{xy}^{H} as a function of the inverse of the number of k-points in the Brillouin zone (N_k). The dashed lines are a polynomial fit to the calculated values to get the extrapolated value of σ_{xy}^{H} at N_k = 0. For comparison, the theoretical value by Wang et al. [11] and the experimental value by Ye et al. [15] are also shown as the horizontal dotted and dot-dashed lines, respectively.

![Graph](image)

**FIG. 1.** (Color online) Calculated anomalous Hall conductivity σ_{xy}^{H} (Ω⁻¹·cm⁻¹) as well as spin magnetic moment m_s (μB/atom) and orbital magnetic moment m_o (μB/atom). Superscripts of SIC indicate that the values are obtained from the GGA+U calculations with the SIC double counting correction scheme (see Sec. II). The corresponding experimental values as well as previous theoretical σ_{xy}^{H} are also listed for comparison.

| Method         | GGA | GGA+U U=1.9eV | GGA+U U=2.5eV | Expt. |
|----------------|-----|---------------|---------------|-------|
| σ_{xy}^{H}     | -2200 | -1066         | -960          | -646  |
| m_s            | 0.639 | 0.661         | 0.675         | 0.57d |
| m_o            | 0.051 | 0.066         | 0.071         | 0.05d |

*References: [14] [10] [15] [16] [32].

Table I lists the calculated anomalous Hall conductivity σ_{xy}^{H} (Ω⁻¹·cm⁻¹) as well as spin magnetic moment m_s (μB/atom) and orbital magnetic moment m_o (μB/atom) along with the experiments (Table I). Nonetheless, with U = 1.9 eV and J = 1.2 eV, the theoretical spin and orbital magnetic moments are still in reasonable agreement with the experiments (Table I).

The theoretical and experimental values of σ_{xy}^{H}, m_s, and m_o are listed in Table I. Also listed in Table I are the available experimental and previous theoretical σ_{xy}^{H}, m_s, and m_o.

Table I shows that the theoretical σ_{xy}^{H} from the GGA is -2200 Ω⁻¹·cm⁻¹, and is much larger than the experimental value of -646 Ω⁻¹·cm⁻¹ at room temperature [14] and also the experimentally derived intrinsic value of -1100 Ω⁻¹·cm⁻¹ at low temperatures [13]. Nevertheless, the present result is in very good agreement with the previous GGA calculations [10], as it should be (see Table I and Fig. 1). Interestingly, when the on-site Coulomb interaction is taken into account via the GGA+U scheme, the calculated σ_{xy}^{H} reduces significantly. In particular, when U = 1.9 eV and J = 1.2 eV which were found to give rise to the correct easy axis and magnitude of the magnetocrystalline anisotropy energy [22], the theoretical σ_{xy}^{H} becomes -1066 Ω⁻¹·cm⁻¹. This good agreement between the present GGA+U calculation and the low temperature measurements [13] (Table I and Fig. 1) indicates that the intrinsic AHE dominates in nickel and that the 3d electron-electron correlation in itinerant magnets such as nickel has an important effect on the AHE, although being only moderate. Further increasing U to 2.5 eV reduces the calculated σ_{xy}^{H} slightly (Table I and Fig. 1). As expected, the GGA+U calculations increase the theoretical spin and orbital magnetic moments, and hence enlarge somewhat the discrepancies between the calculations and experiments (Table I). Nonetheless, with U = 1.9 eV and J = 1.2 eV, the theoretical spin and orbital magnetic moments are still in reasonable agreement with the experiments (Table I).

Theoretical calculations of the magnetocrystalline anisotropy energy of bulk magnets [18], a very fine k-point mesh is needed for the anomalous Hall conductivity calculation. Therefore, we perform the present calculations using several extremely fine k-point meshes with the finest k-point mesh being 106 × 106 × 106. The calculated σ_{xy}^{H} is plotted as a function of the inverse of the number (N_k) of k-points in the first Brillouin zone in Fig. 1. The calculated values of σ_{xy}^{H} are fitted to a polynomial to get the extrapolated value of σ_{xy}^{H} at N_k = 0. For comparison, the theoretical value by Wang et al. [11] and the experimental value by Ye et al. [15] are also shown as the horizontal dotted and dot-dashed lines, respectively.

**III. RESULTS AND DISCUSSION**

Like the calculation of the magnetocrystalline anisotropy energy of bulk magnets [18], a very fine k-point mesh is needed for the anomalous Hall conductivity calculation [9, 10]. Therefore, we perform the present result is in very good agreement with the previous GGA calculations [10], as it should be (see Table I and Fig. 1). Interestingly, when the on-site Coulomb interaction is taken into account via the GGA+U scheme, the calculated σ_{xy}^{H} reduces significantly. In particular, when U = 1.9 eV and J = 1.2 eV which were found to give rise to the correct easy axis and magnitude of the magnetocrystalline anisotropy energy [22], the theoretical σ_{xy}^{H} becomes -1066 Ω⁻¹·cm⁻¹. This good agreement between the present GGA+U calculation and the low temperature measurements [13] (Table I and Fig. 1) indicates that the intrinsic AHE dominates in nickel and that the 3d electron-electron correlation in itinerant magnets such as nickel has an important effect on the AHE, although being only moderate. Further increasing U to 2.5 eV reduces the calculated σ_{xy}^{H} slightly (Table I and Fig. 1). As expected, the GGA+U calculations increase the theoretical spin and orbital magnetic moments, and hence enlarge somewhat the discrepancies between the calculations and experiments (Table I). Nonetheless, with U = 1.9 eV and J = 1.2 eV, the theoretical spin and orbital magnetic moments are still in reasonable agreement with the experiments (Table I).

To help understand how the on-site electron-electron correlation affects the electronic band structure and AHE in nickel, we plot in Fig. 2 and Fig. 3 the relativistic energy bands along the high symmetry lines in the Brillouin zone calculated both without and with on-site Coulomb interaction U. The relativistic band structure may be regarded as the result of a superposition of the corresponding scalar-relativistic spin-up and spin-down band structures with many accidental band-crossings (degeneracies) lifted by the SOI. In nickel, nevertheless, these SOI-induced splittings are generally much smaller than the exchange splittings, and thus can be treated as a perturbation [33]. Also, including the SOI would lower the symmetry of the system. In the present work, the magnetization is assumed to be along the [001] direction, and the symmetry of the system becomes the tetragonal one. The six high symmetry X points which are equivalent in the nonrelativistic (or scalar-relativistic) case, now form two inequivalent groups, namely, two equivalent ± Z points and four equivalent ± X and ± Y points. Therefore, the energy bands near the Z point are slightly different from that near the X and Y points, as shown in Fig. 2 and Fig. 3. The relativistic band structure and Fermi surface of Ni has been reported by several researchers before by using different band structure calculation methods (see, e.g., Refs. [11, 18, 23, and 33]. In
particular, Wang and Callaway analysed in detail the energy band characters and Fermi surface sheets. The present GGA relativistic band structure (the red dashed curves in Fig. 2 and Fig. 3) is very similar to that reported by Wang and Callaway. For example, in both cases, there are five bands (bands 8-12) crossing the Fermi level, and there is a small down-spin-dominant hole pocket (band 8, noted as X2) centered at the X(Y,Z) symmetry point (see Figs. 2-3, and also Fig. 1 in Ref. 33). However, the small X2 hole pocket was not found in the de Haas-van Alphen experiments.

Now let us focus on the changes in the relativistic band structure caused by including the on-site Coulomb interaction. A pronounced change is that all the d-dominant bands are lowered in energy relative to the Fermi level, when the on-site Coulomb interaction is taken into account in the GGA+U scheme (see Figs. 2-3). In other words, the binding energy of the d-dominated valence bands is increased by about 0.3 eV. In particular, the down-spin band 8 is now pushed completely below the Fermi level calculated without the on-site Coulomb interaction, giving rise to a pronounced contribution to the anomalous Hall conductivity, and thus the absence of the X2 hole pocket in the GGA+U band structure is also the main cause for the significant reduction of the calculated anomalous Hall conductivity.

In Fig. 3, we display the anomalous Hall conductivity \(\sigma_{xy}^H\) and the number of valence electrons \(N_e\) as a function of the Fermi level \(E_F\), together with the relativistic band structure, from the GGA+U calculations using \(U = 1.9\) eV and \(J = 1.2\) eV. The fine k-point mesh of 106 x 106 x 106 is used. Clearly, the magnitude of the \(\sigma_{xy}^H\) peaks just above the true Fermi level \((E_F = 0\) eV\), with a large value of -1.420 \(\Omega^{-1}\) cm\(^{-1}\) at 0.16 eV \((N_e = \sim 10.3)\). The peak may be related to the flat band (band 10) along the W-L-\(\Gamma\) line near the \(L\) point just above the Fermi level (see Fig. 3a). As the Fermi level is further artificially raised, the size of the \(\sigma_{xy}^H\) decreases steadily and becomes rather small (within 200 \(\Omega^{-1}\) cm\(^{-1}\)) above 0.75 eV. When the \(E_F\) is artificially lowered, the magnitude of the \(\sigma_{xy}^H\) initially decreases gradually, and then drops sharply starting at \(E_F = -0.35\) eV. The resultant shoulder at -0.35 eV can be attributed to the presence of the top segment of band 8 near the \(X\) \((Y,Z)\) point in this energy region (see Fig. 3a). The \(\sigma_{xy}^H\) then changes its sign at -0.38 eV \((N_e = \sim 9.57)\). As the \(E_F\) is further lowered, the \(\sigma_{xy}^H\) increases sharply and then peaks at -0.74 eV \((N_e = \sim 8.90)\) with a large value of 2635 \(\Omega^{-1}\) cm\(^{-1}\). Beyond this point, the \(\sigma_{xy}^H\) decreases and fluctuates but remains positive as the \(E_F\) is further lowered, and it then changes its sign again at -2.28 eV \((N_e = \sim 4.78)\). For the \(E_F\) being below -3.80 \(\Omega^{-1}\) at -2.28 eV, the \(\sigma_{xy}^H\) is small. Note that the energy bands below -4.0 eV and above 0.5 eV are predominantly of 4s4p character and thus the effects of the SOI and exchange interaction are small.

Experimentally, the measured anomalous Hall resistivity \(\rho_{xy}\) is often analyzed in terms of two distinctly different resistivity \(\rho_{xx}\)-dependent terms\(^{[1]}\), i.e., \(\rho_{xy} = \alpha \rho_{xx} + b \rho_{xx}^2\). Since usually \(\rho_{xy} \ll \rho_{xx}\), the total anomalous Hall conductivity \(\sigma_{xy} = -\rho_{xy}/(\rho_{xx}^2 + \rho_{xx}^2) \approx \alpha \rho_{xx} + b\),
recent experimental finding of the negligible $\sigma_{xy}$ in fcc Fe.$^{[35]}$ The second scattering-independent term $b$ can be several times larger than the $b$, and can also differ in sign.$^{[35]}$ The second scattering-independent term $b$ was usually further separated into the intrinsic contribution $\sigma_{xy}^I$, which can be obtained from band structure calculations,$^{[1]}$ as done here for nickel, and also the extrinsic side jump mechanism ($\sigma_{xy}^{SJ}$). Therefore, when one compares the calculated $\sigma_{xy}^H$ with the experimental scattering-independent term $b$, one should be aware of the possible side jump contribution $\sigma_{xy}^{SJ}$ even though it has been shown that the $\sigma_{xy}^H$ would dominate in ferromagnet metals such as Fe and Co.$^{[4,10]}$ Recent theoretical calculations for the 2D Rashba and 3D Luttinger Hamiltonians using a Gaussian disorder model potential suggested that the AHE in the (III,Mn)V ferromagnetic semiconductors at low temperatures could be dominated by the $\sigma_{xy}^{SJ}$. However, more recent $ab$ initio calculations show that in fcc Fe$_{1-x}$Pd$_x$ and Ni$_{1-x}$Pd$_x$, the $\sigma_{xy}^{SJ}$ is generally two-order of magnitude smaller than both the $\sigma_{xy}^{SK}$ and $\sigma_{xy}^H$, thus being negligible.$^{[35]}$ This is consistent with the recent experimental finding of the negligible $\sigma_{xy}^{SJ}$ in bcc Fe$^{[13]}$ and fcc Ni$^{[13]}$ using a newly established empirical $\sigma_{xy}$-scaling formula for $\sigma_{xy}^{SK}$.$^{[3]}$

Let us now return to the calculated $\sigma_{xy}^H$ as a function of the $E_F$ described above and also presented in Fig. 3. We find that it can explain qualitatively within the rigid band approximation the known AHE experiments on 3d transition metal ferromagnets and their alloys.$^{[11-14]}$ For example, the sign of the $\sigma_{xy}^H$ of both Fe and Co were found to be positive.$^{[11,13]}$ Interestingly, the calculated $\sigma_{xy}^H$ at $N_e = 8.0$ ($E_F = -1.02$ eV) is $758 \, \Omega^{-1} \text{cm}^{-1}$, being in very good agreement with the previous $ab$ initio calculations for Fe$^{[9]}$. Of course, this nearly perfect agreement may be accidental because Fe crystallizes in the bcc structure and also has a much larger spin magnetic moment, and hence the rigid band model should not work very well here. We note that the calculated $\sigma_{xy}^H$ is $2548 \, \Omega^{-1} \text{cm}^{-1}$ at $N_e = 9.0$ ($E_F = -0.71$ eV), which is much larger than the previous theoretical $\sigma_{xy}^H$ value for fcc Co$^{[37]}$ and also the experimental $\sigma_{xy}^H$ value for polycrystalline hcp Co$^{[38]}$. Nickel forms substitutional alloys in fcc structure with low concentration of Fe, Co and Cu. The present calculations (Fig. 3) predict that fcc Ni$_{1-x}$Co(Fe,Cu)$_x$ alloys (x being small) would have the negative intrinsic $\sigma_{xy}^H$ with the magnitude being in the range of $500 \sim 1400 \, \Omega^{-1} \text{cm}^{-1}$. Of course, when comparing this prediction with the AHE experiments on fcc Ni$_{1-x}$Co(Fe,Cu)$_x$ alloys, one should take into account the non-negligible skew scattering contribution $\sigma_{xy}^{SK}$.$^{[32]}$ Indeed, as mentioned earlier, Fig. 3b shows a sign change of $\sigma_{xy}^H$ at $N_e \approx 9.570$ ($E_F = -0.38$ eV), whereas the sign change of the $\sigma_{xy}$ in fcc Ni$_{1-x}$Fe$_x$ alloys with $x \approx 0.13$ (i.e., $N_e \approx 9.74$) was experimentally found$^{[2]}$. This experimentally found sign change at the lower Fe concentration could be attributed to the presence of the positive $\sigma_{xy}^{SK}$ in fcc Ni$_{1-x}$Fe$_x$ alloys.

As mentioned before, the most pronounced effect of including the on-site Coulomb interaction is that all the
In fact, Fig. 3 shows that as the segment of band 8 near the X(Y,Z) point in this en-
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E
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σxy exhibits a pronounced negative peak right at the
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F with the value being about -2178 Ω
−1cm−1 (see Fig. 3b).

The remarkable difference in the
σxy at 0.0 eV from the GGA and GGA+U calculations can be attributed to the presence of the above mentioned X2 hole pocket in the GGA calculation (Fig. 3). Another major difference is the absence of the clear shoulder at -0.35 eV in the GGA σxy spectrum. This is because of the absence of the top segment of band 8 near the X(Y,Z) point in this energy region in the GGA band structure (see Fig. 3a).

In fact, Fig. 3 shows that as the
E F is lowered from the true Fermi level (0 eV), the GGA σxy decreases in magnitude steeply to zero and changes its sign at -0.30 eV (Nxc = 9.55). Nevertheless, in both cases, the σxy changes its sign near Nxc = 9.56. The other pronounced difference (by about 700 Ω
−1cm−1) in the two calculated σxy spectra is the height of the positive peak near -0.70 eV. In the rest of the energy region, the two σxy spectra look rather similar.

IV. CONCLUSIONS

In summary, we have calculated the electronic structure and intrinsic anomalous Hall conductivity of nickel with both the GGA and GGA+U schemes. The highly accurate all-electron FLAPW method is used. We find that the theoretical anomalous Hall conductivity (σxy) obtained from the GGA calculations (Table I) is about 100 % larger than the intrinsic σxy recently measured at low temperatures. In contrast, the theoretical σxy from the GGA+U calculations with U = 1.9 eV and J = 1.2 eV is in almost perfect agreement with the measured one (Table I). This indicates that, as for other SOI-induced magnetic phenomena in 3d itinerant magnets such as the orbital magnetic magnetization and magnetocrystalline anisotropy, the on-site electron-electron correlation, though moderate only, should be taken into account properly in order to get the correct anomalous Hall conductivity. The most significant effect of including the on-site Coulomb interaction is that all the d-dominant bands are lowered in energy relative to the
E F by about 0.3 eV. Consequently, the down-spin band 8 is now pushed completely below the
E F and the small X2 hole pocket disappears. The presence of the small X2 hole pocket in the LDA and GGA calculations is found to be the main reason why the large discrepancy in the σxy between theory and experiment exists. The intrinsic anomalous Hall conductivity (σxy) has also been calculated as a function of the Fermi level
E F (or the number of valence electrons Nxc) in the GGA+U scheme. A sign change is predicted at
E F = -0.38 eV (or Nxc = 9.57), and this explain qualitatively why the theoretical and experimental σxy values for Fe and Co are positive. Finally, the present calculations (Fig. 3) indicate that fcc Ni(1−x)Co(Fe,Cu)x alloys (x being small) would also have the negative intrinsic σxy with the magnitude being in the range of 500 ~ 1400 Ω
−1cm−1.

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[1] N. Nagaosa, J. Sinova, S. Onoda, A. H. MacDonald, and N. P. Ong, Rev. Mod. Phys. 82, 1539 (2010)
[2] J. Smit, Physica (Amsterdam) 21, 877 (1955)
[3] L. Berger, Phys. Rev. B 2, 4559 (1970)
[4] R. Karplus and J. M. Luttinger, Phys. Rev. 95, 1154 (1954); J. M. Luttinger, Phys. Rev. 112, 739 (1958)
[5] M.-C. Chang and Q. Niu, Phys. Rev. B 53, 7010 (1996); G. Sundaram and Q. Niu, ibid. 59, 14915 (1999)
[6] T. Jungwirth, Q. Niu, and A. H. MacDonald, Phys. Rev. Lett. 88, 207208 (2002)
[7] M. Onoda and N. Nagaosa, J. Phys. Soc. Jpn. 71, 19 (2002)
[8] D. Xiao, M.-C. Chang, and Q. Niu, Rev. Mod. Phys. 82, 1959 (2010)
[9] Y. Yao, L. Kleinman, A. H. MacDonald, J. Sinova, T. Jungwirth, D.-S. Wang, E. Wang, and Q. Niu, Phys. Rev. Lett. 92, 037204 (2004)
[10] X. Wang, D. Vandervilt, J. R. Yates, and I. Souza, Phys. Rev. B 76, 195109 (2007)
[11] P. N. Dheet, Phys. Rev. 156, 637 (1967)
[12] T. Miyasato, N. Abe, T. Fujii, A. Asamitsu, S. Onoda, Y. Onose, N. Nagaosa, and Y. Tokura, Phys. Rev. Lett. 99, 086602 (2007)
[13] Y. Tian, L. Ye, and X. F. Jin, Phys. Rev. Lett. 103, 087206 (2009)
[14] J. M. Lavine, Phys. Rev. 123, 1273 (1961)
[15] L. Ye, Y. Tian, X. F. Jin, and D. Xiao, arXiv:1105.5664
[16] G. Y. Guo and H. H. Wang, Chin. J. Phys. 38, 949 (2000)
[17] G. Y. Guo, Phys. Rev. B 55, 11619 (1997)
[18] G. Y. Guo, W. M. Temmerman, and H. Ebert, Physica B 172, 61 (1991)
[19] M. S. S. Brooks, Physica B B 130, 6 (1985)
[20] V. I. Anisimov, J. Zaanen, and O. K. Andersen, Phys. Rev. B 44, 943 (1991)
[21] V. I. Anisimov, I. V. Solovyev, M. A. Korotin, M. T. Czyzyk and G. A. Sawatzky, Phys. Rev. B 48, 16929 (1993)
[22] M. T. Czyzyk and G. A. Sawatzky, Phys. Rev. B 49, 14211 (1994)
[23] I. Yang, S. Y. Savrasov, and G. Kotliar, Phys. Rev. Lett. 87, 216405 (2001)
[24] O. K. Andersen, Phys. Rev. B 12, 3060 (1975)
[25] M. Marder, Conddensed Matter Physics (John Wiley, Sons, Inc., New York, 2000).
[26] G. Y. Guo, Y. Yao, and Q. Niu, Phys. Rev. Lett. 94, 226601 (2005)
[27] G. Y. Guo and H. Ebert, Phys. Rev. B 51, 12633 (1995)
[28] P. Blaha, K. Schwarz, G. Madsen, D. Kvasnicka, and J. Luitz, WIEN2K, An Augmented Plane Wave Local Or-
bitals Program for Calculating Crystal Properties (Technische University Wien, Austria, 2002)
[29] J. P. Perdew, K. Burke, and M. Ernzerhof, Phys. Rev. Lett. 77, 3865 (1996)
[30] C. Kittel, Introduction to Solid State Physics, Seven edition
[31] P. E. Blöchl, O. Jepsen and O. K. Andersen, Phys. Rev. B 49, 16223 (1994)
[32] Landolt-Börnstein, New Series, Vol. III/19a, edited by H. P. J. Wein (Springer, Berlin 1988)
[33] C. S. Wang and J. Callaway, Phys. Rev. B 9, 4897 (1974)
[34] D. C. Tsui, Phys. Rev. 164, 669 (1967)
[35] S. Lowitzter, D. Ködderitzsch, and H. Ebert, Phys. Rev. Lett. 105, 266604 (2010)
[36] A. A. Kovalev, J. Sinova, and Y. Tserkovnyak, Phys. Rev. Lett. 105, 036601 (2010)
[37] E. Roman, Y. Mokrousov, and I. Souza, Phys. Rev. Lett. 103, 097203 (2009)
[38] J. Kötzler and W. Gil, Phys. Rev. B 72, 060412(R) (2005)