

Intrinsic anomalous spin Hall effect

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I. THEORETICAL DERIVATIONS OF ASHE FROM SPIN-POLARIZED DFT

The basic variables of spin-polarized density functional theory (DFT) are the scalar electron density $n(\vec{r})$ and the vector of the magnetization density $\vec{m}(\vec{r})$. The 2×2 spin density matrix $n^{\alpha\beta}(\vec{r})$ can be defined, with α and β denote spin index, $\uparrow$ for spin up (majority spin) or $\downarrow$ for spin down (minority spin). The connection between $n(\vec{r})$, $\vec{m}(\vec{r})$, and $n^{\alpha\beta}(\vec{r})$ is given by

$$n(\vec{r}) = \sum_{\alpha} n^{\alpha\alpha}(\vec{r}) = n^{\uparrow}(\vec{r}) + n^{\downarrow}(\vec{r}) \quad (1)$$

$$\vec{m}(\vec{r}) = \sum_{\alpha\beta} \vec{\sigma}^{\alpha\beta} n^{\alpha\beta}(\vec{r}) \quad (2)$$

where $\vec{\sigma} = (\sigma_x, \sigma_y, \sigma_z)$ is a vector consisting of the Pauli spin matrices.

For simplicity, here, we only consider collinear magnetic structures. In this case, the spin-up $\psi_i^{\uparrow}(\vec{r})$ and spin-down $\psi_i^{\downarrow}(\vec{r})$ orbitals relate to $n^{\uparrow}(\vec{r})$ and $n^{\downarrow}(\vec{r})$ by¹

$$n^{\uparrow}(\vec{r}) = \sum_i |\psi_i^{\uparrow}(\vec{r})|^2 \quad (3)$$

$$n^{\downarrow}(\vec{r}) = \sum_i |\psi_i^{\downarrow}(\vec{r})|^2 \quad (4)$$

The general Kohn-Sham equations for orbitals $\psi_i^{\uparrow,\downarrow}(\vec{r})$ can be written as¹

$$\left(-\frac{\hbar^2}{2m} \nabla^2 + e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}' + V_{ext}^{\uparrow}(\vec{r}) + V_{xc}^{\uparrow}(\vec{r}) \right) \times \psi_i^{\uparrow}(\vec{r}) = \epsilon_i^{\uparrow} \psi_i^{\uparrow}(\vec{r}) \quad (5)$$

$$\left(-\frac{\hbar^2}{2m} \nabla^2 + e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}' + V_{ext}^{\downarrow}(\vec{r}) + V_{xc}^{\downarrow}(\vec{r}) \right) \times \psi_i^{\downarrow}(\vec{r}) = \epsilon_i^{\downarrow} \psi_i^{\downarrow}(\vec{r}) \quad (6)$$

where $-\frac{\hbar^2}{2m} \nabla^2$ and $e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}'$ are the kinetic energy and the Hartree (or Coulomb) potential, respectively. They are spin-independent. In an externally applied magnetic field, the external potential $V_{ext}^{\uparrow,\downarrow}(\vec{r})$ contains a field term ($\mp \mu_B H$), with negative and positive signs for spin-up and spin-down electrons, respectively. The exchange-correlation potential $V_{xc}^{\uparrow,\downarrow}(\vec{r})$ is defined by the functional derivative of the exchange-correlation energy

$$V_{xc}^{\uparrow,\downarrow}(\vec{r}) = \frac{\delta E_{xc}[n^{\uparrow}(\vec{r}), n^{\downarrow}(\vec{r})]}{\delta n^{\uparrow,\downarrow}(\vec{r})} \quad (7)$$

which can have different values for the two spin characteristics even without an external magnetic field. If one ignores the correlation part and only considers the exchange part of E_{xc} , then a simple form of the potential can be derived from the Hartree-Fock formula^{2,3}

$$E_{xc}[n^{\uparrow}(\vec{r}), n^{\downarrow}(\vec{r})] = E_x = -\frac{3}{2} \left(\frac{3}{4\pi} \right)^{\frac{1}{3}} \left[(n^{\uparrow}(\vec{r}))^{\frac{4}{3}} + (n^{\downarrow}(\vec{r}))^{\frac{4}{3}} \right] \quad (8)$$

Therefore, if there is no external magnetic field, i.e., $V_{ext} = 0$, the Hamiltonian for the spin-up electrons is

$$H^{\uparrow}(\vec{r}) = -\frac{\hbar^2}{2m} \nabla^2 + e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}' - \frac{3}{2} \left(\frac{3}{4\pi} \right)^{\frac{1}{3}} n^{\uparrow}(\vec{r})^{\frac{4}{3}} \quad (9)$$

Note that the first two terms on the right side, i.e., $-\frac{\hbar^2}{2m} \nabla^2$ and $e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}'$, are spin independent. Similarly, the Hamiltonian for the spin-down electrons can be expressed as

$$H^{\downarrow}(\vec{r}) = -\frac{\hbar^2}{2m} \nabla^2 + e^2 \int \frac{n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}' - \frac{3}{2} \left(\frac{3}{4\pi} \right)^{\frac{1}{3}} n^{\downarrow}(\vec{r})^{\frac{4}{3}} \quad (10)$$

Thus, the gradient of the Hamiltonian is

$$\vec{\nabla} H^{\uparrow\downarrow}(\vec{r}) = e^2 \int \frac{\vec{\nabla} n(\vec{r}')}{|\vec{r} - \vec{r}'|} d\vec{r}' - 2 \left(\frac{3}{4\pi} \right)^{\frac{1}{3}} n^{\uparrow\downarrow}(\vec{r})^{\frac{1}{3}} \vec{\nabla} n^{\uparrow\downarrow}(\vec{r}) \quad (11)$$

Hence, a spin-dependent electric field induced by magnetism can be obtained

$$\vec{E}^p(\vec{r}) = -[\vec{\nabla} H^{\uparrow}(\vec{r}) - \vec{\nabla} H^{\downarrow}(\vec{r})] = 2 \left(\frac{3}{4\pi} \right)^{\frac{1}{3}} \left[n^{\uparrow}(\vec{r})^{\frac{1}{3}} \vec{\nabla} n^{\uparrow}(\vec{r}) - n^{\downarrow}(\vec{r})^{\frac{1}{3}} \vec{\nabla} n^{\downarrow}(\vec{r}) \right] \quad (12)$$

Note that the magnetism induced spin-dependent electric field differs from the conventional electric field, i.e., the conventional electric field can be defined as $\vec{E}(\vec{r}) = -[\nabla H^{\uparrow}(\vec{r}) + \nabla H^{\downarrow}(\vec{r})]$, while the spin-dependent electric field is from $-\nabla H^{\uparrow}(\vec{r}) - \nabla H^{\downarrow}(\vec{r})$. The spin-dependent electric field represents the difference of electric fields between spin-up and spin-down electrons.

It is known that in a ferromagnetic (FM) or AFM material, $n^{\uparrow}(\vec{r}) \neq n^{\downarrow}(\vec{r})$ for the charge density at position $\vec{r}$. In addition, in solid materials, expecting $\nabla n^{\uparrow}(\vec{r}) \neq 0$ and $\nabla n^{\downarrow}(\vec{r}) \neq 0$ for the charge density at most coordinates is natural. Thus, one can have a non-zero spin-dependent electric field $\vec{E}^p(\vec{r})$ according to Eq. (12). Note that $\vec{E}^p(\vec{r})$ becomes increasingly sizeable as $\vec{r}$ approaches the atomic position due to the increasingly significant difference between $n^{\uparrow}(\vec{r})$ and $n^{\downarrow}(\vec{r})$.

However, if all the magnetic atoms in a material have identical magnetic momentum, then the distributions of spin-polarized charge-density (SPCD), i.e., $[n^{\uparrow}(\vec{r}) - n^{\downarrow}(\vec{r})]$ around each magnetic atom would be the same. So the symmetry of the system is only determined by the geometric symmetry. This means that if a material has geometric inversion symmetry, then the summation of $\vec{E}^p(\vec{r})$ over space would be zero due to symmetry protection. For example, in BCC Fe, although the distribution of local magnetic momentum $[n^{\uparrow}(\vec{r}) - n^{\downarrow}(\vec{r})]$ and thus the magnetism-induced spin-dependent electric field $\vec{E}(\vec{r})$ are non-zero at position $\vec{r}$, the effective spin-dependent electric field $\vec{E}_{eff}^p$ in the system is still zero, i.e., $\vec{E}_{eff}^p = \int \vec{E}(\vec{r}) d\vec{r} = 0$ with integration throughout the simulated cell. Therefore, one cannot expect the ASHE to originate from the collinear magnetic order $\mathbf{M}$ in BCC Fe only via the spin charge density distribution. Our DFT calculations using the spin Berry phase method show that the ASHE conductance is particularly small, with the maximum value being -3.14 ($\hbar/e$) S/cm, approximately 0.5% of the maximum SHE conductance (Table I in the maintext). Note that the non-zero values can be attributed to other factors contributing to the ASHE, which are beyond the scope of the non-zero spin-dependent electric field discussed above.

In contrast, if two atoms in a material do not have the same magnetic momentum, such as in AFM order, then $[n^{\uparrow}(\vec{r}) - n^{\downarrow}(\vec{r})]$ around the two atoms are not spatially equivalent, making the two magnetic atoms inequivalent. Thus, the magnetic order $\mathbf{M}$ can lead to an additional symmetry breaking. As a result, one can obtain a non-zero integration of $\vec{E}^p(\vec{r})$ along a certain direction in a material with geometric inversion symmetry. Let us take TET Mn₂Au, which has an AFM ground state, as an example. From the viewpoint of atomic structure, there are only two types of atoms, i.e., Mn and Au, thus the inversion symmetry is well preserved [see Fig. 1 (a) in the maintext]. However, from the viewpoint of SPCD distributions, the two Mn atoms with different magnetization directions are actually inequivalent. This feature leads to a further broken of symmetry beyond the atomic structure. As indicated in Fig. 1(a), the inversion symmetry of Mn₂Au would be broken when the two Mn atoms with opposite magnetizations are inequivalent, even though the magnetization direction is in $\vec{x}$ direction. As a result, the non-zero effective spin-dependent electric field in $\vec{z}$ direction (E_{eff}^{pz}) would appear. As discussed below, the non-zero E_{eff}^{pz} can induce a sizeable ASHE.

In the SOI representation, the position-dependent Hamiltonian at $H^s(\vec{r})$ induced by the spin-dependent electric field in the $\vec{z}$ direction can be expressed as

$$H^s(\vec{r}) = \frac{\hbar e}{2mc^2} [\vec{\sigma} \times \vec{E}^p(\vec{r})] \cdot \vec{v}(\vec{r}) = \frac{\hbar e}{2mc^2} [\sigma_y \cdot E^{pz}(\vec{r}) \cdot v_x(\vec{r})] - \frac{\hbar e}{2mc^2} [\sigma_x \cdot E^{pz}(\vec{r}) \cdot v_y(\vec{r})] \quad (13)$$

where $v_x(\vec{r})$ and $v_y(\vec{r})$ are the electron velocities in the $\vec{x}$ and $\vec{y}$ directions, respectively. Here $H^s(\vec{r})$ has a meaning of the difference between spin-up and spin-down Hamiltonians induced by the SOI. Thus, one can obtain an expression for the effective Hamiltonian induced by SOI (H_{eff}^s), which is obtained by integration of Eq. (13) throughout the space

$$H_{eff}^s = \int H^s(\vec{r}) d\vec{r} = \frac{\hbar e}{2mc^2} (\sigma_y \cdot E_{eff}^{pz} \cdot v_x) - \frac{\hbar e}{2mc^2} (\sigma_x \cdot E_{eff}^{pz} \cdot v_y) \quad (14)$$

by assuming that the velocity of conduction electrons is independent of position $\vec{r}$. The non-zero effective Hamiltonian H_{eff}^s means that the spin-up and spin-down electrons receive different forces in the $\vec{x}$ (σ_x) and $\vec{y}$ (σ_y) directions.

Hence, the velocity of electrons with $\sigma_y^\uparrow$ ($\sigma_x^\uparrow$) and $\sigma_y^\downarrow$ ($\sigma_x^\downarrow$) spin polarizations would be different when $\sigma_y \cdot E_{eff}^{pz} \cdot v_x \neq 0$ ($\sigma_x \cdot E_{eff}^{pz} \cdot v_y \neq 0$). As a result, one can obtain a non-zero ASHC with spin polarization in the $\vec{y}$ ($\vec{x}$) direction, as discussed in the maintext. As shown in Table I in the maintext, the ASHCs with spin polarizations in the $\vec{x}$ and $\vec{y}$ directions are significantly larger than those with spin polarization in the $\vec{z}$ direction. In particular, when the charge current (or external electric field) is applied in the $\vec{x}$ direction, a sizeable ASHC with spin current and spin polarization both in the y direction (σ_{yx}^y) would be generated in Mn_2Au . Our DFT calculations show $\sigma_{yx}^y = 15.57$ ($\hbar/e$) S/cm, reaching 19% of that of its SHE counterpart [$\sigma_{zx}^y = -81.11$ ($\hbar/e$) S/cm]. In contrast, according to Eq. (14), σ_z does not directly interact with E_z^p , meaning that σ_{zx}^z would be particularly small. Our DFT calculations also show that σ_{zx}^z approaches zero [0.02 ($\hbar/e$) S/cm], agreeing well with the above discussions.

On the other hand, the crystal structure of nonmagnetic material $\text{T}_d\text{-WTe}_2$ [Fig. 3(a) in the maintext] breaks the mirror symmetry with respect to the xz plane, and thus leads to an inversion symmetry broken. The broken symmetry induces a non-zero conventional electric field in $\vec{y}$ direction (E_{eff}^y). As a result, the non-zero effective Hamiltonian $H_{eff} = \frac{\hbar e}{2mc^2} (\sigma_z \cdot E_{eff}^y \cdot v_x + \sigma_x \cdot E_{eff}^y \cdot v_z)$ would be induced, which leads to the energy splitting between $\sigma_z^\uparrow$ and $\sigma_z^\downarrow$. Similar to the case of Mn_2Au , a sizeable ASHC with spin current and spin polarization both in the $\vec{z}$ direction would be generated in $\text{T}_d\text{-WTe}_2$, which is confirmed by our DFT calculations [$\sigma_{zx}^z = 18.99$ ($\hbar/e$) S/cm]. For the charge current in the $\vec{y}$ direction, since E_{eff}^y does not directly interact with v_y , a small σ_{zy}^z is expected. Our DFT calculations further show that $\sigma_{zy}^z = 2.08$ ($\hbar/e$) S/cm, which is much smaller than σ_{zx}^z . The above results also agree well with experimental observations^{4,5}.

II. THE K-POINT MESH CONVERGENCE IN SHC AND ASHC CALCULATIONS

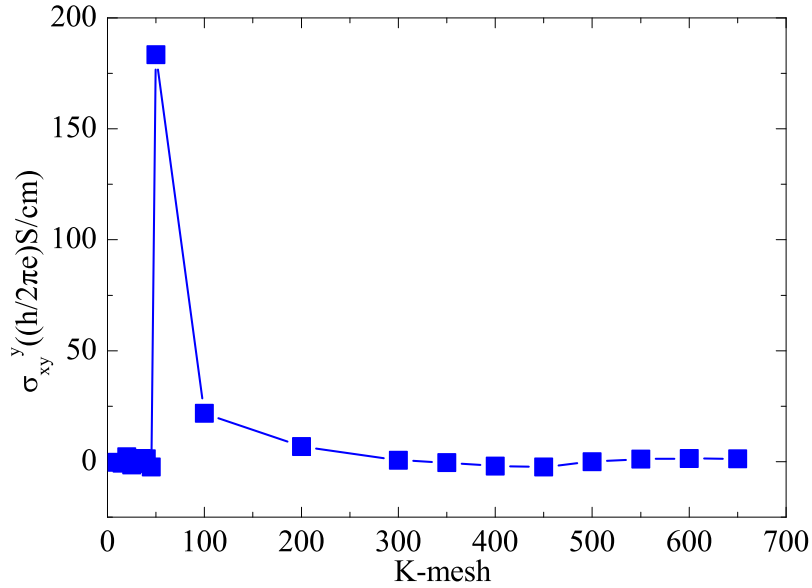


FIG. S1. Calculated k-mesh dependence of σ_{xy}^y of Fe.

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