Sign Changes of Intrinsic Spin Hall Effect in Semiconductors and Simple Metals: First-Principles Calculations

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First-principles calculations are applied to study spin Hall effect in semiconductors and simple metals. We found that intrinsic spin Hall conductivity (ISHC) in realistic materials shows rich sign changes, which may be used to distinguish the effect from the extrinsic one. The calculated ISHC in n-doped GaAs can be well compared with experiment, and it differs from the sign obtained from the extrinsic effect. On the other hand, the ISHC in W and Au, which shows opposite sign respectively, is robust and not sensitive to the disorder.

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The existence of Berry phase in systems with spin-orbit coupling (SOC) can act as gauge field in the momentum space, which in turn affects the transport behavior of electrons in real space, and produces the fascinating new phenomena in solid crystals. One typical example is the intrinsic spin-Hall effect (ISHE) proposed recently.\textsuperscript{3} The ISHE is an effect that, for non-magnetic materials with SOC, a transverse pure spin transport can be induced by external electric-field in the absence of magnetic field (even at room temperature). It is distinguished from the extrinsic spin-Hall effect (ESHE), which is due to impurity scattering.\textsuperscript{3} The obvious advantages of ISHE, especially for the field of spintronics, have stimulated extensive studies recently, both theoretically and experimentally.\textsuperscript{1,2,4,5,6,7,8,9,10,11,12,13,14,15,16,17} Up to now, most of the theoretical studies were done based on certain model Hamiltonians, like Luttinger, Rashba, or Dresselhaus Hamiltonians. These studies provide pictures for the understanding of deep physics (such as the effects of vertex correction\textsuperscript{21,22}), but on the other hand, neglect the band details, which could be very important (as will be addressed in the present paper) due to the topological nature of ISHE. Two experimental evidences have been provided for the existence of spin Hall effect (SHE)\textsuperscript{18,19}. The SHE on the 2D hole gas\textsuperscript{18} is likely of the intrinsic origin, however, the intrinsic or the extrinsic origin of the SHE in the 3D electron film\textsuperscript{19} is still under debate.\textsuperscript{21,22} In order to have close comparison between theory and experiment, parameter-free considerations including all band details are highly desirable, and will be the main focus of this paper. In particular, our calculations make reliable predictions on the sign of the ISHE, which in some cases differs from the sign obtained from the ESHE. This qualitative difference can be used to determine the origin of the effect.

In this letter, we will consider realistic materials by using first-principles calculations to study the ISHE in various systems, including semiconductors (Si and GaAs), and simple metals (W and Au). The main difficulty of such study comes from the accurate evaluation of Berry curvature. We have recently developed a technique to evaluate such property accurately, and have applied it to the calculation of the anomalous Hall effect (AHE) in Fe and SrRuO\textsubscript{3}\textsuperscript{23,24}. Here we apply it to study the ISHE. Besides the quantitative evaluation, we will concentrate on the sensitivity of ISHE to band details and the rich sign changes of ISHE in various materials, which will provide strong support for future experiments to identify the ISHE.

First-principles calculations have been done based on standard density functional theory using accurate FLAPW (full potential linearized augmented plane wave) method, in which the relativistic SOC has been treated fully self-consistently. The exchange-correlation potential was treated by the generalized gradient approximation (GGA), whose validity for the systems we consider here has been shown by many other studies. The underestimated band gap in GGA for semiconductors does not produce problems for our purpose here, because the concerns are for the d.c. limit (ω=0). Accurate k-point integration has been done by tetrahedron method or adaptive mesh refinement.\textsuperscript{24} The convergence of calculated results with respect to the number of k-point has been carefully checked. In general, the number of k-point required to achieve accuracy of 5% is around 1,000,000 in the Brillouin Zones (BZ). For all the calculations presented here, experimental lattice parameters are used.

Suppose the external electric field is applied along the y direction, then the linear response of the spin (σ\textsubscript{x}) current along the x direction can be obtained from the Kubo formula by evaluating the spin Hall conductivity tensor,

\begin{equation}
\sigma_{xy}(\omega) = \frac{e}{\hbar} \int_{V_0} \frac{d^3k}{(2\pi)^3} \Omega(k)
\end{equation}

where \(\sigma_{xy}(\omega)\) is the spin Hall conductivity, e is the elementary charge, \(\hbar\) is the reduced Planck constant, and \(\Omega(k)\) is the magnetic field correction.
where $|nk\rangle$ is the eigen wave function of Bloch state with eigen value $E_n$, and Fermi occupation number $f_{nk}$. $v_y$ is the velocity operator, $j_x$ is the spin current operator, which is defined as $\frac{i}{\hbar}(\sigma_z v_y + v_x \sigma_y)$. $\Omega_n$ is the spin Berry phase connection of the Bloch state, and is responsible for the anomalous transverse transport we studied. The important point here is that all band details and SOC are self-consistently taken into account (no adjustable parameters), no approximation beyond linear response theory has been used. It is also straightforward to take into account the impurity scattering effect by allowing finite life time broadening $\delta$, and the finite temperature effect in the Fermi distribution $f_{nk}$. For the definition of sign, positive $\sigma_{xy}$ means that spin-up ($s_z=1/2$) component flows to the positive $x$ direction. For the convenience of comparison, in our following discussions, we convert spin conductivity into the unit of charge conductivity by multiplying a factor $\frac{2\hbar}{e^2}$ to the calculated values.

$\Omega(k) = \sum_n f_{nk} \Omega_n(k)$

$$= \sum_n f_{nk} \sum_{n' \neq n} -\frac{2\text{Im} \langle nk| j_x |n'k \rangle \langle n'k| v_y |nk \rangle}{(E_{n'} - E_n)^2 - (\hbar \omega + i\delta)^2} \sigma_z$$

FIG. 1: The calculated (a) band structure; (b) spin-Hall conductivity $\sigma_{xy}$ as function of Fermi level position for GaAs. We define the converged Fermi level without doping as energy zero point, and evaluate $\sigma_{xy}$ by rigidly shift the Fermi level position. The panel (c) gives the $\sigma_{xy}$ of n-GaAs as functions of carrier density after subtracting the part that does not contribute to spin accumulation (see the text part for explanation). Note the factor of 10 for the $T=30K$ and $\delta=16meV$ curve.

Semiconductors: Fig.1 and Fig.2 show the calculated band structures and $\sigma_{xy}$ as functions of Fermi level ($E_F$) position for GaAs and Si respectively. In the following, we will first concentrate on the zero-temperature and clean limit ($T=0K$, $\delta=0eV$), then the effects of disorder and elevated temperature will be addressed later.

For hole-doped GaAs (Fig.1(b)), the calculated $\sigma_{xy}$ is large, it reaches about 300 $\Omega^{-1}cm^{-1}$, which is about the same order of magnitude as that estimated from Luttinger model [1]. For hole-doped Si (Fig.2(b)), on the other hand, the maximum of $\sigma_{xy}$ is about 50 $\Omega^{-1}cm^{-1}$. This can be understood by taking into account the fact that the strength of SOC in Si is about 1/7.7 of that in GaAs. In both cases, the $\sigma_{xy}$ first increases with increasing hole density, after reaches maximum it goes down. By carefully calculate the Berry curvature distribution $\Omega(k)$, we found that the contributions from the light hole and split-off bands are mostly negative, which will compensate the positive contributions from the heavy hole band, and finally suppress the $\sigma_{xy}$ for large hole density.

For the electron-doped GaAs (Fig.1(b)), the situation is complicated and far beyond what we understood from model analysis. The calculated $\sigma_{xy}$ shows sign changes as electron density varies: negative (positive) for low (high) density. Such behavior is related to a small splitting of the conduction band due to the lack of inversion symmetry in GaAs. For n-GaAs, the conduction band bottom has mostly $s$ orbital character. Due to the $s$-$p$ hybridization, some $p$ characters exist in this band, leading to the Dresselhaus type SOC in the simplified model (therefore the vertex correction is not important [20,22]).

For electron-doped Si (Fig.2(b)), on the other hand, the obtained $\sigma_{xy}$ at clean limit is quite large (although the strength of SOC in Si is small), and has negative sign. In this case, the conduction-band-bottom is neither around $\Gamma$ point nor $s$ orbital like. The calculated Berry phase contributions to ISHE are mostly related to the conduction bands around $X$ point of the BZ. Considering the factor that the spin relaxation time in electron-doped Si is typically much longer than that in hole-doped case [22], our results suggest the possibility to realize ISHE in Si, which is the most important semiconductor material.
For both GaAs and Si, the effects of temperature and disorder are important for the electron-doped cases, while not so dramatic for hole-doped ones. As shown in Fig.2(b) for Si, by putting $\delta=20\text{meV}$ and $T=300\text{K}$ in our calculations, the $\sigma_{xy}$ is significantly reduced for n-Si, while not so much for p-Si. This is also true for GaAs as shown in Fig.1(c). The disorder and temperature will even cause sign change for $\sigma_{xy}$ in n-GaAs as will be discussed later in comparison with experimental results.

For the insulating GaAs and Si ($E_F$ located in the gap), however, the calculated $\sigma_{xy}$ is non-vanishing (about 43 and 7 $\Omega^{-1}\text{cm}^{-1}$ for GaAs and Si respectively). First we have to emphasize that this is not due to numerical error, which is four orders of magnitude smaller. Our results can be regarded as a generalization of the concept of “Spin Hall Insulator” [16]. Murakami et.al. studied the spin Hall effect in narrow-gap and zero-gap semiconductors like PbTe and HgTe, which have “special” band structures (such as the inverted light hole and heavy hole bands in HgTe), and demonstrate the existence of spin Hall insulator. However, the results here suggest that such “special” band structure is not necessary in general. In real materials, there always exist finite hybridizations, which will produce non-vanishing ISHE in insulators. Note that ISHE is not quantized [11], in qualitative difference with the AHE. Nevertheless, we should emphasize that the existence of such ISHE in insulator will not produce any spin accumulation due to the lack of broken time reversal symmetry [16].

To make comparison with experimental results on n-doped GaAs [19], we show in Fig.1(c) the calculated $\sigma_{xy}$ as functions of electron density, with the subtraction of the part that does not contribute to the spin accumulation (the value within the gap). Now it is very clear that the $\sigma_{xy}$ (for $T=0\text{K}, \delta=0\text{meV}$) is negative for small doping, but change sign to be positive for large doping. We also notice that such fluctuation of $\sigma_{xy}$ are suppressed by introducing disorder and temperature effects. This, on the one hand, is the nature results of topological origin of ISHE, and on the other hand, suggests complication of ISHE in realistic materials. For the experimental doping density ($3 \times 10^{16}\text{cm}^{-3}$) [19], the calculated $\sigma_{xy}$ ($\delta=0\text{meV}, T=0\text{K}$) has the same sign (negative) as that obtained in experiment, and also agrees with Ref. [21]. This is in sharp contrast with the extrinsic SHE, which has opposite sign as estimated in Ref. [21]. The absolute value of calculated $\sigma_{xy}$ at clean limit is two order of magnitude larger than experimental value ($-0.005\Omega^{-1}\text{cm}^{-1}$), however a compatible number ($-0.01\Omega^{-1}\text{cm}^{-1}$) can be obtained by introducing a finite lifetime broadening $\delta=2\text{meV}$ as shown in Fig.1(c). Unfortunately experimental parameter $\delta$ for unstrained sample is unclear [12]. Using the measured $\rho_{xx}$ (300 $\Omega\mu\text{m}$) for strained sample, we estimate the $\delta=16\text{meV}$, which give positive $\sigma_{xy}$ ($T=30\text{K}$, see Fig.1(c)). Nevertheless, considering the uncertainty of experimental parameters, this issue remains to be checked in the future.

![FIG. 3: The same notation as shown in Fig.2, but for W.](image)

![FIG. 4: The same notation as shown in Fig.2, but for Au.](image)

Simple Metal: The ISHE in simple metal has not been studied yet, although it is not surprising to expect that ISHE exists in such systems, due to the same mechanism. We chose elemental W and Au as examples because of the relatively larger SOC. For W the charge conductivity mostly comes from 5$d$ states around Fermi level, while for Au it is mostly from 6$s$ states. The calculated band structures and $\sigma_{xy}$ are shown in Fig.3 and Fig.4 for W and Au respectively. Besides the very strong ISHE obtained (the $\sigma_{xy}$ can reach as high as $-1390\ \Omega^{-1}\text{cm}^{-1}$ for W, and $731\ \Omega^{-1}\text{cm}^{-1}$ for Au), we notice that $\sigma_{xy}$ is negative in W, but positive in Au. This again suggests the rich sign changes of ISHE in realistic materials. What is more interesting is that the ISHE in W and Au are robust and not sensitive to the disorder (in opposite to GaAs or Si). By adding a very large disorder effect ($\delta=0.5\text{eV}$), the calculated $\sigma_{xy}$ only change slightly. Given these special characters, we suggest that both W and Au are nice candidates for future experimental examination of ISHE. Especially for Au, where the conduction electrons have
mostly s characters, relatively long spin relaxation can be expected.

Sign Issue and Discussions: As presented in our above results, the ISHE shows very rich sign changes, which are independent of the carrier type and the sign of impurity potential: (1) for Si, the sign of ISHE is the same as ordinary Hall effect, i.e., positive for hole doping and negative for electron doping; (2) for W and Au, however, the sign of ISHE is opposite with their carrier type: W (Au) has hole (electron) type conductivity but negative (positive) ISHE; (3) for n-doped GaAs, the sign of ISHE changes with increasing doping. Such rich sign changes are more than what we can expect from extrinsic scattering mechanism. Two mechanisms, namely skew scattering and side-jump, were mainly discussed in literature. For a simple discussion, we consider the skew scattering mechanism, which dominates over side-jump contribution for the weak disorder limit. In this case, the sign of ESHE depends on the sign of scattering potential. It is natural to expect that the sign of skew scattering does not change with changing impurity density. However, we predict that the sign of ISHE can change with the same type of doping (in n-GaAs). Such difference may be used to distinguish ESHE from intrinsic contributions. We can also use simple metal W and Au for such purpose, because opposite signs of ISHE (which has nothing to do with scattering potential) are predicted. Nevertheless, the sign issue should be regarded as a very important aspect of SHE, and can be used in future experiments.

In summary, we have performed detailed studies on the ISHE for various realistic materials based on accurate parameter-free first-principles calculations, and predict rich sign changes of ISHE. Furthermore, we demonstrate that the ISHE in semiconductors (GaAs and Si) is highly sensitive to band-details and disorder, while ISHE in simple metals (W and Au) is robust and not sensitive to disorder. The calculated ISHE for n-GaAs can be well compared with experimental results, while the extrinsic spin Hall contribution has opposite sign.

[1] S. Murakami, N. Nagaosa, and S. C. Zhang, SCIENCE 301, 1348 (2003). ibid, Phys. Rev. B 69, 235206 (2004); S. Murakami, Phys. Rev. B 69, 241202 (2004).
[2] J. Sinova, D. Culcer, Q. Niu, and et. al., Phys. Rev. Lett. 92, 126603 (2004).
[3] J. E. Hirsch, Phys. Rev. Lett. 83, 1834 (1999).
[4] E. I. Rashba, Phys. Rev. B 68, 241315 (2003).
[5] J. Schliemann, D. Loss, Phys. Rev. B 69, 165315 (2004).
[6] S. Q. Shen, Ma, X. C. Xie, and et. al., Phys. Rev. Lett., 92, 256603 (2004); S. Q. Shen, Phys. Rev. B 70, 081311 (2004); L. B. Hu, J. Gao, S. Q. Shen, Phys. Rev. B 68, 153303 (2004).
[7] D. Culcer, J. Sinova, N. A. Sinitsyn, and et. al., Phys. Rev. Lett., 93, 046602 (2004).
[8] N. A. Sinitsyn, E. M. Hankiewicz, W. Teizer W, and et. al., Phys. Rev. B 70, 081312 (2004).
[9] J.I. Inoue, G. E. W. Bauer, L. W. Molenkamp, cond-matt/0402442.
[10] O. V. Dimitrova, cond-mat/0405339.
[11] S. Zhang, Z. Yang, cond-mat/0407704.
[12] L. Sheng, D. N. Sheng, C. S. Ting, cond-mat/0409038.
[13] R.V. Shchelushkin, Arne Brataas, cond-mat/0409130.
[14] A.G. Malshukov, K.A. Chao, cond-mat/0410607.
[15] A. Shekhter, M. Khodas, A. M. Finkelstein, cond-mat/0411239.
[16] S. Murakami, N. Nagaosa, S. C. Zhang, Phys. Rev. Lett., 93, 156804 (2004).
[17] G. Y. Guo, and et.al., (to be published).
[18] J. Wunderlich, B. Kaestner, J. Sinova, T. Jungwirth, Phys. Rev. Lett. 94, 047204 (2005).
[19] Y.K. Kato, R.C. Myers, A.C. Gossard, D.D. Awschalom, Science 306, 1910 (2004).
[20] K. Nomura, J. Sinova, N.A. Sinitsyn, A.H. MacDonald, cond-mat/0506189.
[21] H. A. Engel, B. I. Halperin, E. I. Rashba, cond-mat/0505535.
[22] B. A. Bernevig, S. C. Zhang, cond-mat/0412550.
[23] Z. Fang, N. Nagaosa, K. S. Takahashi, and et. al., SCIENCE, 302, 92 (2003).
[24] Y. Yao, L. Kleinman, A. H. MacDonald, and et. al., Phys. Rev. Lett. 92, 037204 (2004).
[25] I. Zutic, J. Fabian, and S. D. Sarma, Rev. Mod. Phys., 76, 323 (2004).
[26] M. Onoda, N. Nagaosa, Phys. Rev. Lett., 90, 206601 (2003); M. Onoda, N. Nagaosa, J. Phys. Soc. Jpn., 71, 19 (2002).
[27] R. C. Fivaz, Phys. Rev. B 183, 586 (1969); A. Crépieux, P. Bruno, Phys. Rev. B 64, 014416 (2001); J. Smit, Physica 24, 39 (1958).