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 [1,2]. The ISHE is an effect that, for nonmagnetic 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 [3]. The obvious advantages of ISHE, especially for the field of spintronics, have stimulated extensive studies recently, both theoretically [4–17] and experimentally [18,19]. Up to now, most of the theoretical studies were done based on certain model Hamiltonians, like Luttinger, Rashba, or Dresselhauss Hamiltonians. These studies provide pictures for the understanding of deep physics [such as the effects of vertex correction [20]], but on the other hand, neglect the band details, which could be very important (as will be addressed in the present Letter) due to the topological nature of ISHE. Two experimental evidences have been provided for the existence of spin Hall effect (SHE) [18,19]. The SHE on the 2D hole gas [18] is likely of the intrinsic origin; however, the intrinsic or the extrinsic origin of the SHE in the 3D electron film [19] is still under debate [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 Letter. 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 $SrRuO_3$ [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 full potential linearized augmented plane wave (FLAPW) 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 dc limit ($\omega = 0$). Accurate $\mathbf{k}$-point integration has been done by tetrahedron method or adaptive mesh refinement [24]. The convergence of calculated results with respect to the number of $\mathbf{k}$ points has been carefully checked. In general, the number of $\mathbf{k}$ points 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_z) current along the x direction can be obtained from the Kubo formula by evaluating the spin Hall conductivity tensor,

$$\sigma_{xy}(\omega) = \frac{e}{\hbar}\int_{V_G}\frac{d^3k}{(2\pi)^3}\Omega(\mathbf{k}) \tag{1}$$

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

where $|nk\rangle$ is the eigenwave function of Bloch state with eigenvalue E_n and Fermi occupation number f_{nk}. v_y is the velocity operator and j_x is the spin current operator, which is defined as $\frac{\hbar}{4}(\sigma_z v_x + v_x \sigma_z)$. Ω_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 lifetime broadening δ, and the finite temperature effect in the Fermi distribution f_{nk}. For the definition of sign, positive σ_{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 $\frac{2|e|}{\hbar}$ to the calculated values.

Semiconductors.—Figures 1 and 2 show the calculated band structures and σ_{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 ($T = 0$ K, $\delta = 0$ eV), then the effects of disorder and elevated temperature will be addressed later.

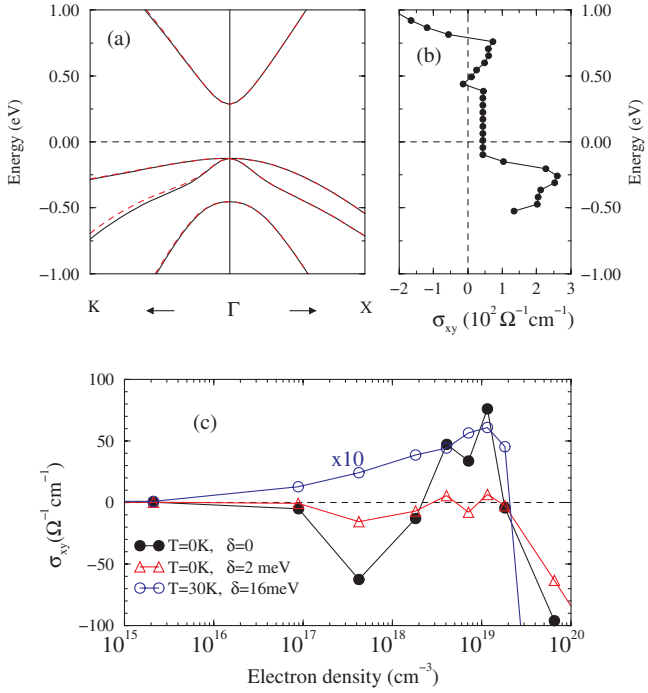

FIG. 1 (color online). The calculated (a) band structure, (b) spin Hall conductivity σ_{xy} as function of Fermi level position for bulk GaAs. We define the converged Fermi level without doping as energy zero point, and evaluate σ_{xy} by rigidly shifting the Fermi level position. The panel (c) gives the σ_{xy} of n-GaAs as functions of carrier (electron) 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 $T = 30$ K and $\delta = 16$ meV curve.

For hole-doped GaAs [Fig. 1(b)], the calculated σ_{xy} is large; it reaches about 300 $\Omega^{-1}\,\mathrm{cm}^{-1}$, which is about the same order of magnitude as that estimated from Luttinger model [11]. For hole-doped Si [Fig. 2(b)], on the other hand, the maximum of σ_{xy} is about 50 $\Omega^{-1}\,\mathrm{cm}^{-1}$. 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 σ_{xy} first increases with increasing hole density; after it reaches maximum it goes down. By carefully calculating the Berry curvature distribution $\Omega(\mathbf{k})$, 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 σ_{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 σ_{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. Because of the s-p hybridization, some p characters exist in this band, leading to the Dresselhaus type SOC in the simplified model [please refer to [20,22] for the possible vertex correction].

For electron-doped Si [Fig. 2(b)], on the other hand, the obtained σ_{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 Γ 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 [25], our results suggest the possibility to realize ISHE in Si, which is the most important semiconductor material.

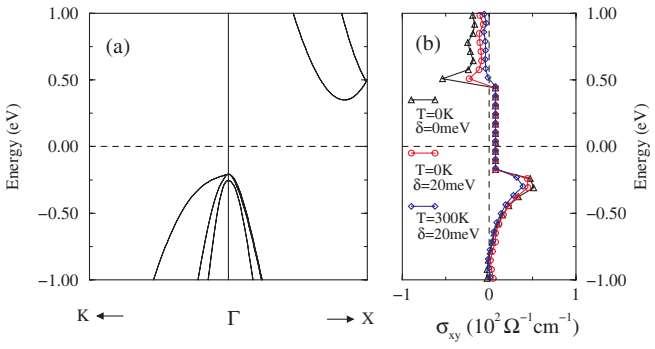

FIG. 2 (color online). The calculated (a) band structure, (b) spin Hall conductivity σ_{xy} as function of Fermi level position for bulk Si.

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 $\delta = 20$ meV and $T = 300$ K in our calculations, the σ_{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 σ_{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 σ_{xy} is nonvanishing (about 43 and 7 $\Omega^{-1}\,\mathrm{cm}^{-1}$ for GaAs and Si, respectively). First we have to emphasize that this is not due to numerical error, which is 4 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 nonvanishing ISHE in insulators. Note that ISHE is not quantized [1], 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 a comparison with experimental results on n-doped GaAs [19], we show in Fig. 1(c) the calculated σ_{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 σ_{xy} (for $T = 0$ K, $\delta = 0$ meV) is negative for small doping, but changes sign to be positive for large doping. We also notice that such fluctuations of σ_{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 (3×10^{16} cm^{-3}) [19], the calculated σ_{xy} ($\delta = 0$ meV, $T = 0$ K) has the same sign (negative) as that obtained in experiment, and also agrees with Ref. [22]. This is in sharp contrast with the extrinsic SHE, which has opposite sign as estimated in Ref. [21]. The absolute value of calculated σ_{xy} at clean limit is 2 orders of magnitude larger than experimental value (-0.005 $\Omega^{-1}\,\mathrm{cm}^{-1}$); however, a compatible number (-0.01 $\Omega^{-1}\,\mathrm{cm}^{-1}$) can be obtained by introducing a finite lifetime broadening $\delta = 2$ meV as shown in Fig. 1(c). Unfortunately, experimental parameter δ for unstrained sample is unclear [19]. Using the measured ρ_{xx} (300 $\Omega\,\mu$m) for strained sample, we estimate the $\delta = 16$ meV, which give positive σ_{xy} [$T = 30$ K, see Fig. 1(c)]. Nevertheless, considering the uncertainty of experimental parameters, this issue remains to be checked in the future.

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 $5d$ states around Fermi level, while for Au it is mostly from $6s$ states. The calculated band structures and σ_{xy} are shown in Figs. 3 and 4 for W and Au, respectively. Besides the very strong ISHE obtained (the σ_{xy} can reach as high as -1390 $\Omega^{-1}\,\mathrm{cm}^{-1}$ for W, and 731 $\Omega^{-1}\,\mathrm{cm}^{-1}$ for Au), we notice that σ_{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 ($\delta = 0.5$ eV), the calculated σ_{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 [25].

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 [3]. Two mechanisms, namely, skew scattering and side jump, were mainly discussed in the literature [26].

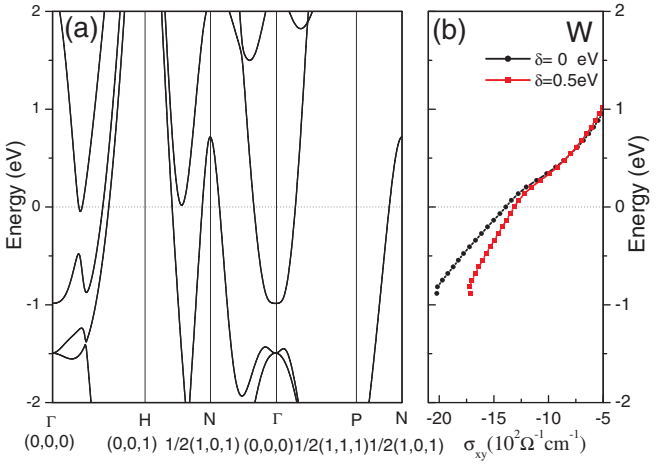

FIG. 3 (color online). The same notation as shown in Fig. 2, but for W.

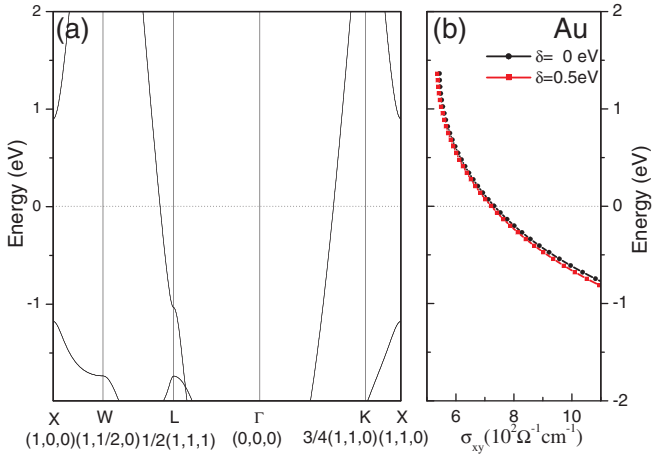

FIG. 4 (color online). The same notation as shown in Fig. 2, but for Au.

For a simple discussion, we consider the skew scattering mechanism [26], 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 [26]. 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 purposes, 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.

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