Ab initio calculation of intrinsic spin Hall conductivity of Pd and AuG. Y. Guo^{a)}*Department of Physics and Center for Theoretical Sciences, National Taiwan University, Taipei 106, Taiwan*

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An *ab initio* relativistic band structure calculation of spin Hall conductivity (SHC) (σ_{xy}^z) in Pd and Au metals has been performed. It is found that at low temperatures, intrinsic SHCs for Pd and Au are, respectively, ~ 1400 and ~ 400 ($\hbar/e$)(Ω cm) $^{-1}$. The large SHC in Pd comes from the resonant contribution from the spin-orbit splitting of the doubly degenerated *4d* bands near the Fermi level at symmetry Γ and *X* points, and the smaller SHC in Au is due to the broad free-electron-like *6s6p* bands. However, as the temperature increases, the SHC in Pd decreases monotonically and reduces to ~ 330 ($\hbar/e$)(Ω cm) $^{-1}$ at 300 K, while the SHC in Au increases steadily and reaches ~ 750 ($\hbar/e$)(Ω cm) $^{-1}$ at room temperature. This indicates that the gigantic spin Hall effect [$\sigma_{xy}^z \approx 10^5$ ($\hbar/e$)(Ω cm) $^{-1}$] observed recently in the Au/FePt system [T. Seki *et al.*, *Nature Mater.* **7**, 125 (2008)] is due to the extrinsic mechanisms such as the skew scattering by the impurities in Au.

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Spin transport electronics (spintronics) has recently become a very active research field in condensed matter and materials physics because of its potential applications in information storage and processing and other electronic technologies¹ and also because of many fundamental questions on the physics of electron spin.² Spin current generation is an important issue in the emerging spintronics. Recent proposals of the intrinsic spin Hall effect (SHE) are therefore remarkable.^{3,4} In the SHE, a transverse spin current is generated in response to an electric field in a metal with relativistic electron interaction [spin-orbit coupling (SOC)]. This effect was first considered to arise extrinsically, i.e., by impurity scattering.^{5,6} The scattering becomes spin dependent in the presence of SOC, and this gives rise to the SHE. In the recent proposals, in contrast, the SHE can arise intrinsically in hole-doped (*p*-type) bulk semiconductors³ and also in electron-doped (*n*-type) semiconductor heterostructures⁴ due to intrinsic SOC in the band structure. The intrinsic SHE can be calculated and controlled, whereas the extrinsic SHE depends sensitively on details of the impurity scattering. Therefore, the intrinsic SHE is more important for applicational purposes in comparison with the extrinsic SHE.

In semiconductors, there have been experimental reports on the SHE in *n*-type GaAs,⁷ *p*-type GaAs,⁸ and *n*-type InGaN/GaN superlattices⁹ in recent years. These experimental results have been discussed theoretically, and it is now recognized that the SHE in *n*-type GaAs is due to extrinsic mechanisms, i.e., skew scattering and side-jump contributions,^{10,11} while that in *p*-type GaAs is mainly caused by an intrinsic mechanism.^{12,13} This conclusion is supported by a theoretical analysis of the impurity effect and vertex correction to SHE in the Rashba and Luttinger models.^{10,12} In *p*-GaAs the fourfold degeneracy at the Γ point of the valence bands acts as the Yang monopole, which enhances the SU(2) non-Abelian Berry curvature.¹⁴

On the other hand, the SHE in metallic systems is currently attracting interest, stimulated by latest experimental reports on the SHE or inverse SHE, i.e., the transverse voltage drop due to the spin current.^{15–17} The spin Hall conductivity (SHC) in metals is much larger than that in the semiconductors. Naively, this may be due to the large number of carriers, but the band structure is very important.¹⁸ Furthermore, the Fermi degeneracy temperature is much higher than room temperature, and hence, the quantum coherence is more robust against the thermal agitations compared with the semiconductor systems.

In recent experiments for metallic systems,^{15,17} platinum shows a prominent SHE surviving even up to room temperature, whereas aluminum and copper show relatively little SHE. However, the mechanism of the SHE in metals has been assumed to be extrinsic. In Ref. 16 this difference is attributed to a magnitude of SOC for each metal. However, platinum seems to be special even among heavy elements. This material dependence strongly suggests a crucial role of intrinsic contributions. Therefore it is highly desired to study the intrinsic SHE of platinum as a representative material for metallic SHE. If this analysis successfully explains the experiment, it will open up the possibility to theoretically design the SHE in metallic systems. Therefore, we recently carried out *ab initio* calculations for the SHC in platinum.¹⁸ We found that the intrinsic SHC is as large as ~ 2000 ($\hbar/e$)(Ω cm) $^{-1}$ at low temperature and decreases down to ~ 200 ($\hbar/e$)(Ω cm) $^{-1}$ at room temperature. It is due to the resonant contribution from the spin-orbit splitting of the doubly degenerated *d* bands at high-symmetry *L* and *X* points near the Fermi level. We showed, by modeling these near degeneracies by an effective Hamiltonian, that the SHC has a peak near the Fermi energy and that the vertex correction due to impurity scattering vanishes, indicating that the large SHE observed experimentally in platinum¹⁶ is of intrinsic nature.

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FIG. 1. (Color online) (a) Relativistic band structure and (b) SHC of fcc Pd. The zero energy and the dotted line is the Fermi level. The dashed curves in (a) are the scalar-relativistic band structure.

In this paper, we study the intrinsic SHE in Pd and Au metals with first-principles band structure calculations. The band structures of Pd and Au have been calculated using a fully relativistic extension¹⁹ of the all-electron linear muffin-tin orbital method²⁰ based on the density functional theory with local density approximation.²¹ The experimental lattice constants used for Pd and Au are 3.89 and 4.08 Å, respectively. The basis functions used are *s*, *p*, *d*, and *f* muffin-tin orbitals.²⁰ In the self-consistent electronic structure calculations, 89 *k* points in the fcc irreducible wedge (IW) of the Brillouin Zone (BZ) were used in the tetrahedron BZ integration. The SHC is evaluated by the Kubo formula, as described in Ref. 22. A fine mesh of 60 288 *k* points on a larger IW (three times the fcc IW) is used. These numbers correspond to the division of the ΓX line into 60 segments (see Fig. 1).

Figure 1 shows the relativistic band structure of Pd and also the SHC (σ_{xy}^z) as a function of E_F . Clearly, the SHC peaks just below the true Fermi level (0 eV), with a large value of $\sim 1400 (\hbar/e)(\Omega \text{ cm})^{-1}$. This large value of the SHC is smaller than that of Pt.¹⁸ Nonetheless, it is still orders of magnitude larger than the corresponding value in *p*-type semiconductors Si, Ge, GaAs, and AlAs.^{22,23} Note that the SHC in Pd decreases monotonically as the E_F is artificially raised and becomes rather small above 3.0 eV. When the E_F is artificially lowered, the SHC first peaks just below the E_F (−0.3 eV), then decreases considerably, and changes its sign at −1.2 eV. As the E_F is further lowered, the SHC increases in magnitude again and becomes peaked at −3.0 eV with a large value of $-2600 (\hbar/e)(\Omega \text{ cm})^{-1}$. The SHC decreases again when the E_F is further lowered and finally becomes very small below −5.0 eV. Note that the bands below −5.5 eV and above 0.5 eV are predominantly of 5*s* character and the effect of the SOC is small.

We notice that a peak in the SHC appears at the double degeneracies on the *L* and *X* points near E_F in the scalar-relativistic band structure (i.e., without the SOC), while the other peak at −3.0 eV occurs near the double degeneracies at the *L* and Γ points (see Fig. 1). The double degeneracy (bands 5 and 6) at *L* is made mostly of $d_{x'z'}$ and $d_{y'z'}$ (z' : threefold axis), consistent with the point group D_{3d} at *L*. The double degeneracy (bands 4 and 5) at *X* consists mainly of

FIG. 2. (Color online) (a) Relativistic band structure and (b) SHC of fcc Au. The zero energy and the dotted line is the Fermi level.

$d_{x'z'}$ and $d_{y'z'}$ (z' : fourfold axis), consistent with the point group D_{4h} . These double degeneracies are lifted by the SOC, with a rather large spin-orbit splitting. As in Pt, the large SHC in Pd may be attributed to these double degeneracies.¹⁸

The relativistic band structure of Au and also the SHC (σ_{xy}^z) as a function of E_F are displayed in Fig. 2. It is clear that both the shape and the amplitude of the SHC versus E_F curve [Fig. 2(b)] of Au are very similar to that of Pd [Fig. 1(b)] and Pt.¹⁸ This is because the band structure of Au [Fig. 2(a)] is rather similar to that of Pd [Fig. 1(a)] and Pt.¹⁸ However, because Au has an extra valence electron and hence its *d* band is completely filled, the E_F falls in the broad 6*s*6*p* band, where the SOC is relatively small. As a result, the SHC in Au is relatively small at low temperatures [$\sigma_{xy}(T=0 \text{ K}) \approx 400 (\hbar/e)(\Omega \text{ cm})^{-1}$], consistent with the previous *ab initio* calculation.²³ Nonetheless, it is a few times larger than the SHC in semiconductors.^{22,23}

The SHC can be written in terms of Berry curvature $\Omega_n^z(\mathbf{k})$ as

$$\sigma_{xy}^z = \frac{e}{\hbar} \sum_{\mathbf{k}} \Omega^z(\mathbf{k}) = \frac{e}{\hbar} \sum_{\mathbf{k}} \sum_n f_{\mathbf{k}n} \Omega_n^z(\mathbf{k}),$$

$$\Omega_n^z(\mathbf{k}) = \sum_{n' \neq n} \frac{2 \text{Im}[\langle \mathbf{k}n | j_x^z | \mathbf{k}n' \rangle \langle \mathbf{k}n' | v_y | \mathbf{k}n \rangle]}{(\epsilon_{\mathbf{k}n} - \epsilon_{\mathbf{k}n'})^2}, \quad (1)$$

where the spin current operator $j_x^z = \frac{1}{2} \{s_z, v_x\}$, with spin s_z given by $s_z = (\hbar/2) \beta \Sigma_z$ (β, Σ_z : 4×4 Dirac matrices).²² $f_{\mathbf{k}n}$ is the Fermi distribution function for the *n*th band at $\mathbf{k}$. Ω_n^z can be regarded as an analog of the Berry curvature for the *n*th band and it is enhanced when other bands come close in energy (i.e., near degeneracy). The SHC for Pd and Au calculated as a function of temperature using Eq. (1) is shown in Fig. 3. Figure 3 shows that the SHC in Pd decreases substantially as the temperature (*T*) is raised above 100 K, although it increases with temperature below 100 K. This rather strong temperature dependence is also due to the near degeneracies since the small energy scale is relevant to the SHC there. Nevertheless, the SHC at room *T* [$\sigma_{xy}(T=300 \text{ K}) = 350 (\hbar/e)(\Omega \text{ cm})^{-1}$] is still rather large. Interestingly, in contrast, the SHC in Au increases steadily with temperature to reach a value of $750 (\hbar/e)(\Omega \text{ cm})^{-1}$. This is because the E_F cuts across the broad 6*s*6*p* band in Au. As a

FIG. 3. (Color online) Temperature dependence of the SHC σ_{xy}^z in Pd and Au metals.

result, the intrinsic SHC in Au at room temperature is in fact larger than that of Pd and Pt.

Excitingly, giant SHE at room temperature has been recently observed in a multiterminal device with a Au Hall cross and an FePt perpendicular spin injector.²⁴ The measured $\sigma_{xy}^z \approx 10^5 (\hbar/e)(\Omega \text{ cm})^{-1}$ is orders of magnitude larger than the intrinsic SHC in bulk Au reported above. This obviously indicates that the SHC due to intrinsic SOC in the band structure of pure Au is not the dominant mechanism in the Au/FePt system. Indeed, Seki *et al.*²⁴ attributed the giant SHE to the extrinsic mechanism of the skew scattering by impurities in Au. Nonetheless, its microscopic origin remains a puzzle and is currently under intensive investigation.²⁵

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