Giant enhancement of the intrinsic spin Hall conductivity in $\beta$-tungsten via substitutional doping

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A key challenge in manipulating the magnetization in heavy-metal/ferromagnetic bilayers via the spin-orbit torque is to identify materials that exhibit an efficient charge-to-spin current conversion. *Ab initio* electronic structure calculations reveal that the intrinsic spin Hall conductivity (SHC) for pristine $\beta$-W is about 60% larger than that of $\alpha$-W. More importantly, we demonstrate that the SHC of $\beta$-W can be enhanced via Ta alloying. This is corroborated by spin Berry curvature calculations of $W_{1-x}Ta_x$ ($x \approx 12.5\%$) alloys which show a giant enhancement of the spin Hall angle of up to $\sim 0.5$. The underlying mechanism is the synergistic behavior of the SHC and longitudinal conductivity with the Fermi level position. These findings not only pave the way for enhancing the intrinsic spin Hall effect in $\beta$-W, but also provide guidelines to exploit substitutional alloying to tailor the spin Hall effect in various materials.

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Current-driven spin-orbit torques (SOTs) in heavy-metal/ferromagnetic heterostructures have drawn increasing attention because they can provide an efficient way of manipulating the magnetization [1–3]. The SOT arises from the transfer of spin angular momentum between the charge current and the local magnetization in the presence of spin-orbit coupling (SOC). The underlying origin of the SOT is still under debate: It may arise from either the bulk spin Hall effect (SHE) [4–7] in the heavy metal or the interfacial SOC [8,9], or both [10,11]. In the SHE, an in-plane charge current density $J_s$ flowing in the heavy metal with strong SOC generates a transverse spin current $J_H$. The SHE efficiency and the critical current density for magnetization switching is expressed in terms of the spin Hall angle (SHA), $\theta_{SH} = J_s / J_H$, [4,5], which measures the efficiency of the charge current to spin current conversion. It only depends on the material properties and can be experimentally measured via the inverse spin Hall effect [12,13], the spin Seebeck effect [14,15], the Kerr effect [16], or the spin Nernst effect [17].

Relatively large values of $\theta_{SH}$ have been reported in the 5$d$ elemental solids of Pt (0.07) [2,18] and the high-resistivity $\beta$-Ta ($-0.15$) [2]. More recently, very thin tungsten films in the highly resistive ($\rho_{\beta-W} \sim 100–300 \, \mu\Omega \, cm$) metastable $\beta$ phase (A15 crystal structure) were reported to exhibit a giant spin Hall effect with a $\theta_{SH} \sim -0.3$ to $-0.4$ [19–21], the largest spin Hall angle among simple element transition metals. In sharp contrast, thicker and/or annealed W films formed in the relatively low-resistivity ($\rho_{\alpha-W} \leq 25 \, \mu\Omega \, cm$) $\alpha$-W phase (bcc crystal structure) exhibit a small ($\sim 0.07$) spin Hall angle [19]. These results invite the intriguing question as to the origin in the electronic structure of the dramatically different intrinsic spin Hall conductivities and the spin Hall angles of $\alpha$- and $\beta$-W.

In this Rapid Communication, we compare the intrinsic spin Hall conductivity (SHC) of bulk W in the $\alpha$ (bcc) and $\beta$ (A15) phases using first-principles calculations. The calculations reveal that both phases possess large SHC, where the $k$-resolved spin Berry curvature elucidates the resonant feature of the spin-orbit splitting of the doubly degenerate bands near the Fermi level. For $\beta$-W, the large SHC along with the large resistivity work cooperatively to yield a large SHA. We propose the “acceptor” alloying in $\beta$-W can further enhance the SHA from the analysis of the electronic structure. More specifically, Ta substitution locates the Fermi level inside the SOC-induced band gap, rendering the SHA remarkably gigantic, which suggests a way to increase the SHE.

*Ab initio* electronic structure calculations have been carried out within the framework of the projector augmented-wave formalism [22], as implemented in the Vienna *ab initio* simulation package (VASP) [23–25]. The generalized gradient approximation is used to describe the exchange-correlation functional as parametrized by Perdew-Burke-Ernzerhof [26]. The plane-wave cutoff energy is 500 eV and the first Brillouin zone (BZ) is sampled using a $Gamma$-centered special $k$-point grid with $24 \times 24 \times 24$ for $\alpha$-W and $14 \times 14 \times 14$ for $\beta$-W, respectively. All structures are fully relaxed with force criteria of 0.001 eV/Å. The SOC of the valence electrons is in turn included using the second-variation method [27] employing the scalar-relativistic eigenfunctions of the valence states. Then, density functional theory (DFT) wave functions were projected to maximally localized Wannier functions using the WANNIER90 package [28–30] and the Kubo formula [31] was employed to calculate the SHC. A dense $k$ mesh of $100 \times 100 \times 100$ and $90 \times 90 \times 90$ on the full BZ for $\alpha$-W and $\beta$-W, respectively, is employed to perform the BZ integration for the intrinsic SHC because of the slow convergence caused by the large contributions of both signs to the spin Berry curvature which occur in very small regions of $k$ space. The longitudinal charge conductivity is calculated using the Boltzmann transport equation [32].

In the clean case (impurity potential $V = 0$), the intrinsic SHC within the Kubo formalism involves an integration of the

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spin Berry curvature $\Omega_n^c(k)$ of the occupied bands over the BZ [7,33],

$$\sigma_{ab}^c = e/\hbar \int_{BZ} \frac{dk}{(2\pi)^3} \sum_n f_{kn} \Omega_{n,ab}^c(k),$$  

(1)

where $f_{kn}$ is the Fermi-Dirac distribution function for the $n$th band at $k$ and the Berry curvature of the $n$th band is

$$\Omega_{n,ab}^c(k) = -\frac{2 \text{Im}\{(kn|\hat{c}_{\alpha}^n|kn)\langle kn|\hat{c}_{\beta}^n|kn\rangle\}}{(\varepsilon_{kn} - \varepsilon_{kn'})^2}.$$

(2)

Here, $j^c = \frac{1}{2}(s^c, v_0)$, is the spin current operator, $a$, $b$, and $c$ denote the three Cartesian directions $x$, $y$, and $z$, $s^c = (h/2)\beta\Sigma^c$, where $\beta$ is a $4 \times 4$ matrix and $\Sigma^c$ is the spin operator in the Dirac equation, and $|kn\rangle$ represents the periodic part of the Bloch wave function with energy $\varepsilon_{kn}$. Note that the spin Berry curvature can be large near the band degeneracy. The second-rank SHC tensor $\sigma_{ab}^c$ describes the spin current ($J_c$) generated along the $a$th direction with spin polarization along $c$ due to a charge current ($J_\parallel$) flowing along the $b$th direction. Both $\alpha$- and $\beta$-W crystallize in the centrosymmetric cubic lattices: the $\alpha$ phase in bcc with space group $Im\bar{3}m$ (No. 229) while the $\beta$ phase in the so-called $A15$ structure with space group $Pm\bar{3}n$ (No. 223). Consequently, $\sigma_{xy}^c = -\sigma_{yx}^c = \sigma_{yz}^c = -\sigma_{zy}^c = \sigma_{zx}^c = -\sigma_{xz}^c$ by permutation symmetry.

There has been a longstanding issue of the SHC cancellation in the clean limit (impurity potential $V \to 0$) due to the vertex corrections [34]. We expect that the SHC in the clean limit ($V \to 0$) is given by the intrinsic SHC value ($V = 0$) due to the vanishing vertex corrections under the symmetry of $H(k) = H(-k)$ [35,36].

$\alpha$-W. Figure 1 shows the relativistic band structure and the SHC ($\sigma_{xy}^c$) as a function of the Fermi level position $\Delta\mu = \mu - EF$ (EF is the Fermi level) for $\alpha$-W. At $\Delta\mu = 0$, $\sigma_{xy}^c = -785 (h/e) (\Omega \text{ cm})^{-1}$. Its absolute value is smaller than that in Pt [2200 (h/e) (\Omega \text{ cm})^{-1}] [36] but larger than that in Au [400 (h/e) (\Omega \text{ cm})^{-1}] [37]. The magnitude of SHC decreases monotonically as $\Delta\mu$ is raised to about $+2.0$ eV, where the SHC reverses sign. On the other hand, when $\Delta\mu$ is lowered, the magnitude of the SHC increases considerably, reaching the value of $-1058 (h/e) (\Omega \text{ cm})^{-1}$ at $EF = -0.8$ eV (indicated by the green dotted line labeled by 1). The absolute value of $\sigma_{xy}^c$ reaches its maximum value of 1206 (h/e) (\Omega \text{ cm})^{-1} at $\Delta\mu = -2.4$ eV, as denoted by the green line (label 2) in Fig. 1. Further lowering of $\Delta\mu$ results in a decrease of SHC, which eventually becomes small for $\Delta\mu < -5.0$ eV, a consequence associated with the vanishing SOC of the 5s orbital.

In order to elucidate the origin of the SHC change, we show in Fig. 1(b) the $k$-resolved spin Berry curvature $\Omega_x^c(k)$ of the 5th band, where red (blue) denotes a positive (negative) contribution. As shown clearly, double degeneracies which are lifted by spin-orbit coupling result in large SHC. Below $-3.0$ eV, the dominant contribution arises from regions in the vicinity of the high-symmetry point $P$ with $\Omega_x^c(k) > 0$. As the energy increases, the largest contributions to $\Omega_x^c(k)$ arise from the $\Gamma$-$P$ symmetry direction with a coexistence of positive and negative values, resulting in $\sigma_{xy}^c = -1206 (h/e) (\Omega \text{ cm})^{-1}$ for $\Delta\mu = -2.4$ eV. As the Fermi level position further rises, the sign of $\Omega_x^c(k)$ along the $\Gamma$-$H$ direction reverses, leading to another SHC peak at about $-0.8$ eV, and then its magnitude decreases again.

$\beta$-W. Figure 2(a) shows the band structure and the SHC (H(\Omega \text{ cm})^{-1}) as a function of $\Delta\mu$ for $\beta$-W. Interestingly, the SHC is $-1255 (h/e) (\Omega \text{ cm})^{-1}$ at $\Delta\mu = 0$, which is $60\%$ larger than that of $\alpha$-W. The variation of SHC with the Fermi level position is more complicated compared to that for the $\alpha$ case due to the more complex band structure. Nonetheless, $\sigma_{xy}^c$ increases as $\Delta\mu$ is lowered, reaching its maximum value of $-1565 (h/e) (\Omega \text{ cm})^{-1}$ at $EF = -0.4$ eV [green horizontal line 1 in Fig. 2(a)] due to the elimination of the large positive contribution of Berry curvature in the energy window from $-0.4$ to $0$ eV. Remarkably, when the SHC reaches its maximum value, the Fermi level lies in the gap along the $\Gamma$-$X$, where the band degeneracy is lifted by spin-orbit coupling. Figures 2(b) and 2(c) show the $k$-resolved spin Berry curvature in the two-dimensional (2D) BZ ($k_x, k_z$) at $k_z = 0$ for the Fermi level position at 0 and $-0.4$ eV, respectively. As expected, the Berry curvature depends sensitively on $\Delta\mu$ including a sign reversal throughout a large fraction of the BZ, especially along the $\Gamma$-$X$ and around the $Y$-$M$ directions. These changes and the emergence of an overwhelming negative spin Berry curvature in Fig. 2(c) are consistent with the band structure of $\beta$-W.

Temperature dependence of SHC. The temperature dependence of the SHC enters through the Fermi-Dirac function $f_{kn}$ in Eq. (1). We find a weak temperature dependence of the SHC in both the $\alpha$ and $\beta$ phases (a change of less than $2\%$ from low to high temperature) (see Fig. S1 in the Supplemental Material [38]), in sharp contrast to the strong temperature dependence reported in Pt [36], Pd [37], and the TaAs Weyl
The high value of SHC in tungsten, which can be retained even at high temperature, offers a great advantage for room-temperature applications.

**Spin Hall angle.** The SHA can be expressed as

\[ \theta_{\text{SH}} = \frac{e}{\hbar} \frac{\sigma_{xy}}{\sigma_{xx}}. \] (3)

where \( \sigma_{xx} \) is the longitudinal charge conductivity, \( \sigma_{xy} \) is the transverse spin conductivity (SHC), and the prefactor \( \frac{e}{\hbar} \) renders \( \theta_{\text{SH}} \) dimensionless. Obviously, to obtain a large SHA, large SHC as well as low charge conductivity are equally important.

In order to determine the SHA we have also calculated the longitudinal conductivity \( \sigma_{xx} \) using the Boltzmann transport equations within the constant relaxation time approximation [32]. According to the experimental resistivity values of 25 and 300 \( \mu\Omega \text{cm} \) for \( \alpha’ \)- and \( \beta’ \)-W [20,40–42], we find that the corresponding room-temperature relaxation times are 5.52 and 1.61 fs, respectively. Assuming that the relaxation time is independent of \( \Delta\mu \), we show in Figs. S2(a) and S2(b) (see the Supplemental Material [38]) the variation of the calculated \( \sigma_{xx} \) as a function of \( \Delta\mu \) for \( \alpha’ \)- and \( \beta’ \)-W, respectively.

In addition to the larger spin Hall current in \( \beta’ \)-W, its high resistivity leads to lower Joule heating for the generation of the same amount of spin Hall current. The calculations also reveal that lowering \( \Delta\mu \) \( \sim -0.2 \) eV leads to a much larger increase of \( \sigma_{xx} \) compared to the corresponding increase of SHC [Fig. 1(a)] in \( \alpha’ \)-W, which in turn reduces the SHA to about \(-0.01\). In sharp contrast, lowering \( \Delta\mu \) \( \sim -0.2 \) eV in \( \beta’ \)-W decreases \( \sigma_{xx} \) while it increases the SHC, resulting in a 47% enhancement of the SHA value \( \sim -0.47 \) over its value of \(-0.32 \) at \( \Delta\mu = 0 \) eV, suggesting that the SHA can be enhanced by Fermi level lowering [see Figs. S3(a) and S3(b) in the Supplemental Material [38]]. Table I summarizes values of the SHA at \( \Delta\mu = 0 \) eV, the maximum SHC, the SHA at \( \Delta\mu = 0 \) eV, and the maximum SHA for the \( \alpha’ \) - and \( \beta’ \)-W structures, respectively.

**Mechanism of Ta-induced enhancement of SHC.** To elucidate the underlying mechanism of the Ta-induced enhancement of SHC, Fig. 3(d) schematically illustrates the doubly degenerate band around the \( \Gamma \) without SOC (gray curves). The SOC splits this double degeneracy with positive (in red) and negative (in blue) spin Berry curvature. If the Fermi level position \( \Delta\mu \) crosses either one of the SOC-split bands, the Berry curvature contributions of both signs tend to cancel each other. This is the case of pristine \( \beta’ \)-W where \( E_F(P) \) crosses the upper band. On the other hand, if the Fermi level position lies in the spin-orbit-coupling-induced gap, the contribution from one sign of Berry curvature is excluded [positive in Fig. 3(d)], resulting in an enhancement of the SHC. This is

| \( \Delta\mu \) (eV) | \( \Delta\mu = 0 \) eV | \( \Delta\mu = 0 \) eV | \( \Delta\mu \) (eV) | \( \Delta\mu = 0 \) eV |
|------------------|------------------|------------------|------------------|------------------|
| \( \alpha’ \)-W | \( -785 \) | \( -1206 \) | \( > -0.07 \) | \( -0.03 \) |
| \( \beta’ \)-W | \( -1255 \) | \( -1565 \) | \( -0.32 \) | \( -0.40 \) | \( -0.47 \) |

\( ^a \)Reference [19].

\( ^b \)References [19,21].
FIG. 3. (a) A15 crystal structure of β-W consisting of four nonequivalent sites at the center or corners (purple spheres) and at three atomic chains along the x (yellow spheres), y (green spheres), and z (red spheres) directions, respectively. (b) Spin Hall conductivity $\sigma_{xy}^\lambda$ vs Fermi level position $\Delta\varepsilon_F$ for β-W alloyed with tantalum substituting a host atom along the x direction. (c) $k_z$-resolved spin Berry curvature in the 2D BZ $(k_x, k_y)$ at $k_z = 0$ and for the Fermi level position at 0 eV. (d) Schematic diagram of the band structure around the Γ point without SOC (gray bands) and with SOC (red and blue bands) where the red and blue colors denote bands with positive and negative spin Berry curvature, respectively. $E_F(P)$ and $E_F(D)$ denote the Fermi level positions for pristine β-W and Ta-doped β-W, respectively. We also show schematically the variation of the SHC vs Fermi level position.

The case for Ta-doped β-W where $E_F(D)$ lies in the local gap along the high-symmetry Γ'–X direction [see Fig. S3(a) in the Supplemental Material [38]].

Electronic structure calculations are carried out for Ta-alloyed β-W where Ta substitutes one of the W atoms along the y and z chains and at the center sites. We find that the behavior of SHC with Fermi level position is similar to that of Ta occupying a site along the x chain shown in Fig. 3(c) [see Fig. S3(b) in the Supplemental Material [38]], demonstrating that the intrinsic SHC of $W_1-x\_Ta_x$ is remarkably insensitive to variations in the local atomic environment. It should be emphasized that the rigid-band model which shifts the Fermi level inside the spin-orbit-induced local band gap can be used in various systems. Substitution of Ta for W will also cause a large enhancement of the spin Hall effect. Therefore, we provide guidelines to tailor the intrinsic spin Hall effect by a rigid-band model utilizing alloying either with a deficit of or excessive valence electrons.

In summary, using first-principles calculations, we have revealed the physics origin of intrinsic SHC in both tungsten phases. While the β phase exhibits about 60% larger intrinsic spin Hall conductivity than α-W, a much smaller longitudinal conductivity gives a giant spin Hall angle. Moreover, we predict even more gigantic SHC in β-W via Ta alloying $W_1-x\_Ta_x$ (x ~ 12.5%), where a shift of the Fermi level synergistically enhances SHC and reduces the longitudinal conductivity. Finally, we give a general mechanism to improve the intrinsic SHC: Tuning the Fermi level of a system to a spin-orbit-coupling-induced gap can induce a large enhancement of the spin Berry curvature integral. Our results pave an intriguing way towards enhancing the magnitude of SHA and SOT in ferromagnetic/heavy-metal bilayers using Fermi level variations. Our predictions may motivate further experimental studies of the crucial effect of Ta alloying in β-W so as to enhance the SHA and hence optimize the spin-orbit torque efficiency.

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