Concomitance of superconducting spin–orbit scattering length and normal state spin diffusion length in W on (Bi,Sb)$_2$Te$_3$

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Abstract

We report the observation of an anomalously large in-plane upper critical field, exceeding at least 2.5 times the Pauli paramagnetic limit, in a thin superconducting W film grown on a topological insulator (Bi,Sb)$_2$Te$_3$. This can be accounted for by setting the spin–orbit scattering length of superconducting W to a value ranging from 1 to 2 nm, which is comparable to the spin diffusion length of normal state W. The coupling between the topological surface states of (Bi,Sb)$_2$Te$_3$ and the wave functions of superconducting W may also contribute to the observed giant critical field. Our results suggest the universality of the spin–orbit scattering formalism for describing the transport involving the diffusive carriers as well as the Cooper pairs in systems with strong spin–orbit coupling.

1. Introduction

Tungsten (W) crystallizes in two allotropic forms: body-centered-cubic $\alpha$-W and A15-structured $\beta$-W. Thermodynamically stable $\alpha$-W is mostly found in bulk crystals or in thicker films whereas metastable $\beta$-W can be obtained most easily in thin films under certain deposition conditions. Transport properties of the two W phases are profoundly different. $\alpha$-W exhibits a relatively low resistivity ($\rho \sim 20 \mu\Omega \cdot \text{cm}$) with a metal-like positive temperature coefficient of resistance. $\beta$-W typically shows an order of magnitude larger resistivity ($\rho \sim 100$–200 $\mu\Omega \cdot \text{cm}$) and a weak semiconductor-like negative temperature coefficient of resistance. Similarly the superconducting transition temperature at zero field ($T_c$) is enhanced from $\sim 12 \text{mK}$ for bulk $\alpha$-W [1, 2] to a few Kelvins for $\beta$-W films [3] and nanowires prepared by focused ion beam (FIB) [4]. $\beta$-W is also the elemental material known thus far for exhibiting the largest spin Hall angle ($\theta_{SH} \sim -0.3$) [5, 6]. In contrast, the spin Hall angle of $\alpha$-W is significantly smaller mainly due to its lower resistivity [7, 8]. For sputtered W films grown at ambient temperature on Si/SiO$_2$ substrates, W films of lower thicknesses form amorphous-like $\beta$-phase but undergo a phase transition and become $\alpha$-W when the film thickness is larger than a critical thickness of $\sim 4$–6 nm, as evidenced by an abrupt increase of the sheet conductance [8, 9]. High-efficient charge-spin conversion in $\beta$-W also leads to large spin Hall magnetoresistance [10, 11] and spin Nernst magnetoresistance [12, 13] in W/CoFeB bilayers [14–17].

We have studied, down to milliKelvin temperatures, magneto-transport properties of a heterostructure consisting of a thin and resistive sputtered W film deposited on a molecular beam epitaxy (MBE)-grown (Bi$_{0.7}$Sb$_{0.3}$)$_2$Te$_3$ (BST) topological insulator (TI). The n-type Bi$_2$Te$_3$ and the p-type Sb$_2$Te$_3$ parent compounds are both theoretically predicted [18] and experimentally confirmed [18, 20] to be the simplest three-dimensional TIs with surface states forming a single Dirac cone at the $\Gamma$ point. The opposite majority carrier of the two compounds indicates that the Fermi level $E_F$ is located above (below) the Dirac point for $n$-type Bi$_2$Te$_3$ ($p$-type Sb$_2$Te$_3$). The realization of ternary BST compounds of varying Bi:Sb ratio allows continuous tuning of the relative position between $E_F$, the Dirac point and the bulk band gap. The nominal Bi:Sb ratio of our BST film is 7:3, corresponding to $n$-type conduction with the Dirac point and $E_F$ both lie within the bulk band gap. The Dirac point is expected to be slightly above the upper bound of the bulk
valence band, at about 200 meV below $E_F$ [21]. Here, we show that the superconducting transition temperature at zero field ($T_c$) of the BST/W film is $\sim$4.0 K, consistent with the relatively high $T_c$ of $\beta$-W. The superconducting state is robust against an in-plane magnetic field of up to 17.5 T. The giant in-plane upper critical field far exceeds the so-called Pauli paramagnetic limit by at least a factor of 2.5, which we attribute to the strong spin–orbit coupling of the system. We demonstrate using simple arguments that the associated spin–orbit scattering length of superconducting W (ranging from 1 to 2 nm) is comparable to the spin diffusion length of normal state W at room temperature. Thorough understanding of the properties of a superconductor in proximity with a TI may help to design a suitable superconductor/TI heterostructure platform for the study of topological superconductivity that may host the exotic Majorana fermions [22–26].

2. Experiments

The BST film of 8 quintuple layers (QL) was grown on moderately doped Si(111) substrate by MBE in a chamber with a base pressure of $\sim$1 $\times$ 10$^{-9}$ Torr. Prior to the growth, through a cycle of resistive treatment, a clean Si(111)$−7 \times 7$ surface was prepared and verified using reflection high energy electron diffraction (RHEED). The electron energy was 15 keV with incidence along the [112] direction. The growth of BST film was carried out in a Te-rich condition while maintaining the substrate temperature at $\sim$290 °C. The Bi and Sb ratio was controlled by adjusting the relative growth rate of Bi and Sb. The layer-by-layer growth mode was achieved as evidenced by the periodic oscillation of the RHEED signal. By counting the number of oscillations, a thickness of 8 QL for the resulting BST film can be accurately determined. The sharp RHEED pattern shown in figure 1(b) suggests high-quality BST film with a flat surface was obtained. More details of the growth can be found in reference [27].

An elongated Si substrate with an MBE-grown uncapped BST was cleaved ex situ into two dies with approximately 4 mm $\times$ 4 mm square-shape BST film. One die was immediately transferred to a sputtering chamber for subsequent deposition of 3 W$\mid$2 MgO$\mid$1 Ta (thicknesses in nanometer) structure. The top 1 nm-thick Ta layer oxidizes naturally and does not contribute to the conduction. The remaining die was left uncapped which serves as a control sample. Hereafter, we use ‘BST/W’ and ‘BST’ to denote these two dies, respectively. 2 Ta$\mid$15 Cu was sputtered on the four corners of the two square-shape samples by shadow masking to form the electrical contact pads. The sheet resistance, $R_{sq}$ of the structures was measured in the standard Van der Pauw geometry [28] as schematically illustrated in figure 1(a); a constant ac current $I_{ac}$ was applied across two contacts on one side while the resulting ac voltage $V_{ac}$ across the two contacts at the opposite side was picked up using a lock-in amplifier (LIA). For a uniform square-like film, the sheet resistance $R_{sq} \approx \frac{\pi}{\ln 2} \frac{V_{ac}}{I_{ac}}$. The temperature and magnetic field dependences of $R_{sq}$ were measured in a $^3$He-$^4$He dilution refrigerator equipped with a 18 T magnet.

3. Results

The temperature ($T$)-dependences of $R_{sq}$ for BST/W and BST are plotted in linear scales in figures 1(c) and (d) respectively. $R_{sq}$ of BST is found to be at least an order of magnitude higher than that of BST/W. It is therefore reasonable to assume $R_{sq}$ of BST/W mainly reflects that of the 3 nm-thick W layer. $R_{sq}$ for BST/W bilayer increases monotonically from $\sim$360 $\Omega$ at 300 K to $\sim$580 $\Omega$ at 5 K. Neglecting the conduction contribution of BST, we estimate the lower bound of the resistivity of W is $\rho_W > 108 \mu\Omega$cm at 300 K and $\rho_W$ increases to $>174 \mu\Omega$ cm at 5 K, before a sudden drop of the resistance is observed. These relatively high $\rho_W$ values place the material to be close to the so-called Ioffe–Regel limit, whereby the corresponding electron mean free path $l$ is of the order of the atomic lattice spacing (a few Angstroms) [29]. The high $\rho_W$ and its negative temperature coefficient confirm the thin W film grown on BST is in the amorphous-like $\beta$-phase. $R_{sq}$ for BST film without W exhibits non-monotonic temperature dependence which may be attributed to the competition between the bulk and the surface states as well as the shunting due to the semiconducting moderately doped Si substrate. At higher temperatures ($T > 40$ K), contribution from the semiconducting bulk states and/or Si substrate are dominant. $R_{sq}$ decreases with increasing temperature due to increased population of the thermally-excited carriers. At lower temperatures, the conduction is governed by that of the metallic surface states.

In order to highlight the transition associated with the resistance change at low temperatures ($T < 5$ K), we plot the same temperature dependence of $R_{sq}$ for BST/W and BST in log–log scales in figures 1(e) and (f) respectively. For BST/W, we find the measured resistance drops to the noise floor after experiencing a two-step resistance change with an intermediate $R_{sq}$ plateau of $\sim$30 $\Omega$. We interpret this as an evidence of the onset of the superconductivity in W. Previous studies have established that the $T_c$ of W films can be continuously tuned by varying the deposition conditions, whereby increasing the amount of $\alpha$-W level decreases $T_c$ [30]. The two abrupt steps of the superconducting transition in $R_{sq}$ (corresponding to $T_c$ of
\( R_{sq} \) against temperature \( T \) for BST/W (c) and BST (d) plotted in linear scales. The same data plotted in log-log scales are shown in (e) and (f), respectively.

\(~4.0\) K and \(~1.8\) K, respectively) may reflect the existence of two primary structural phases of our W film, which consist of \( \beta\)-W and \( \alpha\)-W mixture of different ratio. By comparing the measured value of the intermediate \( R_{sq} \sim 30 \Omega \) plateau with that of \( R_{sq} \) at \( 5K \) (\(~550\) \( \Omega \) in the normal non-superconducting state), we conclude that the W phase with a lower \( T_{c0} \) has limited (\(~5\%) contribution to the electronic transport at the vicinity of \( T_c \). We shall focus on the characterization of the tungsten phase with a higher \( T_{c0} \) of \(~4.0\) K, which is likely to be rich in \( \beta\)-W. Nevertheless, we emphasize that the relative transport contribution does not reflect the actual volumic ratio of the two phases. When grains and filaments of a superconducting phase are embedded in a major non-superconducting phase, depending on the aspect ratio and the distribution of the grains, a moderate critical composition is typically sufficient for realizing the zero resistance state. Instead, the sampling of a large number of grains using nanobeam electron diffraction should provide a better estimation of the actual volumic ratio of the two phases.

For the BST reference film, we have verified that no superconducting transition is observed down to the base temperature (\(~20\) mK) achieved in this study. Below 2 K, a gradual decrease of \( R_{sq} \) by \(~20\%) may be attributed to the weak antilocalization (WAL). WAL and weak localization induced resistance correction stems from the quantum interference of coherent back-scattered conduction electrons [31]. At sufficiently low temperature, as the coherence length of electrons gets longer, such a correction becomes increasingly pronounced. Application of a magnetic field introduces a magnetic phase shift that competes with the interference and eventually destroys the latter. In TIs, WAL or a destructive interference is expected for both the surface and bulk contributions to the electronic transport [32].

We next focus on the suppression of the superconductivity in BST/W by an applied magnetic field \( B \). The field dependence of \( R_{sq} \) measured at 25 mK for \( B \) along varying polar angle \( \theta \) in steps of \( 10^\circ \) and the corresponding \( dR/dB \) mapping are shown in figures 2(a) and (b) respectively. At such a low temperature relative to \( T_{c0} \), the extracted characteristic parameters of the superconductivity are sufficiently close to their values at zero-temperature limit. We found the superconductivity of BST/W heterostructure was suppressed by a perpendicular upper critical field \( B_{c0} = 5.9 \) T. The critical field progressively increases with increasing \( \theta \), reflecting the strong anisotropy of \( B_{c0} \) owing to the thin slab geometry of the heterostructure. For \( \theta = 90^\circ \), application of a maximum in-plane field \( B_{||} = 17.5 \) T does not induce noticeable change in \( R_{sq} \), thus setting a lower bound for the in-plane upper critical field \( B_{c0} \).

We now discuss the dimensionality of the superconductivity in \( \beta\)-W. In the zero temperature limit, the superconducting coherence length \( \xi_0 \) can be expressed as a function of \( B_{c0} \) [33]:

\[
\xi_0 = \sqrt{\frac{\Phi_0}{2\pi B_{c0}}} 
\]

Using \( B_{c0} = 5.9 \) T and the magnetic flux quantum \( \Phi_0 = h/(2e) \), where \( h \) and \( e \) are the Planck constant and electric charge, respectively, we found \( \xi_0 \approx 7.4 \) nm which is longer than the thickness of the W film (\(~3\) nm).
Figure 2. (a) $R_{\text{sq}}$ against applied field $B$ for BST/W bilayer measured at 25 mK with the polar angle $\theta$ of the field vector varied in steps of $10^\circ$. (b) $dR/dB$ color mapping in the $B-\theta$ space. Dashed/dotted (red solid) curves are calculated based on equation (2) (equation (4)) using a fixed perpendicular upper critical field $B_{c\perp 0} = 5.9$ T and different in-plane upper critical fields $B_{c\parallel 0} = 25, 38, \text{and } 60$ T (28, 45 T) as parameters. The experimental data are best described by the blue dotted curve corresponding to $B_{c\parallel 0} = 38$ T (28 T).

(c) and (e) $R_{\text{sq}}$ against perpendicular applied field $B_{\perp}$ (c) and in-plane applied field $B_{\parallel}$ (e) measured at various temperatures $T$. (d) $dR/dB$ color mapping in the $B_{\perp}-T$ space. The blue dashed line is a linear extrapolation of the perpendicular critical field $B_{c\perp}$. (f) $B_{\parallel}-T$ phase diagram with the blue shaded region representing the field and temperature ranges examined in this work. Solid lines and green dashed line are obtained from equation (5) using $\tau_0$ values given in the legend. The black horizontal dashed line indicates the Pauli paramagnetism limit of the critical field $B_{p}$ for a superconductor with $T_{c0} = 4.0$ K. All results shown are from BST/W bilayer.

and the electron mean free path (a few Angstroms) in this highly disordered bad metal. We may therefore infer, in the zero temperature limit, that the superconductivity in BST/W is essentially two-dimensional (2D) and it is in the “dirty limit”. In the 2D superconductor limit, the angular dependence of $B_c$ is described by the following implicit relation [34, 35]:

$$\frac{B_c(\theta) \cos \theta}{B_{c\perp 0}} + \left( \frac{B_c(\theta) \sin \theta}{B_{c\parallel 0}} \right)^2 = 1$$

(2)

$B_{c\parallel 0}$ can be estimated by fitting $B_c(\theta)$ in figure 2(b) using equation (2).

Alternatively, one may assume the electronic transport within and normal to the plane of BST/W thin film are anisotropic due to, e.g. broken inversion symmetry at the interfaces and/or the presence of a 2D Rashba-type spin–orbit interaction. Consequently, we may define two sets of coherence lengths $\xi_{\parallel 0}$ and $\xi_{\perp 0}$ (the subscripts represent the corresponding $k$-vector direction). Under such circumstances, while equation (1) gives an estimation of the in-plane coherence length $\xi_{\parallel 0}$ via $\xi_{\parallel 0} \equiv \xi_0$, we have in addition:

$$\xi_{\perp 0} = \frac{\Phi_0}{2\pi B_{c\parallel 0}\xi_{\parallel 0}}$$

(3)

In this three-dimensional (3D) anisotropic superconductor case, the implicit relation describing the angular dependence of $B_c$ is slightly modified [33, 35]:

$$\left( \frac{B_c(\theta) \cos \theta}{B_{c\perp 0}} \right)^2 + \left( \frac{B_c(\theta) \sin \theta}{B_{c\parallel 0}} \right)^2 = 1$$

(4)
Table 1. Summary of the transport characteristic lengths of BST/W. We use $\tau_{so}$ ranging from 20 fs to 38 fs obtained using the 3D anisotropic model (equation (1)) and the 2D model (equation (2)), respectively. (The corresponding $B_{c0}$ ranges from 28 T to 38 T.) We assume $D = 0.51 \text{ cm}^2 \text{s}^{-1}$.

| Transport    | Carrier         | Parameter                          | Length       |
|--------------|-----------------|------------------------------------|--------------|
| Diffusive    | electron spin   | mean free path, $l$                 | 0.7 nm       |
|              |                 | spin diffusion length, $l_s$        | 1.1 nm (from SMR) [14] |
| Superconducting| Cooper pair    | In-plane coherence length, $\xi_{\parallel}$ | 7.4 nm       |
|              |                 | Out-of-plane coherence length, $\xi_{\perp}$ | 1.6 nm       |
|              |                 | spin–orbit scattering length, $l_o$  | 1.0 $\sim$ 1.4 nm |

We estimate $B_{c\parallel}$0 using the two models. We first fit $B_c(\theta)$ in figure 2(b) using equation (2) in the 2D limit. Upon fixing $B_{c\perp} = 5.9$ T, $B_{c\parallel}$0 is the only free parameter of the fit. We find the experimental data is best described by $B_{c\parallel} \sim 38$ T with the corresponding fitted curve shown by the blue dotted line. In order to test the robustness of the analysis, we plot in black dashed lines calculation results based on equation (2) with $B_{c\parallel} = 25$ and 60 T. The fit provides a better constraint to the lower bound of $B_{c\parallel}$0 whereas it is ineffective in setting an upper limit for $B_{c\parallel}$0. The red solid curves in figure 2(b) represent the fits using equation (4) for 3D anisotropic superconductor with $B_{c\parallel} = 18.5$, 28 and 45 T, respectively. For $\theta < 70^\circ$, the two models predict undistinguishable angular dependence of $B_c(\theta)$ (with slightly lower $B_{c\parallel}$0 for the 3D model). Limited by the available magnetic field, fitting on the current experimental data does not allow us to unambiguously identify the dimensionality of the superconductivity in BST/W. Nevertheless, our primary observation that $B_{c\parallel}$0 of BST/W is several times higher than $B_{c\perp}$0 is robust against the choice of the model. The corresponding in-plane and out-of-plane coherence lengths are summarized in table 1.

The $B_{c\parallel}$ dependence of $R_q$ measured at various temperatures are shown in figure 2(c). As evidenced by the corresponding $dR/dB$ mapping in figure 2(d), the sharpness of the superconducting transition for BST/W under an applied $B_\perp$ remains virtually invariant throughout the temperature range in this study. The out-of-plane critical field $B_{c\perp}$ appears to decrease linearly with increasing temperature, as expected from the Ginzburg–Landau theory. However, we should note that the linear extrapolation (blue dashed line) to the $x$-axis yields a critical temperature of $\sim 2.6$ K, which is small compared to $T_{c0} \sim 4 K$ obtained from a $R_q(T)$ scan at zero field. A slope change of $B_{c\perp}(T)$ of yet unknown origin may have occurred in the temperature range not covered by this work. We shall note that the slope change in $B_{c\parallel}(T)$, previously observed in W/Si multilayer [36] and interpreted in terms of the crossover from 2D decoupled to 3D coupled (across the Si spacers) superconducting states is not relevant in our BST/W bilayer.

In conventional superconductors, the quenching of the superconductivity under the application of an external magnetic field is understood as the collective effect of the electron’s orbital motion [33], spin Zeeman splitting [37, 38], and spin–orbit scattering [39, 40]. In the limit of a two-dimensional superconductor, the contribution of the orbital effect is small for an applied $B_\parallel$. If the spin–orbit scattering is negligible, a transition to the normal state occurs when the Zeeman pair-breaking energy due to an in-plane field matches the binding energy of Cooper pairs, i.e. the superconducting energy gap $\Delta_0$. This critical field $B_\parallel$, commonly being referred to as the Pauli paramagnetic limit, reads $B_\parallel = \Delta_0/\sqrt{\mu_B}$. Using $\Delta_0 = 1.76k_BT_{c0}$ based on the Bardeen, Cooper, and Schrieffer (BCS) theory, and assuming an electron $g$-factor of 2, we obtain $B_\parallel = 1.85 T_{c0}$ (in Tesla and Kelvin, respectively). For the two superconducting phases of W, we estimate $B_\parallel = 7.4$ T and 3.3 T, respectively. Comparing $B_\parallel$ and a lower bound of $B_{c\parallel}$0, set by the maximum available field of 17.5 T, it is clear that the Pauli limit is largely surpassed in superconducting W.

The $B_{c\parallel}$ dependence of $R_q$ measured at various temperatures are plotted in figure 2(e). $R_q$ is at the noise floor throughout the field scans, except for $T = 1.6$ K where a slight upturn of $R_q$ in high fields is visible. We note that the resistance of this slight upturn is of the same range as the intermediate $R_q$ plateau observed in the $R_q$ versus $T$ scan at zero applied field in figure 1(e). It is therefore reasonable to assume that this upturn corresponds to the suppression of the superconductivity in the secondary W phase with $T_{c0} \sim 1.8$ K. We have therefore confirmed experimentally that $B_{c\parallel}$0 remains larger than 17.5 T for an extended temperature range up to 1.6 K. This conclusion is robust, regardless of our choice on the critical field definition (whether it corresponds to the field where the resistance drops to a certain percentage with respect to the normal state resistance or where $dR/dH$ is at the maximum). Previously, there are a few reports focusing on the large critical fields of W/Si multilayers near $T_{c0}$ [36, 41], for which $B_{c\parallel}$0 can be extracted by extrapolation. By examining BST/W down to milliKelvin temperatures, here we provide more direct estimation of the giant $B_{c\parallel}$0 which may partly originate from the strong spin orbit coupling and/or surface states of BST.

In view of the strong spin–orbit coupling of W and that the conduction of the supercurrent is in the dirty limit, it is tempting to attribute the anomalous enhancement of $B_{c\parallel}$0 in BST/W to the spin–orbit scattering.
mechanism due to disorder. In such a case, the associated pair-breaking implicit relation reads [33]:

\[
\ln \frac{T_c}{T_{c0}} = \psi \left( \frac{1}{2} \right) - \psi \left( \frac{1}{2} + \frac{(g\mu_B B')^2 \tau_{so}}{4\pi l_0 k_B T_c} \right)
\]

(5)

where \(\psi(x)\) is the digamma function and \(\tau_{so}\) is the characteristic spin–orbit scattering time. In line with the analysis in figure 2(b), for \(T_{c0} = 4.0\) K, we numerically solve equation (5) to find \(\tau_{so}\) that satisfy \(B_{c0} = 28\) and \(38\) T for the 3D anisotropic and 2D superconductor models, respectively. The results are plotted in the \(B\)-\(T\) superconducting phase diagram in figure 2(f). The blue shaded region represents the temperature and the field range accessible in this study, for which the robustness of the superconductivity was experimentally confirmed. We find \(\tau_{so} \sim 20\) and \(38\) fs respectively for the secondary W phase with a lower \(T_{c0} \sim 1.8\) K (figure 2(f), green dashed line). For \(B_{||} \sim 12\) T at 1.6 K, the model predicts \(B_{c0} \sim 30\) T with \(\tau_{so} \sim 15\) fs. Here, the \(\tau_{so}\) estimation is more sensitive to the choices for defining \(B_{||}\) and \(T_{c0}\).

\(\tau_{so}\) is found to be in a range of a few tens of femtosecond, which is an order of magnitude smaller than that found in the gated-induced superconducting monolayer WTe\(_2\) [42, 43]. Considering the spin–orbit coupling of elemental W should be stronger than that of WTe\(_2\) and the higher defect/disorder density in sputtered film, such an enhancement of the spin–orbit scattering rate may not be unreasonable. As a side note, the Pauli limit can be alternatively overcome through the spin-valley-locking Ising pairing mechanism, as being demonstrated in gated MoS\(_2\) thin flakes [44, 45] and monolayer NbSe\(_2\) [46]. However, such a process requires crystal structures with broken inversion symmetry, which is irrelevant for our amorphous polycrystalline heterostructure.

In order to examine the validity of the ultrafast \(\tau_{so}\) and gain more insight into its physical meaning, we attempt to compare this quantity with the very short, nearly temperature-independent spin diffusion length \(l_s \sim 1.2\) nm extracted from the thickness and temperature dependence of spin Hall magnetoresistance in W/CoFeB bilayers [14]. We first note that the characteristic spin–orbit scattering rate is related to a spin–orbit scattering length \(l_{so}\) via \(1/\tau_{so} = D/l_{so}^2\) where \(D\) is the diffusion constant. Similarly, we define the spin relaxation rate \(1/\tau_s = D/l_s^2\). We note that \(D\) of a weakly coupled superconductor in the dirty limit reads [33]:

\[
D = \Delta \phi^2_0 / \hbar = \Delta_0 (2eB_{R\perp,0}) = 0.51 \text{ cm}^2 \text{s}^{-1}
\]

(6)

As pointed out in references [47, 48], \(l_{so} \neq l_s\) since \(l_{so}\) includes both spin-flip and spin-conserving events. Assuming isotropic scattering (such that the ratio of the spin-flip and spin-conserving scattering is 2:1) and noting that the spin relaxation rate is twice the spin-flip scattering rate [47], we can write \(1/\tau_s \approx 4/(3\tau_{so})\) or in terms of the characteristic lengths, \(l_s^2 \approx 3l_{so}^2/4\). Substituting \(\tau_{so}\) and equation (6) into this relation, we obtain within a simplified, single-band picture:

\[
l_s^2 \approx \frac{3}{4} D \tau_{so} \approx \frac{3\Delta_0 \tau_{so}}{8eB_{R\perp,0}}
\]

(7)

For \(\tau_{so} = 38\) fs and \(20\) fs (c.f. legends of figure 2(f)), we obtain \(l_s = 1.2\) nm and \(0.9\) nm (the corresponding \(l_{so}\) is \(1.4\) nm and \(1.0\) nm), respectively, which are well within the range of \(l_s\) obtained from the spin Hall magnetoresistance measurement for W/CoFeB bilayers. For a thin film superconductor with strong spin–orbit coupling, equation (7) provides an estimation of \(l_s\) solely based on the experimental critical fields along two orthogonal directions and the zero-temperature gap. We further note that the squared exponent for \(l_s\) in equation (7) ensures that the extracted \(l_s\) range will be less sensitive to any fluctuation of the parameters such as the value of \(D\) or the validity of the isotropic scattering assumption.

Since the accuracy of equation (7) partly depends on that of \(D\), as an independent veriﬁcation, we apply the Einstein relation \(D = 1/(e^2 / \rho_{so} N^*)\) (\(N^*\) being the effective density of states at \(E_F\) that actively participate in the conduction) and use \(\rho_{so} \approx 180\) \(\mu\)\Omega \text{ cm} to estimate \(D\) for W. We note that contrarily to equation (6), here only the normal state transport parameters are involved. For a typical monovalent transition metal, e.g. fcc-Copper(Cu), \(N^* = 2.5 \times 10^{-28}\) states \(\text{eV}^{-1} \text{m}^{-3}\) or 0.30 states \(\text{eV}^{-1} \text{atom}^{-1}\). For A15 β-W structure with a lattice parameter of 0.505 nm and containing 8 W atoms per unit cell, we ﬁnd \(N^* = 0.41\) states \(\text{eV}^{-1} \text{atom}^{-1}\) and thereby obtain \(D = 1.4\) \(\text{cm}^2 \text{s}^{-1}\). This is slightly larger but still of the same order of magnitude compared to the \(D\) obtained using equation (6). Similar agreement between different estimations of \(D\) has also been reported for WTe\(_2\) monolayer [43].

The valence electronic conﬁguration of Cu is 4s\(^{1}\) 3d\(^{10}\). Since the d-band of Cu is fully ﬁlled, the DOS at the Fermi level of Cu is practically only of s character. A direct consequence of the absence of unoccupied d-band at the Fermi level is the prohibition of s – d interband scattering, making Cu an excellent electrical conductor [49]. The orbital ﬁlling of W, according to the Hund’s laws, is 6s\(^2\) 5d\(^{4}\). In realistic band structure, 

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Y-C Lau et al
the DOS at the Fermi level of W is consisted of states of s and d character. The less-than-half filling of the d-band leads to a large DOS at the Fermi level with highly localized d electrons. Neglecting the contribution from the d-shell and counting only the s conduction electrons, we arrive to the above approximation that the effective N for Cu and W (and many other transition metals) that contributes to the conduction should be comparable.

Lastly, we verify the prerequisite that the mean free path l must be shorter than lso and ls. We estimate l using an expression derived from the free electron model for a three-dimensional system [29]:

\[ l = \frac{3\pi^2 \hbar}{e^2 k_F^2 \rho_N} \]  

with the Fermi wave vector \( k_F = \sqrt{3\pi^2 n} \) and \( n \) is the effective carrier density. Assuming 0.5 conduction electron per W atom, we obtain \( l \sim 0.71 \text{ nm} \), which is indeed approaching the Ioffe-Regel limit, at about 1.4 times the lattice constant \( a \) and is shorter than \( l_s \) or \( l_{so} \).

We next comment on the implications of equation (7). First, although there are a number of reports that have attributed the violation of Pauli limit to a short spin–orbit scattering length \( l_{so} \) or a high spin–orbit scattering rate \( \tau_{so}^{-1} \) in superconductors [50–53], most of the discussions focused on the atomic number \( Z \) dependence. To the best of our knowledge, this work represents the first attempt to relate \( l_{so} \) in the superconducting state to \( l_s \) of the same material in the normal state, which is extracted from other independent experiments in spintronics. These results suggest the characteristic length scales for spin–orbit scattering and spin relaxation of the normal and the superconducting states of W share a common origin.

Such a compatibility is a priori non trivial. As pointed out above, s-electrons dominate the transport in the normal metallic state such that the contributions from d-electrons can often be neglected. This may not be true in the superconducting state. Notably, the presence of a sharp, narrow peak of d-like electron's density of states at the Fermi level has long been regarded as a crucial key for achieving relatively high \( T_{c0} \) in many A15-structured \( A_2B \) compounds [54]. Yet, our results indicate the fermionic s-like electron's wave function in the normal conducting state of W and the bosonic Cooper pair's wave function in superconducting W are subjected to a similar characteristic spin–orbit scattering length scale.

Second, although a more precise determination of \( \tau_{so} \) is hindered, in this study, by the exceptionally high \( B_{||0} \) of W, the quadratic dependence of \( \tau_{so} \) on \( l_s \) allows us to conclude with high confidence that the spin diffusion length of \( \beta \)-W is indeed of the order of 1 nm, even in the superconducting state. The presence of an adjacent ferromagnetic layer used in the normal state spin transport experiments seems to hardly affect \( l_s \) of W. Previously, the presence of heavy metal/ferromagnet interface was thought to be responsible for the disagreement between \( l_s \) of Pt in contact with Cu channel (measured using the non-local spin valve and WAL techniques) [48] and that of Pt/ferromagnet bilayers estimated via ferromagnetic resonance, harmonic Hall or spin Hall magnetoresistance methods [55–58]. Alternatively, Rojas Sanchez et al [59] proposed the apparent high dispersion in literature on the quantification of the spin diffusion length and the spin Hall angle of Pt is due to the intercorrelation between these two quantities and the conductivity of Pt. Here, we do not observe noticeable change of \( l_s \) of superconducting BST/W, compared to that of W/ferromagnet bilayers obtained from spin Hall magnetoresistance [14]. We therefore conclude that the presence of an adjacent ferromagnetic layer does not play a central role in determining the short \( l_s \) of W.

Third, historically, certain compounds/structures e.g. TaS\(_2\) intercalated by Pyridine [52] and more recently Ta\(_2\)Pt\(_5\)S\(_2\) [53] were found to exhibit very large \( B_{||0} \) and consequently very fast \( \tau_{so} \) comparable to that we have found here in BST/W bilayer. A combination of strong spin–orbit coupling, short spin diffusion length and the diffusive transport in the dirty limit makes these materials potential candidates for investigating spin current generation via the spin Hall effect or the detection of spin current via the inverse process. Conversely, an excellent spin Hall material thin film in the dirty limit should exhibit large upper critical in-plane field far exceeding the Pauli limit if it were to become a superconductor. Superconducting WTe\(_2\) monolayer exhibiting a \( B_{||0}/B_p \) ratio > 2 [42, 43] is potentially an excellent example. Although large charge-spin conversion efficiency is yet to be demonstrated in the monolayer limit, high bulk-like spin Hall efficiency in the same material has been reported recently [60].

So far, we have shown that our experimental observations can be quantitatively understood by setting \( l_{so} \) to \( \sim 1 \text{ nm} \). Concerning the role of the underneath BST, the topological surface states of BST may contribute to the high anisotropy of the critical field by directly influencing the spin–orbit scattering rate in W or via the interaction between the superconducting quasiparticles at the interface and those in the bulk, if a 2D topological superconducting layer is formed at the interface. Unfortunately, current experimental data does not allow us to confirm nor exclude these possibilities. We speculate that a careful study of \( B_{c1}(T) \) in both field directions over a series of bilayer samples with varying interface quality may reveal evidences of these second-order effects.
4. Conclusion

In summary, we have investigated the superconducting characteristics of BST/W bilayer. We find a superconducting state exhibiting an in-plane upper critical field far exceeding the Pauli paramagnetism limit. A comparatively short spin–orbit scattering length, of the order of 1–2 nm, can account for the giant critical field. Interestingly, the length scale involved in defining the critical field, i.e. the spin–orbit coupling, is similar in magnitude with that of the spin diffusion length of the normal state W state. These results suggest that the spin–orbit scattering in normal and superconducting W can both be described by the same characteristic length scale, albeit the involvement of carriers that are distinct in nature. It remains an open question, whether the coupling between the topological surface states of BST and the Cooper pairs of superconducting W may influence the upper critical fields. We expect, by further improving the sample preparation (e.g. in situ deposition of the full stack) and controlling the interface quality, stronger coupling between a superconductor with large spin–orbit coupling (e.g. W) and spin-momentum locked surface states of topological insulators (e.g. BST) can be achieved. Since application of a symmetry-breaking magnetic field is a powerful means for studying the properties of novel quantum mechanical states, the establishment of a superconductor/topological insulator hybrid heterostructure platform that is robust against external field perturbations may pave ways towards the realization and facile characterization of more exotic topologically non-trivial phases, including the Majorana fermions.

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