Non-orthogonal spin-momentum locking in spin-orbit coupled materials

Tugrul Hakioglu \textsuperscript{1,1,*} Wei-Chi Chiu\textsuperscript{2,†} Robert S. Markiewicz\textsuperscript{2} Bahadur Singh\textsuperscript{3} and Arun Bansil\textsuperscript{2}

\textsuperscript{1}Energy Institute and Department of Physics, Istanbul Technical University
Maslak 34469, Istanbul, Turkey
\textsuperscript{2}Department of Physics, Northeastern University, Boston, MA 02115, USA
\textsuperscript{3}Department of Condensed Matter Physics and Materials Science, Tata Institute of Fundamental Research, Colaba, Mumbai 400005, India

Spin-momentum locking is a unique intrinsic feature of strongly spin-orbit coupled materials and a key to their promise of applications in spintronics and quantum computation. Much of the existing work, in the context of both topological and non-topological pure materials, has been focused on an orthogonal locking in the vicinity of the \(\Gamma\) point where the directions of spin and momentum vectors are locked perpendicularly. Mechanisms responsible for non-orthogonal spin-momentum locking (NOSML) have drawn little attention, although an NOSML effect has been reported on the topological surface of \(\alpha\)-Sn. Here, we demonstrate that, the presence of the spin-orbit scattering from dilute non-magnetic impurities can produce the NOSML state in the presence of a strong electronic spin-orbit coupling in the pristine material. We also observe an interesting coupling threshold for the NOSML state to occur. The relevant parameter in our analysis of the NOSML is the deflection angle from orthogonality which can be extracted directly from the spin-and- angle-resolved photoemission (S-ARPES) spectra. Our formalism is applicable to all strongly spin-orbit coupled systems with non-magnetic impurities and not limited only to topological ones. An understanding of NOSML effects bears on spin-orbit dependent phenomena more generally, including issues of spin-to-charge conversion and the interpretation of quasiparticle interference (QPI) patterns as well as scanning-tunneling spectra (STS) in general spin-orbit coupled materials.

I. INTRODUCTION

Spin-momentum locking (SML) occurs commonly in spin-orbit coupled low dimensional materials with or without topological bands\cite{1–4}. Its telltale signatures involve forbidden backscattering\cite{5, 6} from non-magnetic impurities (no ‘U-turn’) and enhancement of weak antilocalization effects \cite{7}. SML enables electrical control of spin polarization in nonequilibrium transport and thus plays a key role in spintronics and spin-based quantum information sciences applications \cite{2, 8} by driving the presence of a spin-polarized component of current with polarization perpendicular to the current density \cite{9–11}.

The orthogonal SML (OSML)--see Fig.1.a, is common in materials exhibiting SML\cite{1–3}. The OSML state is the result of in-plane Rashba spin-orbit coupling (SOC) observed first time on the Au (111) surface long before the topological materials were discovered\cite{12}. In topological insulators, OSML with a \(\pi\)-Berry phase is an essential feature of the surface electron bands\cite{13}. It has been utilized in the electrical detection of magnon decay\cite{14}. Violations of the spin-momentum locked state has been widely seen in real materials. In such cases, they are weakly locked within a narrow range of angles constrained by the crystal symmetries. An S-ARPES study of the Au/Ge (111) surface revealed such examples\cite{15} and similar effects have been reported in high-temperature superconductors\cite{16}. In certain topological insulators the spin wiggles around the Fermi surface due to the hexagonal warping but respecting the OSML\cite{17} although deviations from the orthogonal picture were observed experimentally in the \(Bi_{2-\delta}Sb_{\delta}Te_{x}Se_{3-x}\) family\cite{18} as shown in the Fig.1.b. We can call these as type-I violations of the OSML. It has been shown that high-order corrections to the theoretical \(k.p\) Hamiltonian can induce deviations from the orthogonal picture\cite{19}. Many body interactions also cause similar effects, as the electron-phonon interaction in this material\cite{20–23} was recently studied in this context\cite{24, 25}. The triple and septuple windings of the spin vector have also been studied theoretically\cite{26, 27}.

Another type of deviation from the perfect OSML state is not in the locking phenomenon but in the orthogonality, i.e. the non-orthogonal spin-momentum locking (NOSML) as illustrated in Figs.1.c and d (called as the type-II violations of the OSML). Such a state has been reported on the topological surface of strained \(\alpha\)-Sn\cite{28, 29}. Here, S-ARPES and Mott polarimetry reveals the presence of a radial component of the spin (Fig1.c) with a significant inward deviation of \(\Phi_0 - 90^\circ \simeq 20^\circ\) on a circular Fermi surface. The out-of-plane spin \(S_z\) is observed to vanish in conformity with the absence of the out-of-plane SOC. Note that \(\alpha\)-Sn is inversion symmetric in unstrained and strained phases \cite{29–36}. The authors of Ref.\cite{28} also point at the presence of electron-impurity interaction through an analysis of the electronic self-energy.

This last observation is of key importance in our theory of the NOSML. Our approach is not limited to topological surface states but addresses NOSML as a general phenomenon in materials with strong SOC. The presence of inversion and time-reversal symmetries substantially
II. THE THEORY OF INTERACTING SPIN

Our starting point is the time-reversal invariant Hamiltonian in the pseudo-spin $|\sigma\rangle$ basis ($\hbar = 1$)[1, 37, 38]:

$$H_0 = (\xi_k - \mu)\sigma_0 + g_k \cdot \sigma$$  \hspace{1cm} (1)

where $\sigma = (\sigma_x, \sigma_y, \sigma_z)$ is the pseudospin representing the spin-orbit coupled total angular momentum states[1], $\mathbf{k} = (k_x, k_y) = k (\cos \phi, \sin \phi)$ is the electron wavevector relative to the Dirac point at $\mathbf{k} = 0$ and $\xi_k$ and $\mu$ are the spin-independent and isotropic bare electron band and the chemical potential, respectively. The Hamiltonian in Eq.(1) is the most basic Hamiltonian in spintronics as well as topological surfaces. A pair of such Hamiltonians can be used to model states in Dirac and Weyl semimetals as well as Rashba type interface states. The spin-orbit vector $g_k$ is normally composed of an in-plane component $g_k^\parallel = g_0 \hat{z} \times \mathbf{k}$ with $g_0$ as the Rashba type in-plane SOC and $\hat{z}$ as the surface unit normal vector to the $xy$ plane defined by the $\mathbf{k}$ vector, and an anisotropic out-of-plane component $g_k^\perp$. We also represent the in-plane and out-of-plane components of the spin as well as the self-energy vectors below using the same notation. The $g_k^\perp$ as well as the out-of-plane component of the spin are zero in our case due to the azimuthal rotational symmetry. The eigenstates $|\mathbf{k}\lambda\rangle$ of Eq.(1), where $\lambda = \pm$ is the spin-orbit band index, include the chiral spin-$1/2$ state not only attached to the dominant $|p_z\rangle$ orbitals as considered conventionally, but also the in-plane orbitals $|p_x\rangle$ and $|p_y\rangle$. The role played by the in-plane orbitals is strongly material dependent, which has been demonstrated experimentally[39] and theoretically[40]. It is known that these effects do not violate the OSML[13, 17]. For our purposes in this work we ignore these in-plane orbitals and consider that the orbital texture is solely determined by the out-of-plane $|p_z\rangle$ orbitals. Further discussion on this point is made in Section V. With this considered, the pseudospin is given by $\langle J \rangle = (\mathbf{k}|\sigma|\mathbf{k}\lambda) = (\lambda/2)\hat{g}_k$, where $\hat{g}_k$ is the unit spin-orbit vector, which coincides with the actual spin $S^\lambda(\mathbf{k}) = (\lambda/2)\hat{g}_k$. Since the in-plane component $g_k^\parallel$ of the spin-orbit vector $\mathbf{g}_k$ is perpendicular to $\mathbf{k}$, the spin $S^\lambda(\mathbf{k})$ is locked orthogonally to the electron momentum $h\mathbf{k}$ throughout, yielding the OSML.

The Eq.(1) is clearly insufficient to describe all strongly spin-orbit coupled surfaces and additional terms may be present due to the symmetries. For instance, the cubic Dresselhaus SOC is present in the absence of inversion symmetry in ordinary semiconductors[1]. Its realization in $Bi_2X_3$ type strong topological insulators results in the hexagonal warped Fermi surfaces but the OSML is still respected[17]. Note that, NOSML is ideally an isotropic effect and anisotropic warping in the band structure may hinder its observation. Therefore, we ignore the warping anisotropy and examine symmetry allowed Hamiltonians in the study of the NOSML state. The minimal Hamiltonian which can yield a NOSML state is[1, 37, 38]

$$H_1 = \gamma \mathbf{k} \cdot \sigma$$ \hspace{1cm} (2)

This Hamiltonian is known to be present in the Kane model between the $\Gamma_{7c}$ and $\Gamma_{6c}$ bands of zinc-blende structures[1, 41, 42]. The total Hamiltonian $H_0 + H_1$ is equivalent to $H_0$ by a rotation of the spin-orbit constant in the complex space, and effective spin-orbit coupling is defined as $g_k = g_0 \hat{z} \times \mathbf{k} + \gamma \mathbf{k}$. The energy spectrum is linear and spin-momentum pair is locked non-orthogonally at $\Phi_k = \pm (\pi/2 + tan^{-1}\gamma/g_0)$ with the $\pm$ describing the upper and the lower Dirac cones. While, Eq.(2) can easily accommodate a non-orthogonal state of the spin and momentum in zinc-blende structures[42], it is not applicable when the inversion symmetry holds since that requires $\gamma = 0$. This prompts us to think that the NOSML in inversion symmetric systems may have its origin fundamentally beyond the class of symmetry allowed single particle Hamiltonians.

Here we demonstrate that, the impurities in the real materials may provide a striking clue for the general source of the NOSML. It is known that the electron-impurity interaction, combined with the strong SOC, gives rise to the spin-orbit scattering in addition to the scalar scattering channels, which then leads to a number of observable transport phenomena. These are linearly dependent on the spin-orbit scattering strength[43] such as corrections in the mo-
mentum and spin relaxation, spin-dependent diffusion, weak localization/antilocalization[2] and anomalous spin-texture[44]. The spin-orbit scattering between the impurity and the electron bands also provides a platform for NOSML and this is the main focus of this work.

In this work, we use the interacting Green’s function formalism for the renormalized spin[25]. In this approach the spin is given by

\[ S_\lambda(k) = \frac{\lambda}{2} G(K^*) \]  

(3)

Here, the * indicates that \( S_\lambda(k) \) is calculated at the physical energy pole position \( E^* = E_{\lambda k} \) of the full Green’s function[25]. \( G(K^*) = g_k + \Sigma(K^*) \), with \( K^* = (k, iE^*) \) in the Matsubara Green’s function formalism, is the renormalized spin-orbit vector where \( G = G/G \) is its unit vector. Here it is crucial to note that, \( G \) enters as a simple sum of the spin-orbit vector \( g_k \) of the non-interacting structure and the interactions represented by the spin-dependent self-energy \( \Sigma \) (SDSE). The \( g_k \) is aligned perpendicularly to the momentum. The renormalized spin-orbit vector \( G \) however, develops a non-orthogonal component as a result of the interactions. We recently demonstrated this theoretically using the electron-phonon interaction and the Fermi surface warping yielding six-fold symmetric type-I violations of the OSML in the topological insulator Bi2Se3[24, 25] [see Fig.1.(b)]. Here, we demonstrate that, the electron-impurity interaction can have a similar consequence without the need of a warped Fermi-surface yielding a non-orthogonally locked configuration of the spin and momentum [Fig.1.(c) and (d)].

The spin-dependent self-energy (SDSE) vector \( \Sigma(K^*) \) represents the impurity average of the microscopic scattering events between the electron and the impurity (see Appendix A). The full spin-neutral and the spin-dependent part of the self-energies can be combined in the pseudospin matrix form as,

\[ \Sigma(K) = \Sigma_0(K) \sigma_0 + \Sigma(K) \sigma \]  

(4)

where \( \Sigma_0 \) is the spin neutral self-energy (SNSE) and \( \Sigma = (\Sigma_x, \Sigma_y, \Sigma_z) \) is the SDSE as introduced before. The total change in the spin between this interacting model and the non-interacting one is expectedly decided by the total change in the spin between this interacting model and the non-interacting one is given by

\[ \Delta S_\lambda(k) = (\lambda/2) [G(K^*) - g_k]. \]  

Here we took the difference of two cases with and without interactions using Eq.(3). In the weak interaction limit[25] this takes an elegant form with the leading term

\[ \Delta S_\lambda(k) \approx \frac{\lambda}{2} \Sigma_0 \hat{k} \]  

(5)

where \( \Sigma_0 = \Sigma(K^*) \hat{k} \) is the component of the SDSE along the momentum. The Eq.(5) states that the interactions can cause both type-I and type-II violations of the OSML. Since there are strong symmetry considerations in the pure crystal structure, the generation of a finite \( \Sigma_0 \) is not trivial. In this work, we study the electron-non-magnetic impurity scattering as a new mechanism for the non-orthogonally locked type-II state as shown in Fig.1 (c) and (d).

### III. THE SPIN-ORBIT IMPURITY SCATTERING

Whatever the mechanism is, the formalism in Eqs. (3-5) hinges upon an accurate model for the self-energy in Eq.(4). In our model we consider the electron in an energy band of the Eq.(1) and the impurity atom is spinless. The electron-impurity scattering is represented as a sum of the spin independent and dependent parts by an effective scattering potential as \( V_{\lambda (j)} = V_{\lambda 0}^{(j)} + V_{\lambda o}^{(j)} \) where the spin-neutral part is \( V_{\lambda 0}^{(j)} \) and the second part \( V_{\lambda o}^{(j)} \) is the spin-orbit scattering. In the notation, the superscript \( j \) refers to the \( j \)’th impurity (see Appendix B). The spin-orbit coupling in the pristine sample is assumed to be sufficiently strong compared to the electron-impurity interaction. This enables a perturbative treatment of the latter within each eigen band \( \lambda \) of the Eq.(1) where the energetically unfavorable inter-band mixing can be neglected in the physical states. The inter-band mixing is however, virtually allowed (see below) but much weaker than the intra-band contribution. By this approximation a simple picture can be obtained where the leading order contribution to NOSML can be isolated from the other secondary effects. The scattering matrix between the initial \( |i\rangle = |k \lambda \rangle \) and the final \( |f\rangle = |k' \lambda' \rangle \) states is \( T_{\lambda \lambda'; k' k} = \sum_j \langle k' \lambda' | V_{\lambda (j)}^{(j)} | k \lambda \rangle \) given in the Born approximation by[2, 43, 45–55] and Appendix B by,

\[ T_{\lambda \lambda'; k' k} = \sum_j \langle k' \lambda' | V_{\lambda (j)}^{(j)} | k \lambda \rangle \]  

(6)

where the exponential phase factor accounts for the impurity scattering phase shifts occurring at random centers \( R_j \) and \( t_{\lambda \lambda'}^{(j)}(k, k') \) is the scattering amplitude of the electron off the \( j \)’th impurity from the initial to the final state which can be derived microscopically once the impurity-electron scattering potential is known. We will assume that there is only one kind of impurity and drop the \( j \) index in \( t_{\lambda \lambda'}^{(j)} \). We then write it as

\[ t_{\lambda \lambda'}(k, k') = a_{\lambda \lambda'} + c_{\lambda \lambda'} \hat{k} \times \hat{k'} \]  

(7)

Here, \( a_{\lambda \lambda'}, c_{\lambda \lambda'} \) are complex coefficients describing the spinless and the spin-orbit scattering strengths respectively[50]. These coefficients also depend on \( k, k' \) as well as the details of the electron-impurity interaction [2, 49–52]. Our philosophy is to understand the NOSML in a minimal approach free from material specific features. We therefore allow that these scattering coefficients depend on phenomenological parameters. The quantum mechanical calculation of the \( a_{\lambda \lambda'} \) and \( c_{\lambda \lambda'} \) starting from the spin dependent matrix elements is done in the Appendix B.

We further assume dilute impurity limit \( n_i \ll \lambda_F^{-3} \) where \( n_i \) is the impurity concentration and \( \lambda_F \) is the
Fermi wavelength of the scattered electrons. In this limit, the interference between multiple scatterings can be neglected.

IV. THE SELF-ENERGY DUE TO SPIN-ORBIT SCATTERING

A. The spin-independent self-energy

The OSML is strictly enforced by the \( C_{\infty} \) symmetry near the \( \Gamma \) point in pure crystals\cite{36, 37}. In strongly spin-orbit coupled materials, deviations from this orthogonal picture requires a sufficient impurity scattering and the renormalization of the Bloch states. Including the impurity scattering perturbatively, the effect vanishes in the first order of the perturbation since at this level the electron self energy averages out to zero over the impurities (see Appendix A). The NOSML emerges beyond the second order in the electron self energy as we show here. The Feynman diagrams of the Green’s functions and the self energies for the \( \lambda \) band are summarized in Fig.(4) of the Appendix A.

The NOSML can be concealed by warping or other anisotropy effects. We therefore confine our work to the case when such phenomena are absent or sufficiently weak and consider an isotropic band \( \xi_{\lambda k} = \hbar^2 k^2/(2m) + \lambda g_0 k \).

When the inter-band processes are neglected, the full self-energy in Eq.(4) can be defined for each spin-orbit band independently as \( \Sigma_{\lambda}(K) \). Here it is important that the impurity averaging is properly done at this level (Appendix A). We find\cite{25, 26, 27} that [as shown in Eq.’s (A15) and (A16)],

\[
\Sigma_{\lambda}(K) = \frac{n_i}{2} \int \frac{dk'}{(2\pi)^2} \times |t_{\lambda}(k, k')|^2 \left[ 1 + \lambda \tilde{G}_{\lambda}(k', E, \sigma) \right] \tilde{G}_{\lambda}(k', E) \tag{8}
\]

The \( \tilde{G}_{\lambda}(k, E) = 1/(E - E_{\lambda k}) \) is the Green’s function of the eigenband with index \( \lambda \) and \( E_{\lambda k} = \xi_{\lambda k} + \lambda |\mathbf{G}_{\lambda}(k, E)| \) as the renormalized energy band with \( \xi_{\lambda k} = \xi_{\lambda k} + \text{Re} \{ \Sigma_{\lambda 0} \} \) and \( \mathbf{G}_{\lambda} = g_{\lambda k} + \Sigma_{\lambda} \). The \( n_i \) dependence in Eq.(8) comes from the averaging over the random impurity positions \( R_i \) as given in Eq.(6) as shown in the Appendix A. We note that, the dependence of the self energy on the impurity concentration in Eq.(8) is not linear due to the non-linear dependence of the renormalized spin-orbit vector \( \mathbf{G}_{\lambda} \) on the self-energy. Furthermore these equations can be obtained from our more general theory of the surface electrons interacting with the lattice excitations studied in Ref.[25] when the phonon excitation energy vanishes in the static limit.

The spin-independent and spin-dependent parts of Eq.(8) are extracted as

\[
\Sigma_{\alpha \lambda} = \text{Tr} \{ \Sigma_{\lambda} \} / 2, \quad \Sigma_{\lambda} = \text{Tr} \{ \Sigma_{\lambda} \sigma \} / 2 \tag{9}
\]

We assume that the spin-orbit scattering strength is negligibly small compared to the spin-independent one.

Since the intra-band scattering is also dominant in respect to the inter-band one, \( |c_{\lambda \alpha}| \ll |a_{\lambda \lambda}| \), the \( a_{\lambda \lambda} \) (which we simply denote by \( a \)) can be directly related to the spin-independent self-energy \( \Sigma_{0\lambda} \). Using the Eq’s.(9) we have,

\[
\text{Im}\{\Sigma_{0\lambda}(E)\} \approx \frac{m}{4\hbar^2 n_i |a|^2} \left( 1 - \frac{g_0}{\sqrt{g_0^2 + \frac{2\hbar^2 m}{\hbar^2} E}} \right). \tag{10}
\]

The Eq.(10) is related to the life-time \( \tau = \hbar / \text{Im}\{\Sigma_{0\lambda}\} \) of the electron momentum due to its scattering with the impurities and note that Eq.(10) is independent of \( \lambda \). Another importance of this equation is the connection with the experiment, i.e. \( \text{Im}\{\Sigma_{0}(E)\} \) can be directly extracted from the experimental quasiparticle momentum distribution\cite{28}. We will use the \( \text{Im}\{\Sigma_{0}\} \) as a phenomenological parameter replacing \( n_i \) dependence throughout.

It will be shown in the next section that the SDSE as shown by the second equation in (9) has a different dependence on \( n_i \). This is brought by the renormalized spin-orbit vector on the right hand side in Eq.(8) which leads to the emergence of a critical boundary separating the OSML and the NOSML phases. This is shown in Section IV.B below.

B. The spin-dependent self-energy and the NOSML

We now turn to the spin-dependent component \( \Sigma_{\lambda} \) in Eq.(8), which can be extracted by using the second of the Eqs.(9). Note that \( g_{\lambda k} \) and \( \Sigma_{\lambda \delta} \) are both absent here due to the rotational symmetry. We start by writing \( \Sigma_{\lambda} = (\Sigma_{\lambda x}, \Sigma_{\lambda y}, 0) \) in the polar form using the radial \( \mathbf{k} \) and the azimuthal \( g_{\lambda k} \) unit vectors as

\[
\Sigma_{\lambda} = \Sigma_{\lambda g} \mathbf{g}_{\lambda} + \Sigma_{\lambda k} \mathbf{k} \tag{11}
\]

where \( \Sigma_{\lambda g} = \lambda \mathbf{g}_{\lambda} \) and \( \Sigma_{\lambda k} = \lambda \mathbf{k} \) are defined as the scalar components of \( \Sigma_{\lambda} \) along the \( \mathbf{g}_{\lambda} \) and \( \mathbf{k} \) directions respectively. Note that these components are even complex functions of \( k \) and independent from the direction of \( \mathbf{k} \). Using these scalar components is particularly useful in the impurity averaging since \( \Sigma_{\lambda g} \) and \( \Sigma_{\lambda k} \) are not affected by the scattering directions of \( \mathbf{k} \) vector, a crucial factor in the impurity averaging considering the random orientations in each scattering event [see Appendix A and the explanation below Eq.(A16)]. The Eq.(11) is equivalently written as

\[
\Sigma_{\lambda x} - i \Sigma_{\lambda y} = e^{-i(\phi + \pi/2)} C_{\lambda k} \tag{12}
\]

which is a quite convenient way of writing the SDSE since \( C_{\lambda k} \) is represented in terms of the scalar components of the SDSE elegantly as \( C_{\lambda k} = \Sigma_{\lambda g} + i \Sigma_{\lambda k} \). Then, the real part renormalizes the spin-orbit strength by \( g_0 \rightarrow g_0 + \Sigma_{\lambda g} / k \). This renormalization can be ignored since \( g_0 k \) is sufficiently strong. The imaginary part
ond order scattering matrix elements

We observe in Eq.(14) that the complex scalar self-energy

where we defined a dimensionless quantity

we now define a microscopic spin-deviation angle

measurements and obtain some quantitative estimates,

The \( \text{Im} T_{\lambda \lambda} \) is directly related to the Fourier transform of the sec-

configuration as demonstrated in Eq.(5). Using Eq.(12) in

\( \lambda k \)

\( \phi' = \phi - \phi^\prime \) describing the scattering angle between \( k \) and \( k' \) and

\[ F_\lambda(k') = \frac{g_\lambda + \Sigma_{\lambda_0}(k')}{|G_\lambda(k', E)|} \frac{1}{E - E_{\lambda k'}} \]

We observe in Eq.(14) that the complex scalar self-energy \( C_{\lambda k} \) is directly related to the Fourier transform of the sec-

order in scattering matrix elements, whereas they can be finite in the second order. The Eq.(16) reveals that the contribution of the inter-band scattering to the NOSML has an opposite sign and energetically less favourable in comparison with the intra-band one. The net effect is therefore an effective energy broadening of the bands combined with fluctuations of the spin around the NOSML configuration.

Finally, the Eqs.(3) and (13) also predict that the anomalous component of the spin along the momentum in the upper and lower Dirac cones are opposite to each other as shown in the inset of Fig.2.

V. DISCUSSION AND CONCLUSION

Before we summarize the main results of this work, we briefly discuss here the role of the in-plane orbitals in the NOSML. An important point here is that under the strong spin-orbit coupling the spin and the angular momentum vectors are not individually conserved but the total angular momentum \( \mathbf{J} = \mathbf{L} + \mathbf{S} \) is. This leads to the formation of the orbital texture in which the orbital configurations can affect the spin texture[39, 40]. The dominant bands are usually \( |p_z\rangle \), \( |p_y\rangle \) orbitals which are also non-negligible, a relevant question here is whether they can influence the orthogonality of the spin and momentum[39, 40, 59]. Including the contribution of these in-plane orbitals, the spin-orbital state is given
up to the linear order in $k$ by,

$$
||k\lambda|| = (u_0 - \lambda \nu_1 k) (|p_2 \rangle \otimes |\lambda_\phi\rangle) \\
- \frac{i}{\sqrt{2}} (\lambda \nu_0 - u_1 k - w_1 k) (|p_1 \rangle \otimes |\lambda_\phi\rangle) \quad (18)
$$

where $u_{0,1}, v_{0,1}, w_1$ are material dependent coefficients, $|\lambda_\phi\rangle = (1/\sqrt{2}) [|\uparrow\rangle - \lambda \epsilon |\downarrow\rangle]$ is the chiral spin-1/2 vortex state, $|\lambda_\phi\rangle = |(-\lambda)\phi\rangle$ and $p_j (p_k)$ are the radial (tangential) in-plane combinations of the $p_x, p_y$ orbitals given by the $|p_j(p_k)\rangle = \cos \phi (-\sin \phi) |p_x\rangle + \sin \phi (\cos \phi) |p_y\rangle$. One may consider that $||k\lambda||$ should have been used in this work instead of $|k\lambda||$. Although this is principally correct, the $||k\lambda||$ does not change the spin or the spin-momentum orthogonality at the single-particle Hamiltonian level[13, 17]. However, the in-plane orbitals with finite angular momentum states can be coupled by the electron-impurity coupling which play a role in the spin-orbit scattering strength in Eq.(7).

Determination of the constants $c_{k,k'}$ and $c_{k,k'}$ in Eq.(7) with their full momentum dependence is a fundamentally important problem. Experimentally, the quasi-particle interference (QPI) with spectroscopic STM can be a promising probe of spin-orbit scattering[53, 55]. With this technique the authors in Ref.[53] estimated $\bar{c}/k_F^2 \approx 80 \text{Å}^2$ for the polar semiconductor BiTeI. Here, our discussion on the correlation between electron-impurity scattering and the spin texture provides an alternative method of extracting $\bar{c}$ when the warping

anisotropy is absent. For a system with inversion symmetry, $\bar{c}$ can be found once the $\delta_k$ of the spin texture could be measured by using S-ARPES. We know that $\delta_{jk} \simeq -20^\circ$ in the case of $\alpha$-Sn[60] and the warping is nearly absent in the surface bands. Using Fig.3 and this $\delta_k$, we find that $\bar{c}/k_F^2 \simeq -40 \text{Å}^2$ putting this material as a strong topological spin-orbit impurity scatterer.

In summary, we showed that the presence of the spin-orbit scatterings from non-magnetic impurities, an effect which is expected to be finite when the impurities are present in strongly spin-orbit coupled realistic materials, can provide a mechanism for the deviations from the experimentally and theoretically well-established phenomenon of OSML to the one with non-orthogonal locking. The NOSML angle which can be measured experimentally, is a non-linear function of the impurity concentration with a critical boundary between the OSML and the NOSML states. It will be interesting to explore this new state experimentally in more general topological/non-topological systems at various impurity concentrations, and in fact, it is important to verify the critical boundary that we theoretically observe. Our theory should pave the road for the complete electron-impurity coupling approach to the spin-momentum locking phenomenon also including the magnetic impurities. We end with a final remark that, our study highlights additional richnesses of spin textures brought by the impurity effects in strongly spin-orbit coupled materials.

**Appendix A: IMPURITY AVERAGING**

Here we discuss details of the impurity averaging of the electron self-energy and the Green’s function. Diagrammatically the electron-impurity interaction is described by the Feynman diagrams as shown in Fig.(4).

We consider that the impurity at the random position $\hat{R}_j$ is scattered by electrons with initial and final momenta $k, k'$. By the impurity averaging we mean a two-step process. The first is that the kinetic phase $\epsilon^{(k-k')} \hat{R}_j$ of the electron wavefunction acquired at the $j$’th scattering is randomized by the random position $\hat{R}_j$ of the $j$’th impurity. This leads to the average over the impurity positions as described in a separate section below. The second crucial factor is that the random impurity positions also lead to randomized incidence direction of the electron between two scattering events. In order to avoid averaging over the random initial-final momentum orientations at each scattering, we must form scalar quantities of the self-energy vector as $\Sigma_k = \Sigma \hat{k}$ and $\Sigma_q = \Sigma \hat{g}_k$ as the component of the self-energy along the momentum and along the spin-orbit vector. The $\Sigma_k$ and $\Sigma_q$ are these scalar quantities which are not affected by the random directions of the initial state vector $k$ before each scattering.

We define the average over the impurity positions by

$$
\langle O \rangle_{\mathrm{imp}} = \int d\hat{R} O(\hat{R}) P(\hat{R}) \quad (A1)
$$
expressed in the pseudo-spin representation. The
expression is (temporarily omitting the
self-energy in Eq.(A2) diagrammatically in order
to derive the dependence of Eq.(8) on the impurity con-
centrations. The first term
\[ \langle \Sigma^{\lambda}_{\text{imp}}(k) \rangle = \frac{1}{\Omega} \int d^{3}R \sum_{j=1}^{N_{\text{imp}}} e^{i(k-k').R_{j}} \text{tr}(T_{k\lambda,k'\lambda}) \text{imp} \]
where the average in Eq.(A3) for which we use:

\[ \langle T_{k\lambda,k'\lambda} \rangle_{\text{imp}} = \frac{1}{\Omega} \int d^{3}R \sum_{j=1}^{N_{\text{imp}}} e^{i(k-k').R_{j}} \text{tr}(T_{k\lambda,k'\lambda}) \text{imp} \]

with the \( j \)-index in the summation describing the \( j \)th
impurity. The Feynman diagrams corresponding to the
\( T_{k\lambda,k'\lambda} \) are shown in Fig.(4a). In Eq.(A2) the \( \Sigma_{\lambda}(k',E) \)
is the interacting electron Green’s function of the \( \lambda' \)
band in terms of the 2 x 2 matrix form in the electron-
pseudospin space. In order to find this quantity, we first
start with the Dyson equation

\[ \frac{1}{\Sigma_{\lambda}(k,E)} = \frac{1}{\Sigma_{\lambda_{0}}(k,E)} - \Sigma(k,E) \]

with

\[ \Sigma_{\lambda_{0}}(k,E) = \frac{1}{E - \xi_{k} - g_{k,\sigma}} \]

representing the non-interacting Green’s function and the
\( \Sigma(k,E) = \Sigma_{\lambda_{0}}(k,E)\sigma_{0} + \Sigma_{\lambda}(k,E)\sigma \)
the full electron self-energy. The \( \Sigma_{\lambda}(k,E) \) in the
Eq.(A4) can then be compactly written as

\[ \Sigma_{\lambda}(k,E) = \sum_{\lambda} \Sigma_{\lambda}(k,E) \]

where

\[ \Sigma_{\lambda}(k,E) = \frac{1}{E - E_{\lambda\lambda}} \]

as the exact Green’s function of the quasiparticles in the
eigenband \( \lambda \) of the Hamiltonian in Eq.(1). The Eq.(A7)
itself is the Green’s function of the electrons in the eigen-
band \( \lambda \) expressed in the pseudo-spin representation. The
\( G_{\lambda}(k,E) = g_{k} + \Sigma_{\lambda}(k,E) \) is the renormalized spin-orbit
vertex and \( G_{\lambda} \) is the unit vector of \( G_{\lambda} \). Eq.(A7) is the
direct sum of the contributions from each spin-orbit band
singled out by the physical pole-position of the \( G_{\lambda}(k,E) \)
at \( E = E_{\lambda\lambda} \).

We now leave the Green’s functions aside and examine the
full self-energy in Eq.(A2) diagrammatically in order
to derive the dependence of Eq.(8) on the impurity con-
centration. The first term \( \langle T_{k\lambda,k'\lambda} \rangle_{\text{imp}} \) is the impurity
average in Eq.(A3) for which we use:

\[ \sum_{j=1}^{N_{\text{imp}}} \langle e^{i(k-k').R_{j}} \rangle_{\text{imp}} = \frac{1}{\Omega} \int d^{3}R \sum_{j=1}^{N_{\text{imp}}} e^{i(k-k').R_{j}} \]

where the average impurity concentration is given by

\[ n_{i} = N_{\text{imp}}/\Omega. \]

We therefore have that \( \langle T_{k\lambda,k'\lambda} \rangle_{\text{imp}} =
\]

\[ n_{i} \delta_{k,k'} \]

The main effect of this term is to shift the Fermi energy by a constant and not contribute to the
energy-spin renormalization. We now shift our attention
to the second term in Eq.(A2) describing the intra-band
scattering \( \lambda' = \lambda \) and ignore the inter-band ones, i.e.
\( \langle T_{\lambda\lambda} \Sigma^{\lambda}_{\text{imp}} \rangle_{\text{imp}} \). This requires the knowledge of the full
Green’s function. The result is (temporarily omitting
some indices for simplicity),

\[ \langle T_{\lambda\lambda} \Sigma^{\lambda}_{\text{imp}} \rangle_{\text{imp}} \approx \frac{1}{\Omega} \sum_{i,j} \langle e^{i(k-k').(R_{i}-R_{j})} \rangle_{\text{imp}} \times |t_{\lambda\lambda}(k,k')|^{2} \]

where we used the unitarity of the scattering matrix as
\( t_{\lambda\lambda}(k,k') = t_{\lambda\lambda}(k',k) \). We now work on the relevant
part in Eq. (A10) which depends on the impurity average. By definition
\[ \sum_{i,j} \langle e^{i(k-k').(R_i-R_j)} \rangle_{\text{imp}} = \frac{1}{\Omega} \sum_{i=j} 1 + \sum_{i \neq j} \langle e^{i(k-k').(R_i-R_j)} \rangle_{\text{imp}} \]
(A11)

The impurity averaging over a totally random impurity distribution yields random interference between different impurities when \( R_i \neq R_j \), yielding a vanishing contribution for \( k' \neq k \). This term is therefore (with \( 1 \ll N_{\text{imp}} \))
\[ \sum_{i \neq j} \langle e^{i(k-k').(R_i-R_j)} \rangle_{\text{imp}} = n_{\text{imp}}^2 \delta_{k,k'} \]
(A12)

Hence it averages out to zero when \( k \neq k' \) like the first order impurity average in Eq. (A9). The net effect of this term is therefore essentially the same as the first order impurity-vertex. The net effect of the impurity averaging in Eq. (A11) is therefore provided by the first term on the right hand side as \( (1/\Omega) \sum_i 1 = N_i / \Omega = n_i \). Eq. (A10) is therefore given by
\[ \langle T \Sigma \rangle_{\text{imp}} = n_i |t_{\lambda\lambda}(k,k')|^2 \Sigma_{\lambda}(k',E) \]
(A13)

Using this result in Eq. (A2) we find
\[ \langle \Sigma_{\lambda}(K) \rangle_{\text{imp}} = n_i \int \frac{d\kappa'}{2\pi} |t_{\lambda\lambda}(k,k')|^2 \Sigma_{\lambda}(k',E) \]
(A14)

The Eq. (A14) is the electron self energy for the \( \lambda \) band including the intra-band scattering. Using Eq. (9), the Eq. (A14) yields the Eq. (8) in the manuscript where we simply dropped the explicit impurity averaging symbol \( \langle \ldots \rangle_{\text{imp}} \). The SNSE and the SDSE as used in the main text are
\[ \Sigma_{0\lambda} = n_i \int \frac{d\kappa'}{2\pi} |t_{\lambda\lambda}(k,k')|^2 \Sigma_{\lambda}(k',E) \]
(A15)
\[ \Sigma_{\lambda} = \lambda n_i \int \frac{d\kappa'}{2\pi} |t_{\lambda\lambda}(k,k')|^2 \Sigma_{\lambda}(k',E) \]
(A16)

Next we consider the second type of average which is due to the random orientations of the initial/final momenta. The \( \Sigma_{\lambda\kappa} = \langle \Sigma_{\lambda\kappa} \rangle_{\text{imp}} \) and \( \Sigma_{\lambda\gamma} = \langle \Sigma_{\lambda\gamma} \rangle_{\text{imp}} \) are meaningful quantities for impurity averaging since both are scalars and unaffected by the random directions of the scattered electron momenta. It can be explicitly seen that, the transformation in Eq. (12) separates the random orientation of the \( \hat{k} \) and indeed, the \( C_{\lambda\kappa} = \Sigma_{\lambda\kappa} + i \Sigma_{\lambda\kappa} \) perfectly becomes a complex scalar. After a similar calculation to the Eqs. (A16) above, we easily calculate the \( C_{\lambda\kappa} \) as shown in Eq. (13) of the manuscript.

In this perspective, the Eq.’s (A15) and (A16) are used to derive the Eq.’s (10) and Eq. (13) of the manuscript.
with
\[
\tilde{v}_0(k, k') = \int dr \, e^{i(k-k' \cdot r)} u_{k'}^*(r) v_0(r) u_{k}(r)
\]
(B7)
\[
\tilde{v}_{so}(k, k') = \int dr \, e^{i(k-k' \cdot r)} u_{k'}^*(r) v_{so}(r) u_{k}(r)
\]

Eqs. (B4)-(B10) comprise a generic derivation of Eq. (6) in the main text. This general formulation can be applied to specific cases only when the Bloch state in Eq. (B3) and the electron-impurity potential \( v_0(r) \) are known. However, we can still say more about the \( t_{\lambda \lambda'}(k, k') \) using the derivation of the quantum mechanical spin-orbit scattering in [50].

Here it must be stressed that, in the vicinity of the \( \Gamma \) point, the exact shape of the Bloch wavefunctions in Eq. (B3) is not strictly required. With the Hamiltonian in Eq. (1) representing our system there, we consider the eigenfunctions as
\[
|k\lambda\rangle = |U_k\rangle_{\lambda \sigma} |k\sigma\rangle
\]
(B8)
where \( |k\sigma\rangle \) are the eigenbasis without the spin-orbit interaction [61] and
\[
U_k = \frac{1}{\sqrt{2}} \left( \begin{array}{cc} 1 & i e^{-i\phi} \\ i e^{i\phi} & 1 \end{array} \right).
\]
(B9)
First, we write the Eq. (B6) using the fundamental formulation of the electron-spinless impurity scattering as [50]
\[
t_{\lambda \lambda'}(k, k') = a_{\lambda \lambda'}(k, k') + c_{\lambda \lambda'}(k, k') \hat{k} \times \hat{k}' \hat{z} \tag{B10}
\]
which is the Eq. (7). The Eq. (B10) is obtained with the assumption that the state of the impurity is not changed by the scattering process which is in the essence of the Bohr approximation. Considering that \( k \) and \( k' \) are confined to the \( xy \) plane in our case, the coefficients in Eq. (B10) are given by,
\[
a_{\lambda \lambda'}(k, k') = A_0 \langle k' \lambda' | k \lambda \rangle
\]
\[
c_{\lambda \lambda'}(k, k') = C_0 \langle k' \lambda' | \sigma_z | k \lambda \rangle
\]
(B11)
Here, \( A_0 \) and \( C_0 \) are known complex functions of the scattering angle \( \Delta = \phi - \phi' \) in the scattering plane in terms of the Legendre functions \( P_\ell^m(\Delta) \) and associated Legendre functions \( P_\ell^m(\Delta) \) and the partial scattering phase shifts as given by [50]
\[
A_0 = (a_0 + b_0 \cos \Delta)
\]
\[
C_0 = c_0 \sin \Delta.
\]
Here we considered only the total angular momentum states \( \ell = 0, 1 \) as the relevant ones in this work. In general, \( a_0, b_0 \) and \( c_0 \) are complex coefficients depending on the partial phase shifts. Further information about these parameters can be obtained by calculating the scattering phase shifts for which the knowledge of the specific scattering potentials Eq. (B2) are required. For the sake of obtaining a general argument here, we consider \( a_0, b_0 \) and \( c_0 \) as complex phenomenological constants. Using Eq. (B11), we find
\[
a_{\lambda \lambda'}(k, k') = A_0 |U_k^\dagger U_k\rangle_{\lambda \lambda'}
\]
\[
c_{\lambda \lambda'}(k, k') = C_0 |U_k^\dagger \sigma_z U_k\rangle_{\lambda \lambda'}
\]
(B13)
The Eq.’s (7), (B12) and (B13) are then used in (B10) to find the result in Eq. (14).

[1] R. Winkler, Spin-orbit Coupling Effects in Two-Dimensional Electron and Hole Systems, Springer Tracts in Modern Physics (Springer Berlin Heidelberg, 2003).
[2] M. Dyakonov, Spin Physics in Semiconductors, Springer Series in Solid-State Sciences (Springer International Publishing, 2017).
[3] M. Z. Hasan and J. E. Moore, Annual Review of Condensed Matter Physics 2, 55 (2011), https://doi.org/10.1146/annurev-conmatphys-062910-140432.
[4] A. Bansil, H. Lin, and T. Das, Rev. Mod. Phys. 88, 021004 (2016).
[5] M. Franz and L. Molenkamp, Topological Insulators, ISSN (Elsevier Science, 2013).
[6] F. Ortmann, S. Roche, S. Valenzuela, and L. Molenkamp, Topological Insulators: Fundamentals and Perspectives (Wiley, 2015).
[7] S. Hikami, A. I. Larkin, and Y. Nagaoa, Progress of Theoretical Physics 63, 707 (1980), https://academic.oup.com/ptp/article-pdf/63/2/707/5336056/63-2-707.pdf.
[8] D. Awschalom, D. Loss, and N. Samarth, Semiconductor Spintronics and Quantum Computation, NanoScience and Technology (Springer Berlin Heidelberg, 2013).
[9] O. V. Yazyev, J. E. Moore, and S. G. Louie, Phys. Rev. Lett. 105, 266806 (2010).
[10] A. A. Burkov and D. G. Hawthorn, Phys. Rev. Lett. 105, 066802 (2010).
[11] D. Culcer, E. H. Hwang, T. D. Stanescu, and S. Das Sarma, Phys. Rev. B 82, 155457 (2010).
[12] S. LaShell, B. A. McDougall, and E. Jensen, Phys. Rev. Lett. 77, 3419 (1996).
[13] M. Z. Hasan and C. L. Kane, Rev. Mod. Phys. 82, 3045 (2010).
[14] Z. Jiang, C.-Z. Chang, M. R. Masir, C. Tang, Y. Xu, J. S. Moodera, A. H. MacDonald, and J. Shi, Nature Communications 7, 11458 (2016).
[15] P. Höflner, J. Schäfer, A. Flieszar, J. H. Dil, B. Slomski, F. Meier, C. Loho, C. Blumenstein, L. Patthey, W. Hanke, and R. Claessen, Phys. Rev. Lett. 108,
