Symmetry Conserving Maximally Projected Wannier Functions

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To obtain a local description from highly accurate density functional theory codes that are based on modified plane wave bases, a transformation to a local orthonormal Wannier function basis is required. In order to do so while enforcing the constraints of the space group symmetry the Symmetry Conserving Maximally Projected Wannier Functions (SCMPWF) approach has been implemented in the Full-Potential-Local-Orbital code, FPLO. SCMPWFs represent the zeroth order approximation to maximally localized Wannier functions, projecting a subset of wave functions onto a set of suitably chosen local trial-functions with subsequent orthonormalization. The particular nature of the local orbitals in FPLO make them an ideal set of projectors, since they are constructed to be a chemical basis. While in many cases projection onto the FPLO basis orbitals is sufficient, the option is there to choose particular local linear combinations as projectors, in order to treat cases of bond centered Wannier functions. This choice turns out to lead to highly localized Wannier functions, which obey the space group symmetry of the crystal by construction. Furthermore we discuss the interplay of the Berry connection and position operator and especially its possible approximation, symmetries and the optimal choice of Bloch sum phase gauge in cases where the basis is not explicitly known. We also introduce various features which are accessible via the FPLO implementation of SCMPWFs, discuss and compare performance and provide example applications.

CONTENTS

I. Introduction 1
A. Wannier functions and FPLO 1
B. Outline 2

II. Formalism and Method 2
A. Wannier Orbitals in FPLO 2
B. Accuracy and localization 5
C. Practical application of the method 6

III. Topological aspects 7
A. Position operator 7
B. Berry connection 8
C. Berry curvature 9

IV. Features of Maximally Projected Wannier Functions 10
A. Work flow 10
B. Model extraction 11
C. Band disentangling 12
D. Berry curvature, Anomalous Hall Conductivity 13
E. Molecular orbital projectors 14
F. Molecule Wannier functions 15
G. Post processing 16

V. Performance and Comparison 17
A. Numerical performance 17
B. Comparison with Wannier90 17
C. Comparison with previous reports 18
D. Berry curvature dipole 19

VI. Summary and Conclusions 23

VII. Acknowledgments 23
A. Local basis states 23
1. FPLO basis 23
2. Bloch sums 24

B. Symmetry 24
1. Local basis symmetry 24
2. Space group symmetry/Time reversal 25
3. Bloch theorem 27
4. Berry connection/curvature 28
5. Bloch sum gauge invariance 29

C. Crystal structures 30
1. CaCuO2 30
2. bcc Fe 30
3. B2 FeAl 30
4. HgS 30
5. MgB2 30
6. H2O 31
7. Related citations 31

References 31

I. INTRODUCTION

A. Wannier functions and FPLO

Natural basis states for the treatment of extended systems are extended states which carry a pseudo-momentum quantum number. On the other hand, chemistry is more intuitively described by local basis states, which resemble atomic orbitals, while extended model
Hamiltonians are often built from (implicit) atomic entities (tight-binding). Although several numerical methods exist, which treat molecules and solids with an atom like basis, many contemporary highly accurate density functional theory (DFT) codes are based on modified plane wave bases. To gain back the local description a transformation to a local orthonormal Wannier function basis is desirable.

The usefulness of Wannier functions has gained new momentum in the context of topological properties of extended systems, for whose determination a downfolding of the whole system’s Hamiltonian onto a smaller model is very helpful. While smaller models reduce the often heavy burden of calculating topological quantities, numerical accuracy, especially symmetry conservation, becomes an important issue as well, in particular for higher order transport properties, which involve taking derivatives (e.g. Berry curvature dipole) or for tensorial properties.

Wannier functions (WFs) can be defined in many ways due to the gauge freedom in choosing a unitary transformation among the wave functions to be Wannierized. One way of fixing the gauge, now widely used, is the requirement of maximum localization[1–3] (ignoring the constraints of the space group symmetry). This is an rather intricate algorithm which becomes even more complicated, if space group symmetry is added[4, 5].

Extending the ideas of Ref. 6 to retain full symmetry information we implemented a different scheme into the Full-Potential-Local-Orbital code, FPLO[7], as outlined in Ref. 8. This scheme, which we call Symmetry Conserving Maximally Projected Wannier Functions (SCM-PWF), basically represents the zeroth order approximation of the maximally localized approach[1], in projecting a subset of wave functions onto a set of suitably chosen local trial-functions with subsequent orthonormalization.

The particular nature of the local orbitals in FPLO make them an ideal set of projectors, since they are constructed to be a “chemical” basis. In many cases projection onto the FPLO basis orbitals is sufficient, while the option is there to choose particular local linear combinations (molecular orbitals [MO]) as projectors, in order to treat cases of bond centered Wannier functions. It turns out that our choice leads to highly localized Wannier functions, which obey the space group symmetry of the crystal by construction. In cases where the resulting WFs are not well-localized it is due to a bad choice and can be fixed.

Many similar projective schemes have been proposed before, often including localization criteria (Refs. 9 and 10 and Ref. 11 and references therein) as well as including symmetry considerations[12]. The main difference to our method is that ours is straightforward and simple as the approach of Ref. 6. On top it conserves symmetry and is generally applicable to entangled bands with very few adjustable parameters.

The main challenge of our approach is that the user has to decide where the WFs shall sit and which local symme-

try they shall have. However, this is often exactly what one wants to do, when constructing a Wannier model for a given subset of bands. The strong connection of the FPLO basis and the resulting WFs and the comparatively small basis size allows for automatic Wannierization, which is nothing but constructing the Löwdin orthogonalized FPLO basis. In principle, everything which can be expressed in WFs could be transformed back into the non-orthogonal FPLO basis, which opens the possibility to circumvent WFs altogether, although this is less efficient.

The method described here is used in conjunction with the FPLO density functional (DFT) code, but it can also be used whenever a Hamiltonian is given in a local basis (tight-binding [TB] models), even if the basis is not explicitly known. This can for instance be helpful to further reduce the dimensionality of a TB-model.

The structure of a local basis code also requires modifications to the way the Berry connection/curvature need to be calculated. The connection of these quantities to the position operator was discussed in great detail in Ref. [13] before the recognition of their fundamental nature for topological properties of matter. The local basis formulation of the Berry operators presented here offers insights into the correct way of neglecting the position operator matrix elements and into the correct phase choice of Bloch sums, especially when the Wannier basis is not explicitly known, in particular for tight-binding models.

\section{II. FORMALISM AND METHOD}

\subsection{A. Wannier Orbitals in FPLO}

The FPLO basis consists of local atom like orbitals \( \Phi_{\nu ls}(r) \) localized at site \( s \) in unit cell \( R \), having atom like quantum numbers (qns) \( \nu = nlm\sigma \) without spin orbit coupling and \( \nu = nlj\mu \) in 4-component full-relativistic mode, where \( n \) is the main quantum number, \( lm \) are the qns. of real spherical harmonics, \( \sigma \) is the spin index and
$lj\mu$ are the qns. of a standard (complex) spherical spinor (see Sec. A 1).

From these orbitals Bloch sums can be formed

$$\Phi_{av}^k(r) = \frac{1}{\sqrt{N}} \sum_R e^{ik(R+\lambda s)} \Phi_{Rs\nu}^k(r). \quad (1)$$

where $N$ is the number of unit cells in the Born-von-Kármán (BvK) torus. The chosen normalization is further discussed in Sec. A 2. The parameter $\lambda$ allows to choose the phase gauge and is of interest later. The default gauge in FPLO and hence in constructing the Wannier functions is the relative gauge ($\lambda = 1$) in which matrix elements of an operator between two Bloch sums only depend on the relative vector connecting the two orbital locations in each term of the sum. In post-processing modules which use the WFs the periodic gauge ($\lambda = 0$) can be chosen, in which the Bloch sums are periodic in $k$-space.

The FPLO orbitals from different sites are in general non-orthogonal, which leads to a non-trivial overlap matrix $S^k$. This avoids complicated orbital shapes away from the atomic core region and in fact the SCMPWF transformation is essentially nothing but the orthogonalization of (a subset of) our local orbitals (LO). Introducing the row vector $\Phi^k$ of all orbitals and the matrix of wave function coefficients $C^k$ the overlap and Hamiltonian matrices read

$$S^k = \langle \Phi^k | \Phi^k \rangle$$
$$H^k = \langle \Phi^k | \hat{H} | \Phi^k \rangle$$

and the eigenvalue problem becomes

$$H^k C^k = S^k C^k \epsilon^k, \quad C^k S^k C^k = 1 \quad (3)$$

which gives the row vector of full wave functions

$$\psi^k = \Phi^k C^k. \quad (4)$$

as a linear combination of the Bloch sums of LOs. In the following we will often suppress the orbital/WF indices (and sometimes site indices). The corresponding expressions must then be understood as sub-blocks of vectors-of-functions or matrices.

In order to obtain Wannier functions one needs to Fourier back transform the extended wave functions $\psi^k$ including some unitary matrix $U^k$ which constitutes a general gauge choice

$$\Phi_{c\alpha} = \sum_{Rs} \Phi_{Rs\nu} U_{Rs\nu,ci} \quad (7)$$

sitting at an imagined Wannier center $c$, having an index $i$ from LOs $\Phi_{Rs\nu}$ in the vicinity of $c$. In practice the input $U_{Rs\nu,ci}$ consists of a few lines specifying the center $c$, the contributing site numbers, difference vectors from the center to these sites and weights. In the simplest case $U_{Rs\nu,ci} = \phi_{R+\nu,s,c}\delta_{\nu,0}$ for a subset of orbitals. $U$ can also contain a transformation onto different local quantization axes in which the LOs are rotated, which helps to yield WFs adapted to certain local symmetries.

At this step we insist that the set of MOs transforms properly under the spacegroup. A space group operation with point group matrix $\alpha$ and translation $\tau$ is given in Seitz notation by $g = \{\alpha | \tau\}$. The atom positions transform as

$$g(R+s) = \alpha(R+s) + \tau = \alpha R + gs \quad (8)$$

The transformed site can be backfolded onto the original set of sites via

$$gs = s_g + R_{g,s} \quad (9)$$

where $s_g$ is the site in the original set of sites which $s$ gets mapped onto by $g$ and $R_{g,s}$ is a lattice vector. This back-folding is needed since Bloch sums of the same site in two
different unit cells can differ in phase and since sites are indices to the Bloch sums. Hence, consistency requires that we always consider the functions in the original set of sites. Of course also the Wannier centers $c$ transform this way. Note, that in our formalism the Wannier centers are not an output of the calculation but an input. The most general transformation property of local orbitals or MOs is given by (see Sec. B 2)
\[ \{ \alpha | \tau \} \phi_{ci} = \sum_{c'} \phi_{\alpha R+R_{g,c},c'c'} D_{c,c'}(g) \]
where $D_{c,c'}$ is a matrix which mixes the MOs at center $c$ such that MOs at the transformed center $c'$ are obtained. In essence, if a MO at some center is picked, all MOs which are symmetry related at the same center as well as all equivalent MOs at symmetry related centers must be included into the set of projectors. The sets at related centers can be arbitrarily unitary mixed. Knowing the transformation properties of the FPLO orbitals and the MO matrix $U$ in Eq. (7), $D$ in Eq. (10) is fully defined. By constructing the MOs this way we have explicit information about the symmetry properties of the resulting Wannier functions. In most cases the MOs are identical to a subset of our LOs.

Using Eq. A8 in Sec. A 2, we can calculate the projection of the Hilbert subspace Eq. 6 onto a MO as
\[ \mathcal{H} | \phi_{Re} \rangle = \sum \langle \Psi^k | h^k \rangle \langle \Psi^k | \phi_{Re} \rangle \]
\[ = \sum \left( \langle \Psi^k | h^k C^k S^k U^k \frac{1}{\sqrt{N}} \right) e^{-i k (R+R_\lambda c)} \]
\[ = \frac{1}{\sqrt{N}} \sum \tilde{w}_{ci} e^{-i k (R+R_\lambda c)} \]
which defines the Bloch sums
\[ \tilde{w}_{ci} = \left( \langle \Psi^k | h^k C^k S^k U^k \right)_{ci} \]
of the raw (not orthonormal) Wannier functions as well as the phase gauge to be used in the Fourier back transformation into real space.

This simple projection completely defines $U^k$ in Eq. (5) (besides orthonormality) and is quite intuitive: define the desired subset of bands by specifying $h^k$ and project out the part which has overlap with suitably selected MOs (trial functions). If these MOs span the most part of this Hilbert space the result must be the Wannier functions describing $\mathcal{H}$. The raw WF contains the weights $C^k S^k U^k$ which can be interpreted as the “square root” of the band weights of the MOs in $\Psi^k$: a form of band characters (fat bands) of the considered orbitals, only with the phase information retained. So, we basically sum wave functions according to MO-weights contained in them.

We apply the Löwdin procedure to Eq. (12) in order to obtain orthonormal WFs, which are guaranteed to have the smallest least square deviation from the raw functions: we divide by the square root of the overlap matrix of the raw WFs
\[ O^k = \langle \hat{w}^k | \hat{w}^k \rangle = U^k + S^k C^k (h^k)^2 C^k S^k U^k \]
and obtain orthonormal Bloch sums of maximally projected WFs
\[ w_{ci}^k = \left( \hat{w}^k - \frac{1}{\sqrt{O^k}} \right)_{ci}. \]
The inverse square root of the overlap is calculated using the eigen decomposition $O^k Z^k = Z^k O^k$ according to $(O^k)^{-1/2} = \frac{1}{\sqrt{O^k}} Z^k Z^k$. Furthermore, since we know the symmetry transformation $U^k = U^{k_\lambda} D$, we also know the transformation property $O^k = D^+ O^k D$, which gives $Z^k = D^+ Z^{k_\lambda}$ and $(O^k)^{-\frac{1}{2}} = D^+ (O^{k_\lambda})^{-\frac{1}{2}} D$. So, the WFs $w_{ci}^k$ transforms as the MOs themselves. Finally, we Fourier back transform into real space by inserting Eq. (14) into Eq. (11)
\[ w_{Re}^k = \frac{1}{\sqrt{N}} \sum_k w_{ci}^k e^{-i k (R+R_\lambda c)} \]

In summary our gauge fixing matrix when transforming wave functions into Wannier functions is obtained by basically picking linear combinations of wave functions of a Hilbert subspace which have maximal projection (band weight) for a number of MOs/LOs. The result then is orthogonalized and Fourier back transformed into real space to obtain the Wannier functions, which by construction have maximum resemblance to the MOs. Since the projectors are identical to or are constructed out of our original LOs, which form the basis of the wave functions to begin with, the projection result is optimal as compared to the original idea of Ref. [6] of projecting onto hand-made atom like trial functions, which are not a basis of the wave function formation.

In the extreme case, where we calculate a Wannier basis as large as the FPLO basis (automatic mode), we can use $\phi = \Phi$ for all orbitals and $h^k = 1$ (all bands) which gives $U = 1$ and with $C^k C^k = (S^k)^{-1}$:
\[ \tilde{w}^k = \Phi^k \]
which results in the orthonormalized WFs
\[ w^k = \Phi^k \frac{1}{\sqrt{\langle \Phi^k | \Phi^k \rangle}} \]
which is nothing but the Löwdin orthonormalized FPLO basis. From this it is also clear why we need to use the same $\lambda$ in the Fourier back transformation as in the definition of the orbital Bloch sums Eq. (1). If $\Phi^k$ were orthonormal the Fourier back transformation of Eq. (1) would just yield the (assumed to be orthonormal) local orbitals, i.e. Wannier functions.

Eqs. (12,14) can be written in matrix form as
\[ \tilde{w}^k = \Psi^k \Upsilon^k = \Phi^k \Delta^k \]
which emphasizes either the connection to the wave functions or the basis. While
\[
U^k = \frac{1}{\sqrt{\Omega^k}} \sum_k \mathcal{S}^k U^k
\]  
(19)

is projective unitary $U^k + U^k = 1$,
\[
a^k = C^k U^k
\]  
(20)

fulfills $a^k + \mathcal{S}^k a^k = 1$ since the basis has nontrivial overlap. The Hamiltonian in the Wannier basis then becomes
\[
H^{(w)}(k) = \left\langle w^k | \hat{H} | w^k \right\rangle
\]
\[
= U^k + \varepsilon^k U^k
\]
\[
= a^k + \mathcal{H}^k a^k
\]  
(21)

with $H^k$ according to Eq. (2). A general operator $\hat{B}$ with matrix elements between LO Bloch sums $B^k$ then reads in Wannier basis $B^{(w)k} = \left\langle w^k | \hat{B} | w^k \right\rangle = a^k + \mathcal{B}^k a^k$.

B. Accuracy and localization

If the Wannier transformation is well set up the eigenvalues of Eq. (21) will reproduce the band structure of the targeted subset of bands. For post processing the wannierized operators need to be stored in their real space representation which is obtained from
\[
B^{(TB)}_{0e,k,Rc} = \sum_k f_k e^{-ik(R + \lambda(c' - c))} B^{(w)k}_{c',e,c}
\]  
(22)

where in the simplest case $f_k = \frac{1}{N}$. The superscript stands for tight-binding (TB). From the tight binding representation the WF Bloch representation is recovered via
\[
B^{(BL)}_{c',e} = \sum_R e^{-ik(R + \lambda(c' - c))} B^{(TB)}_{0e,c,Rc}
\]  
(23)

Besides the TB-representation of the operators also the basis representation of the WFs themselves is useful. Inserting Eqs. (1,18) into Eq. (15) one gets
\[
w_{Re} = \sum_{R'} \Phi_R s \phi_{R-s,Re}
\]  
(24)

with the coefficients
\[
\tilde{a}_{0e,R-R',c} = a_{R's,Re} = \frac{1}{N} \sum_k e^{-i(k(R+\lambda c-R'-\lambda s))} a^k
\]  
(25)

These coefficients are a direct measure of localization.

At this step we have two possibilities. Either a real space cutoff $\rho$ is defined such that all matrix elements from a WF center $c'$ to another center at $R + e$ are discarded if $|R + \rho - c'| > \rho$, or one chooses the maximum possible set of center-pairs $c',Rc$ which are consistent with the $k$-mesh used in the Fourier back transformation, which is the mesh used in the self consistent DFT calculation (new since FPLO version 19.00). A cutoff is useful to reduce storage space, to speed up the calculation or to aim at a WF model with minimal number of parameters(not as accurate, of course).

Operators recovered via Eq. (23) at a post processing stage are in general different from their exact Wannier transform $B^{(w)k}_{(w)k}$ for a $k$-point not included in the $k$-mesh. This is the very essence of Wannier interpolation. Hence, the resulting band structure after this transformation through the real space representation will also differ from the band structure obtained directly from the exact transform Eq. (21). The size of this error determines the quality of the Wannier fit.

If the error is unacceptable there are two possible reasons: either the cutoff removed too much information and needs to be increased or the maximum possible cutoff is not big enough, which is equivalent to saying that the underlying $k$-mesh is not fine enough. In practice a cutoff between 25-40 Bohr radii is sufficient to reduce this error to a satisfactory degree. An exception is the automatic mode in which all FPLO orbitals become Wannier functions. Since, the higher lying states are spanned by polarization orbitals which have a larger extent than the valence orbitals the corresponding WFs are also more extended.

To further elucidate this issue we discuss the Fourier back transformation in more detail. If symmetry was of no concern the acceptable $R$-mesh in Eqs. (22,23) is determined entirely by the $k$-mesh[14]. The $k$-mesh is defined as a regular grid defined via $N_k$ subdivisions of the three primitive reciprocal lattice vectors. The $R$-mesh then is the dual mesh, i.e. the smallest parallelepiped supercell in real space for which exp $(i(kR)) \neq 1$. This supercell is the reciprocal cell of the smallest grid micro cell. This mesh is not optimal, instead one folds the vectors of this mesh back to form a set of vectors surrounding the origin and having smallest possible length. Localization of the Wannier functions ensures that the vectors of larger length become more and more unimportant.

If $R$-vectors were included which lie outside of the supercell the Wannier functions will acquire replica features: the contributions of the local orbitals to a Wannier function as measured by $a_{0e,Rc}$ first decrease exponentially with increasing distance of the orbital to the Wannier center until distances are reached which are comparable to multiples of the real space length corresponding to the inverse of the smallest $k$-mesh distance. At these points the orbital contributions start increasing again, i.e. the Wannier functions are quasi-periodic objects in real space with periods defined by the inverse of...
the smallest $k$-mesh distances.

Since we construct symmetry conserving Wannier functions, our $R$-mesh is ideally chosen to reflect the symmetry. This is achieved by backfolding the supercell vectors into the smallest possible spherical volume around the origin for each pair of WF centers $c', Rc$. Then for all vectors of this set all vectors additionally obtained by symmetry are added to the set. Finally, weights are attached to all vectors such that the weights for symmetry related vectors are identical and that the sum of the weights of all vectors which are identical by supercell translations is one. This way we include vectors which violate the supercell condition, but replica do not occur due to the weighting. This algorithm determines the maximal number of real space vectors if no cutoff is used.

In order to reduce computation time the $k$-summation is done for the irreducible part of the mesh, which changes the effective weights to $f_k = \frac{m}{N}$ by multiplying with the multiplicity of the $k$-point. This also requires an explicit symmetrization of the resulting matrix elements Eq. (22). This can easily be done, since the transformation properties of the WFs are explicitly known.

The symmetry which we consider is the full space group or in spin polarized full relativistic mode the group formed by operations which do not invert the magnetic field and by products of time reversal with the operations which invert the field. In non spin polarized full relativistic mode time reversal is added as an extra symmetry.

Without spin orbit coupling the FPLO orbitals are real, which means that WFs which are obtained by projecting onto these orbitals are also real, if inversion symmetry is present. In relativistic mode the angular parts of the orbitals are spherical spinors, which are inherently complex, and so are the Wannier functions.

### C. Practical application of the method

For application of this method the input needs to be discussed. We outlined above that, unless automatic mode is used, a Hilbert subspace of the full band structure needs to be chosen first. This is usually the bands around the Fermi level. Once this decision is made it has to be determined which orbitals contribute to these bands, which can be achieved by inspecting the orbital character of the band structure. At this point one has to make sure that at each point in $k$-space the number of bands highlighted by the orbital character is not smaller than the dimension of the desired Wannier basis. If this condition is not fulfilled is is an indication that other orbitals contribute essentially to the targeted bands and most importantly it means that at these $k$-points the raw WF overlap Eq. (13) will be singular. In cases where bond centered WFs are expected, simple molecular orbitals can be constructed from the relevant orbitals, otherwise the relevant orbitals are the projectors themselves.

In full relativistic mode the option exists to project onto spherical spinors with $\lambda\mu$ quantum numbers (Sec. B1, Eq. (A1)) or onto orbitals which are transformed into pseudo non-relativistic symmetry with $lm\sigma$ quas (Sec. B1, Eq. (B7)). The latter are still four spinors but their large components are mostly resembling real spherical harmonics and they transform as real spherical harmonics. Additionally, local quantization axes can be chosen separately for the orbital angular momentum part as well as the spin part, which facilitates the construction of specialized models and band structure analysis.

The automatic mode can be used in two ways: either all basis orbitals are Wannierized or a reduced set is Wannierized, where all deep lying orbitals (semi-core and deep lying valence orbitals), which do not contribute to the essential band structure are removed.

The last step is the choice of $\xi_k$. We will discuss the case of entangled bands, since this is the most common case. Isolated band complexes are a trivial sub case. The targeted group of bands has a certain energy window $[E_{\text{min}}, E_{\text{max}}]$ in which it is located. When these bands are entangled with other bands it means that the projector’s character is also appearing to a certain degree in the entangled irrelevant bands outside of the core energy window while character of the irrelevant bands flows into the targeted bands inside the core window due to hybridization.

By choosing $\xi_k = 1$ for the core energy window one ensures that all targeted bands are fully included in the Hilbert subspace $\mathcal{H}$. At points where the targeted bands go outside the core energy window $\mathcal{H}$ will suffer a sudden collapse of some of its dimensions. Hence, we apply smooth Gaussian tails

$$\xi_k = \exp \left( -\left( \frac{\xi_k - E_{\text{min/ max}}}{\Delta_{\text{min/max}}} \right)^2 \right)$$ (26)

at the lower and upper end of the energy window, where $\xi_k$ is the band energy of the considered band. The width parameter $\Delta E$ and the core window boundaries $E_{\text{min/ max}}$ need to be adjusted to achieve several objectives. First, the resulting wannierized band structure should not be pulled to lower or higher energies. Secondly, Eq. (13) must not be singular and lastly the projector weight which flows into entangled bands outside the core window must be sufficient sampled by the Gaussian tails.

It turns out that a core window which is smaller than the targeted window supplied with relatively wide tails are the best recipe in most entangled cases. This choice also increases the localization of the resulting WFs. In cases like bcc iron where the $d$-bands are hybridized with $sp$-bands of several tens of electron volts band width the upper tail must be large enough (order 10 eV) to capture some $d$-weight which flows far up into the $sp$-bands. The resulting WFs are astonishingly good (see Sec. IV C). The main reason why the tails can be larger than expected is the fact that we project onto the very orbitals which are the basis of the band structure and hence get large weights from the $d$-projectors in the core window while
the tails collect only small parts of \( d \)-character. In cases of isolated band complexes the tails can be chosen very small and the core window set to the energy window of the isolated band complex.

When picking isolated band complexes for Wannierization it must be avoided that the gaps above and below are topological. Otherwise, the WFs cannot be localized due to topological obstruction. This is usually flagged by the fact that it will be hard to find a set of projectors with basis size equal to the number of targeted bands. In such cases molecular orbitals might look like a possible basis. However, the result would either be singular or not localized (the other option is breaking of symmetry, which we exclude by construction). Essentially, one has to avoid to pick parts of split elementary band representations according to the concepts of Topological Quantum Chemistry\[^{[15]}\].

There is a general recipe which circumvents this issue: always pick a set of projectors which forms a chemical basis, i.e. for instance a \( 4s4p3d \) basis for a transition metal or a \( 5s5p/6s6p \)-basis for many cases with heavy main group elements (sometimes even a simple \( 5p/6p \)-basis might work). In compounds the collection of these minimal chemical bases for all atoms contributing to the targeted bands must be chosen as projectors. This way the probability to pick split elementary band representations is drastically lowered, since the size of the projector set also increases the number of the targeted bands and chemical basis sets are likely to form whole elementary band representations. This strategy increases the size of the resulting WF model, but for many applications one actually wants the whole set of valence bands and the low lying conduction bands, which is actually formed by the chemical basis.

The algorithm described here can also be applied in the molecule mode of FPLO (which is a genuine \( 0d \) mode without the need of a simulation box) to construct basis-reduced Hamiltonian models.

III. TOPOLOGICAL ASPECTS

This section is not essential for the basic understanding of the general method of calculating SCMPWFs. It discusses the position operator matrix elements in the framework of local basis methods, which enter a complete description of the Berry connection and curvature.

A. Position operator

The Berry connection and its relation to the position operator \( r \) has been discussed widely, especially in the context of Wannier functions where it is given as the Fourier back transform of the \( r \)-matrix elements between a WF in the first cell and all other WFs. The representation of the position operator in Bloch and Wannier basis has also been extensively discussed in Ref. 13, where all expressions for Berry connection and curvature are provided although without the realization of their benefit in the study of topological aspects of electronic structures\[^{[16]}\].

Here we will reiterate the representation of the position operator and introduce a basis invariant formulation which allows for consistent and arbitrary basis transformations and naturally lends itself to application in local orbital frameworks. A natural gauge dependent approximation for the Berry connection will be given, which is important in TB frameworks, where the basis is only implicitly known. In the following we will deal with whole connection/curvature matrices to retain the transformation properties. The final Berry connection/curvature is a sub-space trace of these matrices.

We start with the obvious observation that the position operator is not translation invariant, which makes its lattice Fourier transform \( r^{\alpha \beta} \) non-diagonal in \( k \). Furthermore, the limit \( q \to k \) is badly defined (see Eqs. (B.27,B.28) in Sec. B.3)

In the space of extended (Bloch) functions let us introduce the operator identity

\[
r = e^{i k r} i \nabla_k e^{-i k r} - i \nabla_k
\]

which defines the Berry operator (discussed below)

\[
\beta_k = e^{i k r} i \nabla_k e^{-i k r}
\]

Both equations are differential operators with respect to \( k \) for all terms which are multiplied from the right. The action of \( \beta_k \) and \( \nabla_k \) hence must be understood as

\[
\beta_k \langle q^k | \Phi^k \rangle = \langle \beta_k \Phi^k | q^k \rangle + \langle \Phi^k | i \nabla_k \rangle \Phi^k
\]

\[
i \nabla_k \langle q^k | \Phi^k \rangle = \langle i \nabla_k \Phi^k | q^k \rangle + \langle \Phi^k | i \nabla_k \rangle \Phi^k
\]

(29a)

\[(29b)\]

(28)

(using the chain rule), where \( \langle \beta_k \Phi^k | \rangle \) is the isolated action of \( \beta_k \) on \( \Phi^k \).

Matrix elements of \( r \) between Bloch functions (i.e. Bloch sums or linear combinations thereof) can be written as

\[
\langle \Phi^q | r \Phi^k \rangle = \langle \Phi^q | \beta_k \Phi^k \rangle - \langle \Phi^q | i \nabla_k \Phi^k \rangle
\]

\[
\langle \Phi^q | r \Phi^k \rangle = \langle \Phi^q | \beta_k \Phi^k \rangle - \langle i \nabla_k \Phi^q \rangle \langle \Phi^k | \rangle
\]

(30a)

\[(30b)\]

(31)

where in Eq. (30a) the differentiations are not confined in both terms, while in Eq. (30b) the differentiation is confined, i.e. these are normal matrices. The unusual shape of Eq. (30b) is formally correct in that it transforms properly under basis changes such that with \( \Psi^k = \Phi^k C^k \) one gets

\[
\langle \Psi^q | r \Psi^k \rangle = \langle \Psi^q | \beta_k \Psi^k \rangle - \langle i \nabla_k C^{-\dagger} \Psi^q \rangle
\]

by bracketing Eq. (30b) with \( C^k \) from both sides and applying the chain rule Eq. (29) to both terms on the rhs. At this stage it is vital to always consider the full non-diagonal form. Note, that the last term of Eq. (30b)
 reduces to \((i \nabla_k \delta_{q,k})\) for orthonormal \(\Phi^k\), which is basis independent. (For correct transformation behavior the overlap matrix has to be kept until the end result, however.) Hence, the essential part of the position operator is the Berry connection matrix discussed in the following.

### B. Berry connection

Using the Berry operator defined above it can be shown (Sec. B 3, Eq. (B29)) that \(\beta^s_k \Phi^k\) fulfills the Bloch theorem and hence \(\langle \Phi^q | \beta^s_k \Phi^k \rangle\) is diagonal in \(k\). Each Bloch function can be written as \(\Phi^k = e^{ikr} \psi^k\) which leads to

\[
\langle \Phi^q | \beta^s_k \Phi^k \rangle = \delta_{q,k} \langle \psi^k | i \nabla_k \psi^k \rangle
\]

(32) which contains the matrix underlying the Berry connection

\[
A^k_\phi = \langle \psi^k | i \nabla_k \psi^k \rangle
\]

(33) Consequently, the position operator matrix elements in a Bloch basis consist of a \(k\)-diagonal well defined part, the Berry connection matrix, and the gradient of the overlap matrix (delta function for orthonormal bases) (which also contains diagonal elements, implicitly) and this is true in all bases.

An illustrative example of the meaning of Eq. (31) is to formally calculate the dipole integral \(D = \int r n(r) d^3r\) of the density \(n(r) = \sum_{k,n} f_{kn} | \Phi^k_n |^2\) which using Eq. (31) becomes

\[
D = \sum_{k,n} f_{kn} \left[ A^k_{\phi,nn} - \lim_{q \to k} (i \nabla_k \delta_{q,k}) \right] d^3r
\]

(34) of which the first term, the essential part of \(r\), is the polarization \(P^s\) of Ref. 17 up to constant factors.

The Berry connection matrix is Hermitian if the Bloch basis is orthonormal

\[
A^{k+}_\phi = A^k_\phi - i \nabla_k S^k_\phi
\]

(35) e.g. for Wannier functions and Hamiltonian eigen functions, which remedies its asymmetric formulation (derived via \(i \nabla_k S^k_\phi = i \nabla_k \langle \Phi^k | \Phi^k \rangle = i \nabla_k \langle e^{-ikr} \Phi^k | e^{-ikr} \Phi^k \rangle\).

The basis change \(\Psi^k = \Phi^k C\) of the \(k\)-diagonal Berry connection matrix in Eq. (31) reads explicitly

\[
A^k_\psi = C^{k+} A^k_\phi C^{k+} + C^{k+} S^k_\phi i \nabla_k C^{k+}
\]

(36) If one interprets \(\Phi\) as the Bloch sums of local orbitals or Wannier functions and \(\Psi\) as the eigen functions of the Hamiltonian, the Berry connection consists of a term \(C^{k+} A^k_\phi C\), where \(A^k_\phi\) shall be called basis-connection, and the usual gradient-of-coefficients term (with an overlap matrix for the non-orthonormal case). The latter term is always available while the former is not known in most situations involving tight binding models. Below, a proper approximation for the basis-connection will be introduced. Practically, Eq. (36) is not useful due to the random phase issue in \(\nabla_k C^k\). It will be useful in deriving corrections to the Berry curvature later on.

For a plane wave basis \(\Psi^k = \sum_G e^{i (k+G) r} C^k_{G}\) the basis functions are the exponentials and the basis connection matrix is zero:

\[
A^k_{G,G'} = \langle e^{i G' r} | i \nabla_k e^{i G r} \rangle = 0
\]

(37) For a local basis we introduce vector valued functions

\[
(r \Phi)^k_s = (r - R - s) \Phi_s (r - R - s)
\]

(38) with Bloch sums

\[
(r \Phi)^k_s = \frac{1}{\sqrt{N}} \sum_{k} e^{ik(R + \lambda s)} (r \Phi)^k_s
\]

(39) which leads to \((\lambda = 1 - \lambda)\)

\[
(\beta^s_k \Phi^k) = (r \Phi)^k_s + \lambda \Phi^k s
\]

(40) and the basis connection

\[
A^k_{\psi,s',s} = \langle \Phi^s_{s'} | \beta^s_k \Phi^k \rangle = \langle \Phi^s_{s'} | (r \Phi)^k_s \rangle + \lambda S^k_{s's} s
\]

(41) The \(\lambda\)-term is zero for the relative gauge, but non-zero for the periodic gauge of the Bloch sums. The first term on the rhs., which shall be called reduced position operator, can be straightforwardly evaluated as

\[
\langle \Phi^s_{s'} | (r \Phi)^k_s \rangle = \sum_{\lambda} e^{i k (R + \lambda (s-s'))} \langle \Phi^s_{s'} | (r \Phi)^k_s \rangle
\]

(42) from local orbital matrix elements, which are translation invariant:

\[
\langle \Phi^s_{s'} | (r \Phi)^k_s \rangle = \langle \Phi^s_{s'} | (r \Phi)^{R+s-s'}_s \rangle
\]

(43) and Hermitian if \(\Phi\) is orthonormal.

Eq. (43) is very useful due to its invariance, which makes them transferable on a lattice. It is the ideal form to store position operator matrix elements (also for the Wannier functions).

The relation to the position operator follows by expanding the \(r - R - s\) part of Eq. (38) in Eq. (41), using

\[
i \nabla_k S^k_{s's} = - \sum_{\lambda} e^{i k (R + \lambda (s-s'))} S^k_{s's'} R_s \cdot \langle R + \lambda (s-s') \rangle
\]

and reads

\[
A^k_{\psi,s',s} = \frac{1}{N} \sum_{\lambda} e^{i k (R + \lambda (s-s'))} \langle \Phi^s_{s'} | r | \Phi^k_s \rangle
\]

(44)
In the periodic gauge $\lambda = 0$ and for WFs $S_{q,s}^k = \delta_{q,s}$ this is the expression usually given. However, Eq. (41) is the simpler and arguably the more natural choice.

In order to calculate the basis-connection Eq. (41) for the Wannier functions one needs to express the reduced position operator matrix Eq. (42) of the WFs in terms of the basis orbitals. Note, that in our implementation the relative gauge $\lambda = 1$ is used and hence the last term in Eq. (41) is zero and basis-connection and reduced position operator matrix are identical.

Our Wannier functions are related to the LOs via Eq. (18). The Berry connection in the Wannier basis hence is obtained by a basis change Eq. (36) when letting $\Psi \to w$ and $C \to a$:

$$A^k_w = a^k + A^k_0 a^k + a^k + S^k_w i \nabla k a^k$$

(45)

The first term on the rhs. is given by Eq. (41) with $\Phi$ being our LOs, while the gradient needs special treatment. It turned out that a direct calculation via the real-space coefficients $a_{0c}, R_c$ is not very accurate. Instead we use a numerical differentiation technique akin to the one described in Ref. 1. Finally, $A^k_w$ is Fourier back transformed which gives $\langle w_{0c} | (r w)_{R,C} \rangle$ in real space which is stored together with the other WF information for post-processing. From this the Wannier basis connection $A^k_w$ can be retrieved via Eqs. (41,42) with $\Phi \to w$, $S_{q,s} \to \delta_{q,s}$ in either Bloch sum gauge.

The reduced position operator matrix elements Eq. (43), especially for orthonormal bases, are quite intuitive. The on-site elements are the integral of two orbitals multiplied with the position vector from the orbital origin. For two equal orbitals which are eigenfunctions of inversion this integral is always zero due to parity. For other cases and also for the off-site case one can argue that the orthonormality of the orbitals/WFs will somewhat transfer to these matrix element to make them rather small.

Also the reduced elements are Hermitian (for orthonormal bases) and transform according to all symmetries (see Sec. B.4). Hence, they are a reasonable choice to be considered as the physically important matrix. By approximating it by zero we get

$$A^k_{w,q,s,s'} \approx \lambda \delta_{q,s,s'}$$

(46)

for orthonormal bases. Note, that this is nonzero in the periodic gauge, in which the Bloch sums are $k$-periodic.

The basis connection can only be true in one gauge, which forces a choice which matrix to put to zero. We argue that the relative gauge, which is usually not mentioned in the context of Wannier functions is the one in which the WF basis-connection can be neglected. We will show numerical evidence for this later. This might be important in tight-binding models, where the basis is not explicitly known and hence the basis connection is missing. Furthermore, approximation Eq. (46) is needed to preserve symmetry properties (Sec. B.4), which further indicates the correctness of this choice.

C. Berry curvature

In this section we derive the Berry curvature matrix

$$F_\Psi^k = \nabla k \times A^k_\Psi$$

(47)

in eigenstate representation. Starting from the (Wannier) basis connection $A^k_w = \langle w_k | \beta_k w \rangle$ (usually reassembled from the reduced position operator in WF basis during post processing) we make a basis change $\Psi_\Psi = w_k C^k$ using Eq. (36) with $\Phi \to w$:

$$A^k_\Psi = C^k + A^k_w C^k + C^k + S^k_w i \nabla k C^k$$

(48)

where the overlap matrix is the unit matrix for orthonormal WFs. Let’s introduce some short hands

$$\langle A \rangle_C = C^k + A^k_w C^k$$

(49)

$$\langle S i \nabla \rangle_C = C^k + S^k_w i \nabla k C^k$$

(50)

$$\langle f \rangle_C = C^k + (\nabla k \times A^k_w) C^k$$

(51)

Differentiation of the normalization $C^k + S^k_w C^k = 1$ gives

$$\langle S i \nabla \rangle_C^+ = \langle S i \nabla \rangle_C + C^k + (i \nabla k S^k_w) C^k$$

(52)

and Eq. (35) gives

$$\langle A \rangle_C^+ = \langle A \rangle_C - C^k + (i \nabla k S^k_w) C^k$$

(53)

Then using $\nabla \times u w w = (\nabla u) \times v w + u (\nabla \times v) w - w w \times (\nabla w)$, where $u, w$ are matrices and $v$ is a vector of matrices, and the identity $C^k C^k + S^k_w = 1$ and commuting non-vector matrices through the $\times$-symbol one gets

$$A^k_\Psi = \langle A \rangle_C + \langle S i \nabla \rangle_C$$

(54a)

$$F_\Psi^k = \langle f \rangle_C - i \langle A \rangle_C^+ \times \langle A \rangle_C + i A^k_w + A^k_w$$

(54b)

$$= \langle f \rangle_C + i \langle A \rangle_C^+ \times \langle S i \nabla \rangle_C + i \langle S i \nabla \rangle_C^+ \times \langle A \rangle_C$$

$$+ i \langle S i \nabla \rangle_C \times \langle S i \nabla \rangle_C$$

(54c)

Note, that the cross product expressions are matrix products at the same time $(a \times b)_{a_{lm,k}} = \sum_{ij} \varepsilon_{ijk} a_{nl} b_{im,j}$. The second term grouping of $F_\Psi^k$ corresponds to the result of Ref. 18 while the first emphasizes the symmetry between the curvature in both bases and is discussed in the appendix of Ref. 18.

Due to Eq. (35) $A^k_\Psi$ is Hermitian, since $\Psi^k$ is orthonormal, and the basis curvature $F^k = \nabla \times A^k_\Psi$ is Hermitian since $S^k_w$ is smooth and $\nabla \times S = 0$. This makes $\langle f \rangle_C$ and hence also $F_\Psi^k$ Hermitian. Eqs. (54) are true for any basis $w^k$ and hence could also be used directly in the non-orthogonal FPLO basis. (If additionally $\Psi^k$ were to be non-orthonormal $(S^k_w)^{-1}$ needs to be inserted before or after all $\times$-symbols in Eq. (54).)

In practice, $\langle S i \nabla \rangle_C$ is obtained in the parallel transport gauge[18], which is not a periodic gauge and hence makes Eq. (54a) useless for topological applications but
it allows to calculate Eq. (54b). In detail non-degenerate perturbation theory of first order gives
\[ \langle Si\nabla\rangle_{C,mn} = iC^k_m \left( \nabla_k H^k_n \right) C^k_m - C^k_m \left( \nabla_k S^k_n \right) C^k_m z^k_n, \]
\[ \cdot (1 - \delta_{mn}) \]  
where the gradient of the Hamiltonian in Wannier basis reads
\[ \nabla_k H^k_n = i \sum_R e^{ik \left( R + \lambda (s-s') \right)} \left( R + \lambda (s-s') \right) \cdot \left( w_{0s} | \hat{H} | w_{Rs} \right). \]

For an orthonormal basis the gradient of the overlap vanishes. The basis curvature \( f^k \) is obtained by directly taking the curl of the exponential in Eq. (42) (\( \Psi \to w \)) and inserting into Eq. (41). (The curl of the overlap can be obtained in a similar way for non-orthonormal bases.)

We need to discuss the non Abelian (degenerate) case, in which the Berry curvature matrix obtains a covariant correction in each degenerate subspace[19]. If \( P_i \) denotes the projector onto the \( i \)-th subspace (degenerate or not) one needs to write
\[ P_i F_{\Psi n A} \cdot P_i = P_i \left( F_{\Psi}^k - i A^k_\Psi P_i \times P_i A^k_\Psi \right) P_i \]
for each subspace. For a degenerate subspace \( A^k_{\Psi,ii} = P_i A^k_{\Psi,ii} P_i \) is a single vector-valued matrix element and hence \( A^k_{\Psi,ii} \times A^k_{\Psi,ii} = 0 \) and the correction is zero. In a degenerate subspace only the average of the trace is physically meaningful. In the case of an orthonormal basis this allows to introduce a unitary transformation, which ultimately leads to \( i \text{Tr} P_i \langle A \rangle_C P_i \times P_i \langle A \rangle_C P_i = 0 \) and permits to discard such block diagonal terms (if only degenerate subspace averages are considered). Inserting Eq. (54b) into Eq. (57) and introducing \( Q_i = 1 - P_i \) one gets for orthonormal bases
\[ P_i F_{\Psi n A} \cdot P_i = P_i \left( f_C - i \langle A \rangle_C Q_i \times Q_i \langle A \rangle_C + i A^k_\Psi Q_i \times Q_i A^k_\Psi \right) P_i \]
\[ = P_i \left( f_C - i \langle A \rangle_C Q_i \times Q_i \langle A \rangle_C + i A^k_\Psi Q_i \times Q_i A^k_\Psi \right) P_i \]  
\[ (58) \]

This expression contains \( \langle Si\nabla\rangle \) bracketed between \( Q_i \) and \( P_i \) and hence only matrix elements between different subspaces such that the energy denominator in Eq. (55) is never zero. This solves the division by zero issue. In essence the non-Abelian Berry curvature is obtained from Eqs. (54b,54c) by removing the diagonal blocks of all subspaces from \( \langle A \rangle_C \) and \( \langle Si\nabla \rangle_C \).

Ref. 18 discusses the terms of \( F^k \) and argues that the \( \langle Si\nabla \rangle \times \langle Si\nabla \rangle \)-term of Eqs. (54c) is by far the leading term. This would allow to ignore the basis connection/curvature terms, especially if they are not obtainable. However, this is not necessarily correct. In Sec. B 4 it is shown that the term \( \langle Si\nabla \rangle \times \langle Si\nabla \rangle \) does not transform properly under symmetry in the periodic gauge. However, if \( A^k_\Psi \) is approximated by Eq. (46) in Eqs. (49,54a) proper symmetry transformation is restored. This shows that the term grouping Eqs. (54b,58) is to be preferred and in fact under the approximation the only term left is \( i P A^\text{F} \Psi Q \times Q A^\text{F} \Psi P \) with approximated \( A^\text{F} \Psi \), which reduces to the \( \langle Si\nabla \rangle \)-only term in relative gauge.

Another argument for the grouping Eq. (54b) is the fact that both \( i P A^\Psi Q \times Q A^\Psi P \) and \( (f)_C - i \langle A \rangle_C Q_i \times Q_i \langle A \rangle_C \) are invariant under symmetry and with respect to the Bloch sum gauge choice controlled by \( \lambda \) (see Sec. B 5) while the other grouping is not, especially not the approximation of only taking the \( \langle Si\nabla \rangle \times \langle Si\nabla \rangle \)-term. Finally, if the \( \Psi \)-basis is complete \( \langle f \rangle_C - i \langle A \rangle_C Q_i \times Q_i \langle A \rangle_C \) will go to zero and \( F^k_{\Psi n A} = i P A^\Psi Q \times Q A^\Psi P \) is the complete Berry curvature.

In FPLO before version 19 only the periodic gauge was implemented; now both gauges as well as the full basis connection are available. Furthermore, for historical reasons the FPLO implementation contains an overall minus sign for the Berry connection/curvature as if we defined \( A^k = \langle u^k \mid -i \nabla_k u^k \rangle \) instead of Eq. (33). Taking this sign into account we are consistent with the data presented in Ref. 18.

On a final note, in cases of TB-models for which no explicit WFs but the nature/symmetry of these functions are known, instead of approximation Eq. (46) one could derive parametrized analytic expressions for leading matrix elements in Eq. (43) based on the nature of the WFs which inserted into Eq. (41) give a reasonable approximation for the basis connections.

IV. FEATURES OF MAXIMALLY PROJECTED WANNIER FUNCTIONS

The examples shown here are not discussed in terms of their DFT setup. More details can be found in Sec. C.

A. Work flow

The WF creation is controlled by the choice of projectors \( \phi \), the energy window, which can be chosen individually for each projector (although most of the time a global window is used) and the real space cutoff \( \rho \) and a coefficient threshold (to save space), below which WF data are discarded. The latter might need to be changed from its default if the targeted bands are very flat.

The projectors can be single FPLO orbitals, for which only a site number, an orbital descriptor and optionally local quantization axes need to be defined. In full relativistic mode, the projectors can be in four-spinor quantum numbers \( nlj \mu \) or in pseudo-non-relativistic qns. \( nlm \sigma \), in which case separate local spin axes can be defined. Also unnormalized linear combinations of LOs (MOs) can be chosen as projectors, which are defined by picking an imagined Wannier center, from which different vectors to the contributing sites must be de-
fined. Furthermore, for each contribution a site number, a weight and local axes must be specified. The main constraint is that the set of projectors must be closed under all symmetry operations. Band disentangling and the quality of the fit is controlled by proper choice of projectors and energy window.

Alternatively, there is an automatic mode, which picks either all FPLO orbitals as projectors or only the subset which does not clearly form semi-core states. The FPLO basis is relatively small with of 15 to 35 orbitals per atoms for most of the periodic table, which avoids a too large resulting WF basis. Removal of semi-core states further decreases the basis size. The advantage of this mode is application in automated calculations and the fact that no energy window needs to be defined. The cutoff \( \rho \) however might need to be increased (preferably to it’s maximum) due to the larger extent of higher energy WFs.

To construct WFs with FPLO a WF input file needs to be created, which can be done by hand or with the help of a python module, which allows to efficiently create single-orbital projectors for any set of sites/orbitals as well as molecular orbital projectors. Starting from a converged calculation and with the input file present an FPLO-run will produce a file with all the data needed to construct the Wannier functions in a second run (see Fig. 1). At this stage the real space Wannier representation, the symmetry operations and all demanded operators (like reduced position operator, full relativistic spin operators \( \beta \Sigma \) and the exchange-correlation (xc) magnetic field) are written to a separate file for post processing. Additionally, WF output on a real space grid for visualization purposes can be requested.

### B. Model extraction

The first example demonstrates the construction of a single band model of the anti-bonding (AB) \( Cu \, 3d_{x^2−y^2} \) band for the infinite layer cuprate \( CaCuO_2 \). The band structure (Fig. 2) contains the prototypical AB band in the energy interval \([-2, 2]\) eV around the Fermi level highlighted by the FPLO-orbital band character. The bonding part below -3 eV, which is more diffuse, is of course also present.

![FIG. 1. The work flow to construct and use SCMPWFs in the FPLO implementation.](image)

![FIG. 2. (color online) (a) the band structure (black) of \( CaCuO_2 \), the FPLO orbital \( 3d_{x^2−y^2} \) band character (red) and the single band Wannier fit (cyan). (b) the unit cell containing the Wannier function at isovalue 0.04. (c) the maximum of orbital contributions to the WF in logarithmic scale as a function of WF-orbital distance. (d) the WF printed along the (100)-axis through the WF center (black), the \( Cu \, 3d_{x^2−y^2} \) (orange) and the \( O \, 2p_x \) (red) orbitals scaled according to their amplitude.](image)
The $k$-mesh has $12^3$ points. The core energy window is chosen as $[-1,1]$ eV with symmetrical Gaussian tails, Eq. (26), of width $\Delta = 2$ eV as indicated on the right side of the band structure. This window is considerably overlapping with the bonding part, yet the resulting WF (cyan) follows the anti-bonding part, where $h^k$ is dominant. The fit is not perfect, owed to the fact that there are lots of hybridizations interrupting the unbroken flow of the band character. It however samples the essence of the targeted band. The WF fit was calculated from WF of the band character. It however samples the essence of the targeted band. The WF fit was calculated from WF data with a real space cutoff $\rho = 25a_B$. To illustrate the exponential localization of the WF the maximum of the amplitudes $\max(|a_{Rs,0e}|)$ of contributing orbitals around the WF, Eq. (24), as a function of WF-orbital-distance is shown in Fig. 2c for a cutoff $\rho = 70a_B$. Note, the logarithmic scale.

Fig. 2b shows the crystal structure and the WF, which clearly has $x^2 - y^2$ symmetry and sizable oxygen hybridization tails as to be expected for this case. The WF along the (100)-direction through the WF center is plotted in Fig. 2d. Additionally, the Cu 3$d_{x^2-y^2}$- and O 2$p_z$-orbitals scaled according to their amplitude $a_{Rs,0c}$ is shown. The circles show the position of the atoms in the same color as the orbitals. This WF is rather extended since it needs to describe a Cu-O hybridized band via a single WF. It is however exponentially localized as indicated by the inset which shows $|w|$ in logarithmic scale. Note, that the projector consists only of the Cu 3$d_{x^2-y^2}$-orbital (orange), all other contributions to the WF are pulled in due to the projection process.

### C. Band disentangling

A more complex example which demonstrates band disentangling is the 3$d$-only WFs for spin polarized fully relativistic bcc iron (see Sec. C2). Spin is no longer a good qu. although spin orbit coupling and hence spin-mixing is quite small. The complication is the hybridization with the 4$s$-$p$-bands which have a band width of several tens of electron volts, which means that bands which start as $d$-bands at some $k$-point can mutate into free electron bands away from the point. Hence, the predominately 3$d$-character Hilbert space is not restricted to a finite energy window and the corresponding WFs cannot follow the $d$-band structure at all points.

We performed a Wannier fit using 3$d$ orbitals with non-relativistic symmetry as projectors (see Sec. B1, Eq. (B7)). These are obtained from a linear combination of the four spinors Eq. A1, such that the resulting orbital transforms as a real Harmonic (which is an in-built option besides projection onto the relativistic four spinors themselves). The energy window as indicated at the right side of Fig. 3a has a core region $[-5,0]$ eV and Gaussian tails of width $\Delta = 7$ eV. We used a $k$-mesh with 16$^3$ points in the primitive reciprocal cell and a cutoff $\rho = 25a_B$ for the real space representation of the WFs. Although, the energy window is quite large, the resulting Wannier fit (bright green and red) follows the bands with 3$d$-character (milky green and red) very closely at most parts of the Brillouin zone (BZ). The WFs also resolve the original spin character.

![FIG. 3.](image) (color online) (a) band weights of the FPLO calculation in pastel colors and Fe 3$d$ WF fit in bright colors (see text). (b) the 3$d_{x^2-y^2}$ WF printed along the (100)-axis through the WF center (black), the Fe 3$d_{x^2-y^2}$ local orbital (orange). Left inset shows $|w|$ along the same path in logarithmic scale. Right inset shows the maximal orbital contributions to the WFs in logarithmic scale as a function of WF-LO distance.

Fig. 3a shows three regions marked by numbers. In region 1 the highest minority $d$-band at $\Gamma$ becomes a free electron band (milky blue) as it progresses towards the H-point. At the same time an $s$-$p$-band enters the $d$-region from below. So, the WFs must reconnect the manifold at $\Gamma$ to the manifold at $H$ by interpolating through some dispersion not existing in the DFT band structure (the same happens for the majority spin bands).

One of the lowest (degenerate) minority bands (-3 eV) at $H$ bends upwards and turns into an $s$-$p$-band at $N$, as marked by the right arrow in region 2, while the missing $d$-character flows in from above 2 eV. At the N-point all $d$-weight sits in the two bands at 1-1.5 eV. Consequently, the WF (left arrow) must deviate from the DFT band to smoothly interpolate the pure $d$-bands between $H$ and $N$.

A similar situation as in region 1 occurs in region 3 between the H- and $\Gamma$-point and the $\Gamma$- and P-point, re-
spectively. $d$-weight flows to lower energies from $N$ to $\Gamma$ and to higher energies between $\Gamma$ and $P$, which forces the WFs to deviate from the original band structure. Between, $P$ and $H$ the second and forth lowest Wannier band deviate from the DFT bands by 180 meV (not visible in the fat-band plot). Besides these regions the WF fit follows the original bands rather closely.

Fig. 3b plots the WF along the (100)-direction through the Wannier center (black) together with the $3d_{x^2-y^2}$ orbital (orange). The circles denote the atom positions. There are some notable orthogonalization tails at first and second neighbor, but otherwise the WF and the local orbital are nearly identical. The right inset shows the WF along the same direction as the main panel with logarithmic $y$-axis. Clearly, the expansion coefficients as well as the WF itself are exponentially localized and the localization is similar to that of the $d$-orbital.

$4s$-orbital are nearly identical. The right inset shows max$(|\alpha_{Rs,0c}|)$ (Eq. (24)) as a function of WF-LO distance and the left inset shows the WF along the same direction as the main panel with logarithmic $y$-axis. Clearly, the expansion coefficients as well as the WF itself are exponentially localized and the localization is similar to that of the $d$-orbital.

There are some notable orthogonalization tails at first and second neighbor, but otherwise the WF and the local orbital are nearly identical. The right inset shows max$(|\alpha_{Rs,0c}|)$ (Eq. (24)) as a function of WF-LO distance and the left inset shows the WF along the same direction as the main panel with logarithmic $y$-axis. Clearly, the expansion coefficients as well as the WF itself are exponentially localized and the localization is similar to that of the $d$-orbital.

$4s$-orbital are nearly identical. The right inset shows max$(|\alpha_{Rs,0c}|)$ (Eq. (24)) as a function of WF-LO distance and the left inset shows the WF along the same direction as the main panel with logarithmic $y$-axis. Clearly, the expansion coefficients as well as the WF itself are exponentially localized and the localization is similar to that of the $d$-orbital.

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To compare to Ref. 18 (Wang06) and to illustrate the Berry curvature terms and their grouping we show the results for the Berry curvature of full-relativistic spin-polarized bcc Fe in Fig. 5 (for the 3d4s4p-Wannier model of Sec. IV C). By comparing the Fermi surface contour to Ref. 18 it becomes clear that the two band structure codes do not give the exact same result, which is not surprising given that the cited results use a pseudo-potential code and treats the relativistic effects differently from our full 4-component treatment. It is likely that our results compare better for slightly different lattice constants. Nevertheless, the Berry curvature plot and the curvature plotted along the Brillouin zone path $\Gamma(000)$-H(010)-P(111) compare well. Fig. 5b shows the two different ways of grouping the terms. The left panel uses the grouping of Wang06, while the right panel shows the grouping Eq. (54b) (also discussed in the appendix of Wang06), preferred by us for symmetry, gauge invariance and approximation reasons.

In bcc Fe there is only one site at $(000)$ and hence the Wang06 shows that the $\langle S|\nabla \times (S|\nabla) \times (A|C) \rangle$ and $\langle f|C \rangle$ are quite small. The approximation Eq. (46) is zero and hence the approximated curvature is the $\langle S|\nabla \times (S|\nabla) \times (A|C) \rangle$ term itself.

Our preferred term grouping in the right panel shows that $A_\Psi \times A_\Psi$ is even closer to the total result while $\langle f|C \rangle - \langle A|C \rangle \times \langle A|C \rangle$ is rather small. We also integrated the Berry curvature over the ir-
reduce the part of the BZ (with subsequent symmetrization) to obtain anomalous Hall conductivity (AHC). To speed up this quite time consuming calculation we reduced the real space cutoff to $\rho = 25a_B$ which leads to small band energy errors of maximal 200 meV around the N-point, but yields a smaller Wannier model. For a mesh with $300^3$ k-points in the total BZ we obtain $715.4\Omega cm$ and $720.5\Omega cm$ for the $\nabla \times \nabla$-only and the full Berry curvature, respectively. For $600^3$ k-points this becomes $717.9\Omega cm$ and $722.9\Omega cm$. This is a bit smaller than the $756.8\Omega cm$ reported by Ref. 18, which used a finer adaptive integration. However, their smallest mesh which compares to our largest also deviates by about the same amount, which is most likely due to the method/band structure differences discussed above.

To further assess the validity of the general arguments of Sec. III A in favor of our preferred term grouping we show the Berry curvature results for two more cases. The left panel of Fig. 6 shows the Berry curvature along a BZ-path for spin-polarized full-relativistic B2 FeAl, which is obtained from bcc Fe by replacing the iron at the unit cell center by Al (see Sec. C 3), while the right panel shows $\mathbf{F}$ for non-spin-polarized full-relativistic HgS along the same path (different high symmetry point names). The top and middle rows show the results for the term-grouping of Wang06 in the periodic and relative gauge, respectively, while the bottom row shows the preferred term-grouping which is independent of the Bloch sum gauge. Additionally, in the top row the $A_{\Psi} \times A_{\Psi}$-term is shown within the approximation Eq. (46), which is distinct in the periodic gauge and equivalent to the $\langle S \nabla \rangle_C \times \langle S \nabla \rangle_C$-term in relative gauge.

Clearly, for FeAl in the periodic gauge the individual terms in the Wang06 grouping are larger than the total which demonstrates the necessity of including the basis connection terms. Interestingly, the approximated $A_{\Psi} \times A_{\Psi}$-term in periodic gauge (which equals the $\nabla \times \nabla$-term in relative gauge) is quite close to the total. The preferred grouping shows as for bcc Fe that the $A_{\Psi} \times A_{\Psi}$-term is largely dominant.

For HgS the situation is less satisfactory. We first note that all individual terms in the Wang06 grouping in periodic gauge (top right panel) are nonzero along $\Gamma-X$, although the total as well as the approximated $A_{\Psi} \times A_{\Psi}$-term are zero due to symmetry (non-spin polarized). In the relative gauge (middle right panel) as well as in the preferred grouping (bottom right panel) the symmetry is preserved, which confirms the general arguments of Sec. III A. On the line X-L neither term grouping has a dominant term.

E. Molecular orbital projectors

Up to now we always projected onto single orbitals, which creates WFs which look like these orbitals in the...
was chosen such that the most part of the target bands where sampled by a larger upper Gaussian tail with \( \Delta_E = 7 \text{ eV} \). Due to the weak hybridization with the \( 2p_z \)-band away from the high symmetry lines, the WF band energies at the band bottom along \( \Gamma-A \) would be pulled to higher energies, compared to the FPLO bands, if a simple rectangular window encompassing the target bands would be chosen.

The 3 bands of the WF-fit in Fig. 7a accurately follow the FPLO band structure. Fig. 7c shows an iso-surface of one of the bonding \( sp^2 \) WFs together with a semi-transparent density plot of the WF in the plane through it's center. Along a path, indicated by the thin line through the Wannier center, the WF has a profile as shown in Fig. 7b, where the circles mark the positions of the boron atoms. The WF is clearly localized as shown by the logarithmic plot of \( |w| \) along the path in the left inset and the logarithmic plot of the maximal orbital contributions (Eq. (24)) to the WF in the right inset.

F. Molecule Wannier functions

In FPLO molecules are treated without a simulation box, since all orbitals have a finite compact support. The algorithm described in Sec. II A is not restricted to bulk materials. The only places were the extended nature of a sytem comes into play are \( k \)-sums and by using the \( \Gamma \)-point only (effectively no sum) one can also create molecule WFs. To illustrate this we will determine the smallest model Hamiltonian for an \( H_2O \) molecule (see Sec. C6). There are 4 occupied valence MOs formed by \( O-2s/2p \) and \( H-1s \) orbitals of which one is formed to nearly 100\% by the \( O-2p_z \) orbital, whose lobes point out of the \( O-H \) plane and which hence is decoupled by symmetry from the other orbitals in lower orders, so that this orbital can be neglected in a model. To obtain WFs we chose the \( O-2s \), \( O-2p_y \) and the two \( H-1s \) orbitals as projectors. The \( O-2p_x \) orbital will be pulled into the Wannier basis via hybridization tails at the other WFs by choosing an energy window \( [-24,0.3] \text{ eV} \) with an upper \( \Delta = 1 \text{ eV} \), which encompasses the four lowest valence MOs and some unoccupied states. This basis will fit the three mixed occupied MOs and one unoccupied MO. The corresponding Wannier model Hamiltonian Eq. 59 will have the same eigen energies as the corresponding MOs in the DFT calculation Eq. 60 except for the omitted orbital. The Hamiltonian parameters are

\[
H = \begin{pmatrix}
 h_{2s} & h_{2s,2p_x} & h_{2s,1s} & h_{2s,1s} \\
 h_{2s,2p_x} & h_{2p_z} & h_{2p_{1s}} & h_{2p_{1s}} \\
 h_{2s,1s} & h_{2p_{1s}} & h_{1s} & h_{1s,1s} \\
 h_{2s,1s} & h_{2p_{1s}} & h_{1s,1s} & h_{1s}
\end{pmatrix}

\] (59)
Isosurfaces of the resulting WFs in real space are shown in Fig. 8.

\[
E = \begin{pmatrix}
-24.398 \\
-12.509 \\
-8.6791 \\
-6.6177 \\
0.3046
\end{pmatrix}
\text{ (60)}
\]

Isosurfaces of the four WFs of the minimum model for H$_2$O for an isovalue of 0.09. The character is denoted at the side. The atoms are shown as white (H) and red (O) spheres.

**G. Post processing**

For post processing of the Wannier function data a python package is provided. It reads the Wannier data and maps them onto a chosen structure. This can be the original unit cell forming the original lattice, any supercell and corresponding lattice, a semi infinite slab or a finite slab based on a chosen supercell. If the original lattice is 2d the resulting slabs are 1-dimensional. The slabs are idealized in the sense that no surface relaxation effects can be considered; it is a straightforward mapping of the original hoppings/data onto the sites forming the slabs. For the semi-infinite slab the termination can be chosen and for the finite slab both terminations can be chosen and an arbitrary number of atoms can be removed to get a rough estimate of such effects on the surface states.

The python package offers an interface to obtain the Hamiltonian and all requested operators for a user specified \( k \)-point, which provides maximum flexibility. It also offers ready made procedures to calculate several properties depending on the chosen structure mapping. For all but the semi-infinite mapping the band structure, including band weights, and Fermi surfaces and Fermi surface cuts can be calculated. For the original and super cell mappings a bulk-projected band structure can be obtained which integrates the band structure along a chosen \( k \)-direction. This offers the spectral densities with surface effects excluded.

For the semi-infinite slabs a Green’s function method[20, 21] is used to calculate spectral densities as a function of energy and momentum (band structure plot equivalent) or in a \( k \)-plane (Fermi surface cut equivalent). Fig. 9 shows an example of this for the ternary Type-II Weyl semi-metal TaIrTe$_4$[22].

**FIG. 8.** (color online) Isosurfaces of the four WFs of the minimum model for H$_2$O for an isovalue of 0.09. The character is denoted at the side. The atoms are shown as white (H) and red (O) spheres.

**FIG. 9.** (color online) Surface spectral densities (SD) for TaIrTe$_4$ of a semi infinite slab with (001)- (left column) and (001)- (right column) termination. The crosses mark the position of the Weyl point. Upper row: Fermi surface SD, the arrows mark the Fermi arcs. Lower row: the corresponding energy resolve SD along a line \( k_y = 0.125 \). The arrows mark the surface bands forming the arcs.

For the mapping onto the original lattice the symmetry operations and their eigenvalues can be analyzed. Furthermore, an adaptive algorithm for the search for Weyl points (WP) is available. We use a 3d version of the methods of Ref. 23 to determine the chirality of each micro-cell of a regular subdivision of the primitive unit cell. Then in refinement steps for each cell with a non-trivial chirality the cell is subdivided into 8 sub-cells for which the chirality again is determined. The subdivision stops if the sub-cell size falls bellow a specified threshold. For each found Weyl point it’s veracity can be be checked by calculating an integral of the Berry curvature over a small spherical shell around the WP, as well as creating a plot of the Berry curvature field for visual confirmation. The resulting Fermi arks can be analyzed by calculating the spectral density for a mapping onto a semi-infinite slab or by calculating the band structure and Fermi surface cuts for a finite slab.

Also for the original mapping the \( Z_2 \) topological indices can be determined for non-centrosymmetric lattices using a Wannier-center algorithm[24] with automatic determi-
nation of the indices [25]. This algorithm is also directly available from the FPLO code itself, by internally Löwdin orthogonalizing the whole FPLO basis (which results in full-basis Wannier functions), but it is faster to use a Wannier model due to the resulting basis reduction. For centrosymmetric lattices FPLO itself calculates the $Z_2$ indices.

The Berry curvature can be calculated band wise for any non-slab mapping either with approximated reduced position operator as discussed in Sec. III.A or in full form, which also gives access to the anomalous Hall conductivity (Sec. IV.D). By using the symmetry information mirror Chern numbers [26, 27] for topological crystalline insulators can be obtained as well.

Besides Hamiltonian and reduced position operator in full-relativistic mode the spin operators and the exchange correlation magnetic field can be extracted. This is achieved by starting from the xc-term of the Hamiltonian

$$H_B = \langle \Phi^k | \beta \Sigma B | \Phi^k \rangle$$  \hspace{1cm} (61)

where the $4 \times 4$-matrices $\beta$ and $\Sigma$ are defined as

$$\beta = \begin{pmatrix} 1_{2 \times 2} & 0 \\ 0 & -1_{2 \times 2} \end{pmatrix}, \quad \Sigma = \begin{pmatrix} \sigma & \sigma \\ \sigma & -\sigma \end{pmatrix},$$ \hspace{1cm} (62)

$B(r)$ is the xc-field and $\Phi^k$ is the column-vector of the FPLO basis orbitals. Assuming basis completeness the identity

$$1 = |\Phi^k_k\rangle \bigg\langle \Phi^k_k |$$ \hspace{1cm} (63)

can be inserted to separate the field followed by the Wannier transformation Eq. (18) using the identity $1 = S_w^k = \alpha^k \Sigma \sigma \alpha^k$ which leads to Wannier representation

$$H_B^w = \langle \beta \Sigma w | B \rangle w$$ \hspace{1cm} (64)

$$\langle \beta \Sigma w | = \langle w | \beta \Sigma | w \rangle \hspace{1cm} (65)$$

with separate matrix representation for the vector of spin operators and of the xc-field.

The spin operator matrix can be used to add model magnetic fields as was for instance done in Ref. 28 to simulate a canted magnetic field. The python interface provides model fields which are constant on user defined subsets of WFs with the definition

$$H_B^w = \sum \langle \beta \Sigma w | B_i | P_i \rangle P_i$$ \hspace{1cm} (67)

where the diagonal of matrix $P_i$ is one for the targeted WFs and zero otherwise and $B_i$ is a constant vector.

The Wannier functions can also be used in the dHvA-package of FPLO instead of the full FPLO data [29]. Although the dHvA-package already uses an adaptive algorithm to sample the Fermi surface the use of a Wannier model can considerably speed of the process.

V. PERFORMANCE AND COMPARISON

A. Numerical performance

For orientation Table I shows the timing of various stages of the calculations of Sec. IV. We recorded the times for the complete self consistent calculation, for the single loop, which dumps the raw data needed for the WFs, for the actual Wannier function creation run, for the calculation of the WFs on a real space grid and for post processing, where it was used. All calculations are done on a single core.

|                | SCF | WF | WF on grid | post processing |
|----------------|-----|----|------------|----------------|
| CaCuO$_2$     | 53  | 8+2| 9          | 198            |
| Fe (3d4s4p)   | 39  | 11+10 | 76 | 32            |
| FeAl          | 74  | 23+77 | 248 | 14            |
| HgS           | 46  | 18+25 | 142 | 14            |
| MgB$_2$       | 13  | 3+2 | 9          |                |
| H$_2$O        | 2.9 | 0.2+0.2 | 1.3 |                |

B. Comparison with Wannier90

At present, the most popular code in the community is Wannier90 [3, 30, 31]. It supports interfaces with widely used band-structure codes has a growing number of postprocessing options [3]. It is therefore reasonable to use Wannier90 for benchmarking our FPLO results. To this end, we use band-structure tools from the Quantum ESPRESSO package version 6.7 [32] and the wannierization software provided in Wannier90 version 3.1.0. We use serial (single-core) executables compiled using the Intel’s Math Kernel Library (MKL) version 2020.4.304. In contrast to the full-potential code FPLO, Quantum ESPRESSO does not calculate the potential of core electrons explicitly, it is adopted from external pseudopotential files. Here, we used ultra-soft LDA (Perdew-Wang 91 [33]) Rappé-Rabe-Kaxiras-Joannopoulos (RRKJ) pseudopotentials [34] from psi4 version 1.0.0 [35], except for bcc-Fe calculations for which projector-augmented-wave pseudopotentials [36] were employed.

We start with the one-orbital model for CaCuO$_2$. As these calculations do not include the spin-orbit coupling, we can use symmetry-adapted Wannier functions [4] implemented in Wannier90 [3]. The agreement with the
FIG. 10. Left: LDA band structure of CaCuO₂ computed with Quantum ESPRESSO and Wannier fit for a one-orbital model performed using symmetry-adapted scheme implemented in Wannier90. Right: maximal contributions to the WF in logarithmic scale as a function of WF-orbital distance.

FIG. 11. Top: Wannier fit of the LDA band structure of bcc Fe computed with Quantum ESPRESSO. The Wannier model is constructed by projections onto 3d states. Right panel shows maximal contributions to the WF in logarithmic scale as a function of WF-orbital distance. Only the orbital-diagonal contributions are shown. Bottom: same for the model comprising 3d, 4s, and 4p states.

GGA band and the localization of the resulting Wannier functions (Fig. 10) are comparable to that in FPLO (Fig. 2).

Next, we consider full-relativistic calculations of bcc Fe. Again, we distinguish two models: a ten-bands model which includes only the 3d states, and a 18-bands model, which additionally accounts for 4s and 4p contributions. The former provides a generally good description of the LDA band structure, but deviations can be seen with a naked eye (Fig. 11, top left). Inclusion of the 4s and 4p states readily yields an excellent description of the entire valence band and the low-energy region of the polarization band (Fig. 11, bottom left). Also here the degree of localization is comparable with respective models calculated using FPLO. Berry curvature calculations (Fig. 12) are in excellent quantitative agreement with the FPLO results (Fig. 5).

Berry curvature of FeAl (Fig. 13, left) generally agrees with the FPLO result (Fig. 6, left), the discrepancies likely stem from the RRKJ pseudopotential for Fe. For HgS, the calculated Berry curvature does not vanish along the Γ-X line (Fig. 13, right), in contrast with the FPLO result (Fig. 6, right). This discrepancy is however quite small and must be due to either symmetry violation (due to full relativistic mode) and or numerical artefacts, since we argued in Sec. IV D that the total contribution must vanish along this line.

C. Comparison with previous reports

We performed calculations of the anomalous Hall effect, the anomalous Nernst effect, and the spin Hall effect based on the symmetry conserving Wannier functions implemented in FPLO. Our results for the anomalous Hall conductivity in Heusler compounds Co₂FeX (X=Al, Ga, Si, and Ge) are around 60, 210, 260, and 64 S/cm, respectively, by choosing the magnetic moment along the z direction. We obtain good agreement with the previous ab – initio report [37]. In addition to calculations, different groups also performed experimental Hall measurement for Co₂FeAl, Co₂FeGa, and Co₂FeSi. The calculated results fit well with the experimental measurements in Co₂FeGa and Co₂FeGe [38, 39]. Though our results and previous ab – initio reports give similar anomalous Hall conductivity for Co₂FeAl, it is much smaller than the value from Hall measurements [37], which might be due to a large extrinsic scattering contribution in the experimental setup.

For the anomalous Hall effect in typical transition elements such as Fe, Co, and Ni, our calculations show good
agreement with both experimental measurements and previous \textit{ab initio} calculations \cite{40–42}. We also made comparisons with some typical magnetic topological materials from recent reports, such as Fe$_3$Sn$_2$ which features a massive Dirac cone \cite{43, 44}, the first experimentally verified magnetic Weyl semimetal Co$_3$Sn$_2$S$_2$ \cite{45}, and the magnetic nodal line semimetals Rh$_2$MnGa, Rh$_2$MnAl, and Fe$_3$GeTe$_2$ \cite{46, 47}. We find good agreements for all of these magnetic topological systems, see Tables II.

Recently, starting with high throughput calculations, strong anomalous Nernst effects were observed in iron-based ferromagnets Fe$_3$X (X=Pt, Ga, Al, and Si) and Fe$_4$N \cite{49}. From Table III, one can see the good agreement between our calculations and recent reports. We also find good agreements for the anomalous Nernst conductivity in typical magnetic topological materials Co$_2$MnGa \cite{50}, Co$_3$Sn$_2$S$_2$ \cite{51}, and Fe$_3$GeTe$_2$ \cite{52}. All of them show the reliability of the Wannier functions derived from FPLO.

Another widely studied transport property in the linear response regime is the spin Hall effect (SHE). We made a systematic comparison between reported results and that obtained from FPLO. We find that the results for cubic (bcc and fcc) transition metals from our calculations are in good agreement with previous \textit{ab initio} calculations, see Tables IV,V,VI,VII, VIII,IX,X, We also tried to compare our theoretical results to the experimental reports. However, the experimental values from different reports differ a lot, due to details of experimental setup and extrinsic contributions.

As for the hexagonal transition metals, there are some differences between our calculations and that from previous reports \cite{76} (Table XI,XII), but overall, the two calculations are in good agreement. The differences might be due to the different sets of coordinates for the hexagonal lattice vectors. So far, most of the experiments focused on cubic transition metals and some compounds, whereas, to the best of our knowledge, there are almost no experimental reports for hexagonal transition metals. In addition to the transition metals, we also made a comparison for other compounds with available experimental reports and \textit{ab initio} calculations, see Table XIII, and here good agreements can also be found.

### D. Berry curvature dipole

We also calculated the Berry curvature dipole for the type-I Weyl semimetal TaAs and the type-II Weyl semimetal TaIrTe$_4$. The crystal structure of TaAs and TaIrTe$_4$ belong to point group $4mm$ and $mm2$, respectively. According to the symmetry analysis for these two point groups, TaAs has only one independent component with $D_{xy}=-D_{yx}$, and TaIrTe$_4$ has two independent components of $D_{xy}$ and $D_{yx}$. Our calculations fulfill these symmetry restrictions. The results reach convergence at a dense k-grid of $720^3$ and $480^3$ for TaAs and TaIrTe$_4$, respectively, see Fig. 14. The Berry curvature dipole has a value of about 0.37 for the $xy$ component of TaAs, in good agreement with Ref. 83. The Berry curvature dipole in TaIrTe$_4$ can reach up to around -0.19 and -0.50 for the $xy$ and $yx$ components, respectively.
TABLE II. Anomalous Hall Effect.

| Compound | Theory (S/cm) | experiment (S/cm) | this work (S/cm) |
|----------|---------------|-------------------|------------------|
| Co$_2$FeAl | 39 [37]       | 320-360 [48]      | 60               |
| Co$_2$FeGa | 181 [37]      | 50-350 [38]       | 210              |
| Co$_2$FeSi | 189 [37]      | 163, 400-600 [38, 39] | 260 |
| Co$_2$FeGe | 119 [37]      |                   | 64               |
| Fe$_3$Sn$_2$ |             | 200-400 [43, 44]  | 590              |
| Co$_3$Sn$_2$S$_2$ |           | 1100 [45]        | 1130             |
| Co$_2$MnAl | 1631 [46]     |                   | 1520             |
| Rh$_2$MnGa | 1860 [46]     |                   | 1760             |
| Rh$_2$MnAl | 1720 [46]     |                   | 1640             |
| Fe     | 1000 [40]     |                   | 840              |
| Fe$_3$GeTe$_2$ |         | 500 [47]         | 590              |
| Ni     | 2200 [41]     |                   | 2300             |
| Co (hcp) | 470 [41]      |                   | 480              |
| Co (fcc) | 250 [42]      |                   | 270              |

TABLE III. Anomalous Nernst Effect. maximum value with $T < 500K$ for Fe$_3$Pt, Fe$_3$Ga, Fe$_3$Al, Fe$_3$Si and Fe$_4$N.

| Compound | Theory (A/Km) | experiment (A/Km) | this work |
|----------|---------------|-------------------|-----------|
| Fe$_3$Pt | 6.2 [49]      |                   | 4.0       |
| Fe$_3$Ga | 3.0 [49]      |                   | 3.0       |
| Fe$_3$Al | 2.7 [49]      |                   | 3.0       |
| Fe$_3$Si | 2.5 [49]      |                   | 2.7       |
| Fe$_4$N  | 2.4 [49]      |                   | 1.1       |
| Co$_2$MnGa | 6.2 (300K) [50] | 4.4 ($E_F = E_0 + 0.06$ eV) | |
| Co$_3$Sn$_2$S$_2$ | 3 (80K) [51] | 2.6 |
| Fe$_3$GeTe$_2$ | 0.3 (150 K) [52] | 0.38 |
| TABLE IV. Pt, SHC in the unit of \((h/e)(S/cm)\) |
|------------------------------------------|
| Pt                                      |
| \(\sigma_{xy} \)                       |
| This work                                | 2260 |
| Ref.(ab-initio)                          | 2200 [53] |
| Ref.(ab-initio)                          | 2281 [54] |
| Ref.(ab-initio)                          | \(\sim 2200\) [55] |
| Ref.(ab-initio)                          | \(\sim 2500\) [56] |
| Ref.(Exp., \(T=10K\))                   | 1700\(\pm\)400 [57] |
| Ref.(Exp., room temperature)             | \(\sim 5100\) [58] |
| Ref.(Exp., room temperature)             | 310\(\pm\)50 [59] |
| Ref.(Exp., room temperature)             | 870\(\pm\)120 [60] |
| Ref.(Exp., room temperature)             | 1750 [61] |
| Ref.(Exp., room temperature)             | 1900 [62] |

| TABLE V. fcc Au, SHC is in the unit of \((h/e)(S/cm)\) |
|------------------------------------------------------|
| Au                                                   |
| \(\sigma_{xy} \)                                    |
| This work                                            | 377  |
| Ref.(ab-initio)                                      | 400 [63] |
| Ref.(ab-initio)                                      | 470 [56] |
| Ref.(ab-initio)                                      | 350 [64] |
| Ref.(Exp., \(T=4K\))                                | 11100 [65] |
| Ref.(Exp., room temperature)                         | 880\(\pm\)80 [59] |
| Ref.(Exp., room temperature)                         | 500\(\pm\)100 [67] |
| Ref.(Exp., room temperature)                         | 42000 [68] |

| TABLE VI. fcc Pd, SHC in the unit of \((h/e)(S/cm)\) |
|------------------------------------------------------|
| Pd                                                   |
| \(\sigma_{xy} \)                                    |
| This work                                            | 1180 |
| Ref.(ab-initio)                                      | 1200 [63] |
| Ref.(Exp., room temperature)                         | 350 [69] |
| Ref.(Exp., \(T=10K\))                               | 270\(\pm\)90 [57] |
| Ref.(Exp., room temperature)                         | 260\(\pm\)40 [59] |
| Ref.(Exp., room temperature)                         | 300\(\pm\)70 [71] |
| Ref.(Exp., room temperature)                         | 290\(\pm\)50 [67] |

| TABLE VII. bcc Ta, SHC in the unit of \((h/e)(S/cm)\) |
|------------------------------------------------------|
| Ta                                                   |
| \(\sigma_{xy} \)                                    |
| This work                                            | -133 |
| Ref.(ab-initio)                                      | -142 [54] |
| Ref.(Exp., \(T=10K\))                               | 111\(\pm\)3 [59] |
| Ref.(Exp., room temperature)                         | -630 [72] |
| Ref.(Exp., room temperature)                         | -160\(\pm\)120 [73] |

| TABLE VIII. bcc Nb, SHC in the unit of \((h/e)(S/cm)\) |
|-------------------------------------------------------|
| Nb                                                   |
| \(\sigma_{xy} \)                                    |
| This work                                            | -94  |
| Ref.(Exp., \(T=10K\))                               | 100\(\pm\)20 [57] |

| TABLE IX. bcc W, SHC in the unit of \((h/e)(S/cm)\) |
|-----------------------------------------------------|
| W                                                    |
| \(\sigma_{xy} \)                                    |
| This work                                            | -819 |
| Ref.(ab-initio)                                      | -785 [74] |
| Ref.(Exp., room temperature)                         | -1270\(\pm\)230 [75] |

| TABLE X. bcc Mo, SHC in the unit of \((h/e)(S/cm)\) |
|-----------------------------------------------------|
| Mo                                                   |
| \(\sigma_{xy} \)                                    |
| This work                                            | -276 |
| Ref.(Exp., \(K=10T\))                               | -230\(\pm\)50 [57] |
| Ref.(Exp., room temperature)                         | -23\(\pm\)5 [59] |

| TABLE XI. Hexagonal transition metals from [76]. The SHC is in the unit of \((h/e)(S/cm)\) |
|---------------------------------------------------------------|
| Sc                                                           |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | -40  | 7   | 6  |
| Ref.(ab-initio)                                              | \(\sim 50\) | \(\sim -10\) | -  |
| Ti                                                          |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | -2   | -23 | -19 |
| Ref.(ab-initio)                                              | \(\sim 5\) | \(\sim -15\) | -  |
| Zn                                                          |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | -70  | -6  | -6  |
| Ref.(ab-initio)                                              | \(\sim -130\) | \(\sim -20\) | -  |
| Y                                                           |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | 109  | 77  | 68  |
| Ref.(ab-initio)                                              | \(\sim 140\) | \(\sim 40\) | -  |
| Zr                                                          |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | -242 | -48 | -19 |
| Ref.(ab-initio)                                              | \(\sim 300\) | \(\sim -60\) | -  |
| Tc                                                          |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | -160 | -18 | -97 |
| Ref.(ab-initio)                                              | \(\sim 200\) | \(\sim -140\) | -  |
| Ru                                                          |
| \(\sigma_{xy} \) \(\sigma_{yz} \) \(\sigma_{xz} \)          |
| This work                                                    | 90  | 190 | 163 |
| Ref.(ab-initio)                                              | \(\sim 10\) | \(\sim 90\) | -  |
### TABLE XII. Continuation of Tab. XI. The SHC is in the unit of \((\hbar/e)(S/cm)\)

| Element | \(\sigma_{xy}\) | \(\sigma_{yz}\) | \(\sigma_{zx}\) |
|---------|-----------------|-----------------|-----------------|
| La      | 290             | 316             | 308             |
|         | Ref.(ab-initio) | ~150            | ~300            |
| Hf      | -375            | -53             | 112             |
|         | Ref.(ab-initio) | ~800            | ~600            |
| Re      | -325            | -441            | -519            |
|         | Ref.(ab-initio) | ~500            | ~700            |
| Cd      | -93             | 2               | -5              |
|         | Ref.(ab-initio) | ~30             | ~20             |
| Os      | -68             | -162            | -72             |
|         | Ref.(ab-initio) | ~250            | ~300            |

### TABLE XIII. Other compounds, SHC in the unit of \((\hbar/e)(S/cm)\)

| Compound | Phase | \(\sigma_{xy}\) | \(\sigma_{yz}\) | \(\sigma_{zx}\) |
|----------|-------|-----------------|-----------------|-----------------|
| PtO\(_4\) | bcc   | 1420            | 215             |                 |
|         | Cubic |                 |                 |                 |
|         | This work | 1420            | 215             |                 |
|         | Ref.(ab-initio) | [77]            |                 |                 |
| PtTe\(_2\) |       |                 |                 |                 |
|         | This work(largest component \(\sigma_{yz}\)) | 122             |                 |                 |
|         | Ref.(Exp., room temperature) [78] | 100-800          |                 |                 |
| NbSe\(_2\) |       |                 |                 |                 |
|         | This work(largest component \(\sigma_{yz}\)) | 188             |                 |                 |
|         | Ref.(Exp., room temperature) [79] | ~150             |                 |                 |
| WTe\(_2\) |       |                 |                 |                 |
|         | This work | 34              | 85              |                 |
|         | Ref.(ab-initio) [80] | ~20            | ~80             |                 |
|         | Ref.(Exp., room temperature) [80] | 14-96           |                 |                 |
| Bi\(_2\)Se\(_3\) |       |                 |                 |                 |
|         | This work | 105             | 104             | 96              |
|         | Ref.(Exp., room temperature) [81] | ~80             |                 |                 |
|         | Ref.(Exp., room temperature) [82] | 500-1000        |                 |                 |
VI. SUMMARY AND CONCLUSIONS

We thoroughly documented the underlying methods of the FPLO package for the construction of symmetry conserving maximally projected Wannier functions.

The method does not enforce maximum localization but rather results in Wannier functions with (near) maximum projection onto FPLO orbitals or linear combinations thereof, yet yields a very high degree of localization due to the nature of the orbitals while retaining the symmetry properties of the chosen projectors. The simplicity of this algorithm leads to numerical efficiency and a high level of accuracy, which is needed in the context of topological properties, where symmetry conservation is of the essence, especially when taking derivatives (e.g. for the Berry curvature dipole).

For a complete treatment of topological properties the full Berry connection/curvature is desirable. To this end we discussed the position operator matrix elements as they have to be treated in a local orbital context as well as their symmetry properties in the context of possible approximations of this operator. The latter gives valuable insights for the application of such approximations in tight binding models with implicit (unknown) Wannier basis.

The method has been applied to a set of compounds to illustrate various applicational aspects as well as to demonstrate the general quality of the resulting Wannier models and the available post processing tools.

We gave an idea of the efficiency of our method and repeated some of the calculations using the Wannier90 package to convince the reader that our method indeed yields comparable results. We gave an extensive list of results for the anomalous Hall effect, the anomalous Nernst effect, and the spin Hall effect compared to results of other publications.

As a technical reference an extended appendix documents the FPLO basis orbitals and their transformation properties from which the symmetry properties of the Wannier functions result.

In summary, we believe to have demonstrated that maximally projected symmetry conserving Wannier functions based on optimized local orbital methods have similar localization and flexibility as maximally localized Wannier functions, however with minimal computational cost and complexity and explicit symmetry properties.

VII. ACKNOWLEDGMENTS

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Appendix A: Local basis states

1. FPLO basis

An FPLO basis orbital \( \Phi_{Rsv}(r) \) is a solution of a Schrödinger/Dirac equation in an atom centered suitably modified potential with compact support. The boundary conditions are chosen such that there is no leakage of the orbital wave function at the compact support radius (this is not possible exactly in the Dirac case but practically the leakage can be made tiny and irrelevant). The orbitals sit in the unit cell at lattice vector \( R \) at site \( s \) and have quantum numbers \( \nu \). To be precise \( \Phi_{Rsv}(r) = \Phi_{sv}(r - R - s) \) where \( \Phi_{sv}(r) \) is a function of atom site index \( s \) and qns. \( \nu \).

In non- and scalar-relativistic mode the orbitals have the shape \( \Phi_{snlms} = \varphi_{snl}(r) Y_{lm}(\hat{r}) \chi_\sigma \) where \( n \) is the main quantum number, \( Y_{lm} \) is a real spherical harmonic with angular momentum qns. \( l \) and \( m \) and \( \chi_\sigma \) is a spin \( \frac{1}{2} \) basis spinor defined via \( \chi_\sigma = \chi_\sigma^\dagger = \sigma^\dagger \chi_\sigma \) (\( \sigma = \pm 1 \)). The radial basis functions are not spin dependent which is compensated by the variational freedom of the chosen basis set which contains polarization orbitals additionally to the chemical valence (and semi-core) orbitals.

In full relativistic mode the orbitals are solutions to a 4-component Dirac equation and are four spinors

\[
\Phi_{snlj\mu} = \begin{pmatrix} g_{snlj}(r) \chi_{\kappa\mu} \\ if_{snlj}(r) \chi_{-\kappa\mu} \end{pmatrix}
\]  
(A1)

where \( g_{snlj} \) and one small component \( f_{snlj} \) radial function per \( j \)-shell and spherical spinors \( \chi_{\kappa\mu} \), where \( \kappa = (2j + 1) (l - j) \) are the eigenvalues of the spin orbit operator \( \vec{\kappa} = \frac{1}{2} \vec{\sigma} \vec{L} \) with \( \vec{L} \) the angular momentum operator and \( \kappa_{j1 - j\frac{1}{2} - l\frac{1}{2}} = l \) and \( \kappa_{j1 + j\frac{1}{2} + l\frac{1}{2}} = -l \frac{1}{2} + 1 \) and \( \mu = -j, -j + 1, \ldots, j \) is the \( \vec{J}_z \) eigenvalue.

The orbitals can be made orthonormal at each site. However, off-site orthonormality is hard to achieve, since this would mean to explicitly construct Wannier functions from the start. With a proper choice of orbitals a well conditioned overlap matrix

\[
S_{R's'v'} = \langle \Phi_{R's'v'} | \Phi_{Rsv} \rangle
\]

with one large component \( g_{snlj} \) and one small component \( f_{snlj} \) can be achieved. Lattice translations

\[
\hat{T}_{R}\Phi_{Rsv}(r) = \Phi_{Rsv}(r - R') = \Phi_{sv}(r - R - s - R') = \Phi_{R'+R'sv}(r)
\]  
(A2)
yield translation invariance according to
\[ S_{R's'\nu',Rs\nu} = \langle \hat{T}_{-R'} \Phi_{R's'\nu'} | \hat{T}_{-R} \Phi_{Rs\nu} \rangle = \langle \Phi_{0s'\nu'} | \Phi_{R-R's\nu} \rangle = S_{0s'\nu',R-R's\nu} \]  
\[ (A3) \]

2. Bloch sums

In order to form extended basis states Bloch sums of local orbitals are defined as
\[ \phi^k_{as}(r) = \frac{1}{\sqrt{N}} \sum_R e^{i k (R + \lambda s)} \Phi_{Rs\nu}(r) \]  
\[ (A4) \]
where \( \lambda = 0, 1 \) (\( \bar{\lambda} = 1 - \lambda \)) picks a particular phase gauge as discussed in the main text. The normalization contains the number of unit cells \( N \) in the Born-von-Kármán (BvK) torus, which is there for formal correctness but never actually appears in any coded formulas except for the normalization of the Fourier back transform. The Fourier transformed overlap matrix is defined as
\[ \Phi_{Rsk}(r) \]
\[ (B1) \]

The exponential contains the difference vector between the two orbital positions if \( \bar{\lambda} = 1 \), which is why we call it relative gauge.

The overlap of a Bloch sum with a single basis orbital \( \Phi_{Rs\nu} \) reads
\[ \langle \Phi^k_{as}(r') | \Phi_{Rs\nu} \rangle = \frac{1}{\sqrt{N}} \sum_{R'} e^{-i k (R' + \lambda s')} \langle \Phi_{R's'\nu'} | \Phi_{Rs\nu} \rangle = \frac{1}{\sqrt{N}} \sum_{R'} e^{-i k (R' + \lambda s')} S_{R's'\nu',Rs\nu} = \frac{1}{\sqrt{N}} \sum_{R'} \sum_{R} e^{i k (R + \lambda s')} S_{0s'\nu',R-R's\nu} = \frac{1}{\sqrt{N}} \sum_{R} e^{i k (R + \lambda (s - s'))} S_{0s'\nu',Rs\nu} \]  
\[ (A5) \]
which motivates the normalization choice in Eq. (A4). The exponential contains the difference vector between the two orbital positions if \( \bar{\lambda} = 1 \), which is why we call it relative gauge.

The overlap of two Bloch sums with single basis orbitals \( \Phi_{Rs\nu} \) and \( \Phi_{Rs'\nu'} \) reads
\[ \langle \Phi^k_{as}(r') | \Phi_{Rs\nu} \rangle = \frac{1}{\sqrt{N}} \sum_{R'} e^{-i k (R' + \lambda s')} \langle \Phi_{R's'\nu'} | \Phi_{Rs\nu} \rangle = \frac{1}{\sqrt{N}} \sum_{R'} e^{-i k (R' + \lambda s')} S_{R's'\nu',Rs\nu} = \frac{1}{\sqrt{N}} \sum_{R'} \sum_{R} e^{-i k (R - R' + \lambda s')} S_{0s'\nu',R-R's\nu} = \frac{1}{\sqrt{N}} \sum_{R'} \sum_{R} e^{i k (R' + \lambda (s - s'))} S_{0s'\nu',R-R's\nu} e^{-i k (R - R')} = \frac{1}{\sqrt{N}} \sum_{R'} S_{k}^{k}_{s's'\nu',Rs\nu} e^{-i k (R + \lambda s)} \]  
\[ (A6) \]
using \( R' \rightarrow -R' + R \) and Eqs. (A3,A5). Using Eq. (A4) and Eq. (A5) the overlap of two Bloch functions yields the overlap matrix
\[ \langle \Phi^k_{as}(r') | \Phi^k_{as}(r') \rangle = \frac{1}{N} \sum_{R} S_{k}^{k}_{s's'\nu',Rs\nu} = S_{k}^{k}_{s's'\nu',Rs\nu} \]  
\[ (A7) \]

We introduce local linear combinations of orbitals (MOs) for projection purposes
\[ \phi_{Rci} = \sum_{Rs} \Phi_{Rs\nu} U_{Rs\nu,Rci} \]  
\[ (A7) \]
which shall be translation invariant in the same way as the orbitals Eq. (A2), i.e. \( U_{Rs\nu,Rci} = U_{0s\nu',R-R'ci} \) and have Bloch sums
\[ \phi^k_{ci} = \frac{1}{\sqrt{N}} \sum_{R} e^{i k (R + \lambda c)} \phi_{Rci} = \frac{1}{\sqrt{N}} \sum_{R} e^{i k (R + \lambda c - \lambda s)} U^k_{s\nu',ci} \]  
\[ (A8) \]

To calculate the overlap between a MO and an orbital Bloch sum Eq. (A6) gets then modified according to
\[ \langle \Phi^k_{as} | \phi_{Rci} \rangle = \frac{1}{\sqrt{N}} \sum_{R} (S^k_{s's'} U^k_{s\nu',ci} e^{-i k (R + \lambda c)}) \]  
\[ (A9) \]
\[ \text{with} \]
\[ \sum_{k} f_k = 1 \]

If the full mesh is used \( f_k = N^{-1} \), but if only irreducible \( k \)-points are used \( f_k \) is corrected for the multiplicity of the irreducible point. Combining Eqs. (A5,A9) one gets
\[ \sum_{k} f_k e^{i k (R - R')} = \delta_{R,R'} \]
which is only true modulo a lattice vector which describes the BvK periodicity. The back transformation of a Bloch sum itself with correct normalization reads
\[ \Phi_{Rs} = \frac{1}{\sqrt{N}} \sum_{k} e^{-i k (R + \lambda s)} \phi^k_{s} \]
A translation invariant operator in Bloch sum basis (as the overlap) contains two Bloch sums and is diagonal in \( k \)-space. A full Fourier back transformation of both sides of the matrix then reduces to Eq. (A9) with \( \frac{1}{N} \) normalization instead of \( \frac{1}{\sqrt{N}} \).

Appendix B: Symmetry

1. Local basis symmetry

The complex spherical harmonics are defined as
\[ Y_{lm}(\varphi, \theta) = (-1)^{m+l} \sqrt{\frac{2l+1}{4\pi} \frac{(l-|m|)!}{(l+|m|)!}} P^{|m|}_l (\cos \theta) e^{im\varphi} \]  
\[ (B1) \]
with the associated Legendre functions for positive \( m \) as a function of \( z = \cos(\theta) \)

\[
P_l^m(z) = (1 - z^2)^{\frac{m}{2}} \frac{d^m}{dz^m} P_l(z)
\]

\[
P_l(z) = P_l^0(z) = \frac{1}{2^l l!} \frac{d^l}{dz^l} (z^2 - 1)^l
\]

From this the real harmonics are obtained via

\[
Y_{lm}(\theta, \varphi) = \sum_{m'} Y_{lm'}(\theta, \varphi) U_{m'm}^*
\]

\[
= \begin{cases} 
\frac{1}{\sqrt{2}} \left( (-1)^m Y_{l|m|} + Y_{l|-m|} \right) & m > 0 \\
\frac{1}{\sqrt{2}} \left( (-1)^{|m|} Y_{l|m|} - Y_{l|-m|} \right) & m < 0 
\end{cases}
\]

The spherical spinors as introduced after Eq. (A1) are defined as

\[
\chi_{\kappa\mu} = \sum_{s = \pm 1} \chi_s Y_{l|m|} \xi^s_{\kappa\mu}
\]

with basis spinors \( \chi_s \), complex Harmonics Eq. (B1) and Clebsch-Gordon coefficients

\[
\xi^s_{\kappa\mu} = -\left( \frac{s (\kappa + |\kappa|) + (\kappa - |\kappa|)}{2 |\kappa|} \right) \sqrt{\frac{1}{2} - s} \frac{\mu}{(2\kappa + 1)}
\]

which can be written as

\[
\chi_{\kappa\mu} = \sum_{s = \pm 1, m = -l_s}^{l_s} \chi_s Y_{l|m|} T_{ms,k\mu}^{l_s}
\]

\[
\chi_{\kappa\mu} = \sum_{s = \pm 1, m = -l_s}^{l_s} \chi_s Y_{l|m|} (UT)_{ms,k\mu}^{l_s}
\]

\[
T_{ms,k\mu}^{l_s} = \delta_{m,-\mu} \xi^{s}_{\kappa\mu}
\]

with \( l_s = \frac{|2\kappa + 1| - 1}{2} \) where \( T^l \) can be arranged in a unitary matrix for each \( l \)-shell.

Inversion \( \hat{f} \mathbf{r} = -\mathbf{r} \) at the origin of the harmonics with \( \hat{f} \mathbf{r} = f(-\mathbf{r}) \) acts like

\[
\hat{f} Y_{lm} = Y_{lm} (-1)^l
\]

\[
\hat{f} Y_{lm} = Y_{lm} (-1)^l
\]

\[
\hat{f} \chi_{\kappa\mu} = \chi_{\kappa\mu} (-1)^l
\]

\[
\hat{f} \chi_s = \chi_s
\]

A proper rotation \( \hat{\alpha} \) with \( \hat{\alpha} f(\mathbf{r}) = f(\hat{\alpha}^{-1} \mathbf{r}) \) parametrized by the Euler angles is represented by the Wigner-\( D \) functions in the \( l \) or \( ij \) basis which without further details reads

\[
\hat{\alpha} Y_{lm} = \sum_{m'=l} Y_{lm'} D_{lm'm}^l(\alpha)
\]

\[
\hat{\alpha} Y_{lm} = \sum_{m'=l} Y_{lm'} D_{lm'm}^l(\alpha)
\]

\[
\hat{\alpha} \chi_{\kappa\mu} = \sum_{\sigma' = \pm 1} \chi_{\kappa'\mu'} D_{\kappa\sigma'\kappa'}(\alpha)
\]

where \( j = \frac{1}{2} \) and \( \hat{\alpha} \) transforms as non-relativistic complex harmonics \( \mathcal{Y} \).

From Eqs. (B3,B4,B5)

\[
D Z D^j T^l = T^l D^j
\]

which shows that the totality of all four spinors of an \( l \)-shell \( \Phi^l = \left( \Phi^l_{j=-\frac{1}{2}} \Phi^l_{j=+\frac{1}{2}} \right) \) multiplied by \( T^l \) transforms as non-relativistic complex harmonics \( \mathcal{Y} \) (or as real harmonics \( Y \) if \( (UT^l)^+ \) is used).

Finally, the time reversal operator is defined as \( \hat{T} \mathbf{r} = -i\mathbf{r} \mathbf{K}_0 \), where \( \mathbf{K}_0 \) is the \( y \)-component of the Pauli matrix vector and \( \mathbf{K}_0 \) is complex conjugation, which gives the general action \( \hat{\tau} f_i = \sum_{i'} f_i D_{ii'}^i \mathbf{K}_0 \) where \( D^\theta \) can be read off the following relations

\[
\hat{\tau} Y_{lm} = Y_{l-m} (-1)^m \theta
\]

\[
\hat{\tau} Y_{lm} = Y_{l-m} \theta
\]

\[
\hat{\tau} \chi_{\kappa\mu} = \chi_{\kappa\mu} (-1)^{l+j-m} \mathbf{K}_0
\]

\[
\hat{\tau} \chi_s = \chi_{-\sigma} \mathbf{K}_0
\]

\[
2. \text{ Space group symmetry/Time reversal}
\]

The space group contains the set of operations

\[
T_{RG} = T_R \{ \alpha \mid \tau_\alpha \} = \{ \alpha \mid \tau_\alpha + \mathbf{R} \}
\]

where \( T_R \) is a lattice translation and \( g = \{ \alpha \mid \tau_\alpha \} \) is the Seitz symbol containing a proper/improper rotation \( \alpha \) and a non-lattice translation \( \tau_\alpha \). The latter depends on the origin choice and a particular pick among all the vectors \( \tau_\alpha + \mathbf{R} \). A real space vector transforms according to

\[
T_{RG} \mathbf{r} = \{ \alpha \mid \tau + \mathbf{R} \} \mathbf{r} = \mathbf{g} + \mathbf{R} = \alpha \mathbf{r} + \tau + \mathbf{R}
\]
This is especially true for a lattice site \( s + \mathbf{R} \) for which one gets
\[
T_R g (\mathbf{R} + s) = \alpha \mathbf{R} + g s + \mathbf{R} = \alpha \mathbf{R} + \alpha s + \tau + \mathbf{R}
\]
The inverse is given by
\[
\{ \alpha \mid \mathbf{R} + \mathbf{R} \}^{-1} = \{ \alpha^{-1} | - \alpha^{-1} (\tau + \mathbf{R}) \}
\]

When setting up a structure a certain set of sites will be generated. They do not need to lie in the first unit cell. The set of transformed sites \( gs \) will in general not be identical to this original set. A simple lattice translation
\[
gs = s_g + R_{g,s}
\]
will map the transformed set back onto the original one. If this is done consistently, the site numbers \( s, s_g \) can be used as indices to orbitals and Bloch sums.

The vector of all orbitals (and of Wannier functions) at a site transforms according to
\[
g \Phi_{Rs} (r) = g \Phi_s (r - \mathbf{R} - s)
\]
which transforms orbitals at site indexed by \( s \) into orbitals at site indexed by \( s_g \). For the FPLO basis these matrices are the same at each site and are obtained according to Sec. B 1. The last row, Eq. (B10b), is a useful short hand for Eq. (B14) in deriving symmetry properties since \( g (\mathbf{R} + s) \) is a complete set of lattice sites which can serve as summation variable.

In case of molecular orbitals as WF projectors the symmetry properties of the WFs is an implicit input in our method an they are required to transform akin to Eq. (B10b). The input matrix \( U \) in Eq. (A7) together with the transformation properties of the orbitals \( D^\Phi_s \) determine the transformation properties of \( \phi_c \) completely. In practice one chooses \( U \) in Eq. (A7). The code looks if application of symmetry replicates the set of Wannier functions and if the resulting matrices \( D^\Phi_c \) are unitary. If not the input is invalid. This requires the following relations
\[
g \phi_c = \sum_{Rs} g \Phi_{Rs} U_{Rs,0c}
\]
or
\[
U_{Rs,0c} = (D^\Phi_s (\alpha))^+ U_{g(Rs),gc} D^\Phi_c (\alpha)
\]
where again we need to replace \( g (\mathbf{R} s) \) by \( \alpha \mathbf{R} + R_{g,s}, s_g \) and \( gc \) by \( R_{g,c}, c_g \).

Additionally, useful operators need to be lattice periodic
\[
T_R \hat{B} (r) T_R^{-1} = \hat{B} (r - \mathbf{R}) = \hat{B} (r)
\]
and invariant under the space group
\[
g \hat{B} (r) g^{-1} = \hat{B} (g^{-1} r) = \hat{B} (r) D^B (\theta)
\]
where \( D^B (\theta) \) is the representation matrix of the operator. E.g. \( D^B (I_0) = \pm \) \( \alpha \) if \( \hat{B} \) is a polar \( (+ \alpha) \) or axial \( (- \alpha) \) vector and \( \alpha \) a proper rotation. This does however not apply to the position operator \( \hat{r} \), since it is not translational invariant. For all point group operations which leave the origin invariant \( \hat{r} \) holds, but for a general space group operation one gets \( g \hat{r} g^{-1} = \alpha^{-1} (\tau - \mathbf{R}) = (\mathbf{r} - \mathbf{R}) \).

If time reversal applies it yields
\[
\theta \hat{B} (r) \theta^{-1} = \hat{B} (r) D^B (\theta)
\]
where \( D^B (\theta) \) is e.g. +1 for the Hamiltonian and −1 for the spin and magnetic field. In full relativistic spin polarized mode products of time reversal with operations, which flip the magnetic field, are group elements. We apply the latter only to the subset of the actual space group, which preserves the magnetization axis, i.e. we do not implement full magnetic groups.

Using the shorthand Eq. (B10b) with its meaning Eq. (B10a) matrix elements transform as
\[
B_{Rs',Rs} = \langle \Phi_{Rs} | \hat{B} | \Phi_{Rs'} \rangle
\]
and
\[
\theta \Phi = \Phi D^\theta \theta_0
\]
\[
\Phi^+ \theta^{-1} = \theta_0 D^\theta + \Phi^+
\]
one gets the general behavior of matrix elements under time reversal
\[
B_{Rs',Rs} = \langle \Phi_{Rs'} | \theta^{-1} \hat{B} \theta^{-1} \Phi | \Phi_{Rs} \rangle
\]
\[
B_{Rs',Rs} = K_0 D^\theta + B_{Rs',Rs} D^\theta \theta_0 \theta_0
\]

With the general behavior (details in Eq. (B8))
\[
\theta \Phi = \Phi D^\theta \theta_0
\]
\[
\Phi^+ \theta^{-1} = \theta_0 D^\theta + \Phi^+
\]
one gets the general behavior of matrix elements under time reversal
\[
B_{Rs',Rs} = \langle \Phi_{Rs'} | \theta^{-1} \hat{B} \theta^{-1} \Phi | \Phi_{Rs} \rangle
\]
\[
B_{Rs',Rs} = K_0 D^\theta + B_{Rs',Rs} D^\theta \theta_0 \theta_0
\]

(B16)
Now, the transformation of Bloch sums of LOs/WFs is obtained from Eqs. (A4,B10a) where the use of back-folded sites \( s_g \) is of the essence

\[
g\Phi^k_s = \frac{1}{\sqrt{N}} \sum_R e^{ik(R+\lambda s)} g\Phi^k_{Rs} = \frac{1}{\sqrt{N}} \sum_R e^{ik(R+\lambda s)} \Phi_{\alpha R + R_g, s_R} D^\Phi_s
\]

\[
\Phi^k_s D^\Phi_s = \frac{1}{\sqrt{N}} \sum_R e^{i\alpha k(R+\lambda s)} \Phi_{\alpha R + R_g, s_R} D^\Phi_s = \frac{1}{\sqrt{N}} \sum_R e^{i\alpha k(R+\lambda s)} \Phi_{\alpha R + R_g, s_R} D^\Phi_s
\]

\[
\Phi^k_s D^\Phi_s = \Phi^k_s e^{i\alpha k(\lambda s - R_g, -s_R)}
\]

\[
= (\Phi^k D^\Phi)^s
\]

with

\[
D^\Phi_s = \delta_{s, s'} D^\Phi_s e^{-i\alpha k(\lambda (s_g - s_{g'}) + \lambda \tau)}
\]

from this one sees that the relative gauge \( \lambda = 0 \) leads to simpler expressions, since all the phase factors in Eqs. (B20) vanish.

Eqs. (A4,B15a) gives the time reversal of the orbital Bloch sums

\[
\theta \Phi^k_s = \theta \frac{1}{\sqrt{N}} \sum_R e^{ik(R+\lambda s)} \Phi^k_{Rs} = \frac{1}{\sqrt{N}} \sum_R e^{-ik(R+\lambda s)} \theta \Phi^k_{Rs}
\]

\[
= \frac{1}{\sqrt{N}} \sum_R e^{-ik(R+\lambda s)} \Phi^k_{Rs} D^\Phi^\theta_s K_0
\]

\[
= \Phi^k_s e^{-i\alpha k(\lambda s - R_g, -s_R)}
\]

and together with Eqs. (B15b,B13)

\[
\langle \Phi^k_s | \hat{B} | \Phi^k_s \rangle = \langle \Phi^k_s | \theta^{-1} \hat{B} \theta^{-1} \theta | \Phi^k_s \rangle
\]

\[
= \left( D^\Phi^\theta_s \langle \Phi^k_s | \hat{B} | \Phi^k_s \rangle \right)
\]

If the operation is a product \( \theta g \) this gets modified by carefully inserting Eq. (B20) for \( -k \) into Eq. (B22) which is not spelled out due to the unwieldy phase factors.

Essentially we let \( D^\Phi \rightarrow D^\Phi D^\theta \), \( B \rightarrow B D^\theta \) and \( \alpha k \rightarrow -\alpha k \) in Eq. (B20) and apply complex conjugation to the whole rhs.

In order to symmetrize the matrix elements when needed we make use of the fact that the sum of the rhs. of Eq. (B14) over all group operations divided by the number of group elements is a projector. So, for all pairs of lattice sites we perform this sum over all transformed pairs. In case if time reversal is contained in the operation the corresponding formulas Eq. (B16) or a mix of this with Eq. (B14) must be used.

On a final note, in relativistic case the ambiguity of choosing the rotation or rotation+2\( \pi \) as pointgroup part of \( g \) cancels out in all formulas which transform matrix elements, since they are bilinear in the orbitals.

3. Bloch theorem

Eq. (B17) can be specialized to pure translations by using \( \alpha = 1, \tau = R \) i.e. \( g = \{ E | R \} \) for which \( g s = s + R \) and \( D^s_s (\alpha) = 1 \) which gives the Bloch theorem

\[
T_R \Phi^k_s = \Phi^k_s e^{-ik R}
\]

which can be applied to matrix elements of translational invariant operators Eq. (B11) between Bloch sums Eq. (A4) of different \( k \)-vectors (the most general case) to get (dropping all non-essential indices)

\[
\langle \Phi^q | \hat{B} | \Phi^k \rangle = \langle T_R \Phi^q | T_R \hat{B} T_R^{-1} | T_R \Phi^k \rangle = \langle \Phi^q | \hat{B} | \Phi^k \rangle e^{-ik(q - q') R}
\]

For \( q \neq k \) in the first BZ the phase cannot become one and hence

\[
\langle \Phi^q | \hat{B} | \Phi^k \rangle = \delta_{q,k} \langle \Phi^q | \Phi^k \rangle
\]

For the position operator which fulfills

\[
T_R T_R^{-1} = r - R
\]

Eq. (B24) becomes

\[
\langle \Phi^q | r | \Phi^k \rangle = \langle \Phi^q | r | \Phi^k \rangle - \delta_{q,k} S^k R e^{-i(k-q) R}
\]

which specializes to

\[
\langle \Phi^k | r | \Phi^k \rangle = \langle \Phi^k | r | \Phi^k \rangle - S^k R
\]

for the \( k \)-diagonal terms, which shows that the limit \( q \rightarrow k \) is badly defined and that the operator is essentially \( k \)-non-diagonal.

The Berry operator Eq. (28) transforms like the position operator itself:

\[
T_R \beta_k T_R^{-1} = e^{ikr} e^{-ik R \partial_k e^{-ik R}} = e^{ikr} i \partial_k e^{-ik R} + e^{-ik R} (i \partial_k e^{ik R}) = \beta_k - R
\]
which then gives
\[ T_R | \beta_k \psi^k \rangle = | T_R \beta_k T_R^{-1} T_R \psi^k \rangle \]
\[ = (| \beta_k - R \rangle \Phi^{k \alpha \beta} R^\dagger) e^{-i k R} \]
\[ = | \beta_k \psi^k e^{-i k R} - | \psi^k \rangle e^{-i k R} \]

Applying Eq. (29a) to the first term of the rhs. yields
\[ T_R | \beta_k \psi^k \rangle = | \beta_k \psi^k \rangle e^{-i k R} \]
and hence Eqs. (B24,B25) apply and \( \beta_k \) is \( k \)-diagonal.

4. Berry connection/curvature

The generalization of the vector valued functions Eq. (38) is obtained by using Eq. (B9) and \( g f (r) = f \left( g^{-1} r \right) \) as in Eq. (B10) which results in
\[ (g^{-1} r - R - s) = (r - \alpha R - gs) \alpha \]
for the \( r \)-factor and hence in
\[ g_r \Phi_s = \Phi_s (r) D_s^\Phi \]
where the last \( \alpha \) acts on the vector structure.

The reduced position operator matrix elements Eq. (43) transform as Eq. (B14) with \( D^B \to \alpha \), i.e. as LO matrix elements in the orbital indices and as a polar vector in the position operator indices. Bunch sums of Eq. (B30) lead to the transformation of Eq. (42) according to
\[ \langle \Phi^k | (r \Phi)^k \rangle = D^{\Phi k} + \langle \Phi_a k | (r \Phi)^a \rangle D^{\Phi k} \]
with \( D^{\Phi k} \) from Eq. (B18).

Eqs. (41,B31) then lead to
\[ A^k_b - S^k_s D^{b k} A^k_s - S^a_s k D^{a k} = \]
\[ = A^k_b - S^k_s D^{b k} A^k_s - S^a_s k D^{a k} \]
where \( s \) is the diagonal matrix containing the site indices of the corresponding orbitals. By using Eq. (B18) and Eq. (B20) with \( B = 1 \) this can be transformed into
\[ A^k_b = D^{b k} A^k_s + S^a_s k (s - s_g) \alpha \]
(B33)

From this it is clear that for WF \( S^k_s = 1 \) approximation Eq. (46) fulfills the transformation law Eq. (B32) trivially while setting the basis connection to zero \( A^k_b = 0 \) will in general not. Acting with \( g \) on \( r \) in Eq. (28) and using \( \nabla_k = \nabla_a k \alpha \) we get
\[ g \beta_k g^{-1} = (\beta_k - \tau) \alpha \]
which allows to derive the general transformation properties of \( A^k_b \) directly when assuming a general law \( g \Phi^k = \Phi^a k \Phi^k D^{b k} \Phi^k \) with some representation matrices \( D^{b k} \).

\[ A^k_b = \langle \Phi^k | (r \Phi)^k \rangle \]
\[ = \langle \Phi^k | (r \Phi)^a \rangle D^{a k} \]
\[ = \langle \Phi^k | (r \Phi)^a \rangle D^{a k} + i \alpha \nabla_k D^{a k} \]
\[ - S^a_s k \tau \alpha \]
\[ = \langle \Phi^k | (r \Phi)^a \rangle D^{a k} + i \alpha \nabla_k D^{a k} \]
\[ - S^a_s k \tau \alpha \]
\[ = D^{b k} (A^k_b + S^a_s k i \nabla_k) D^{b k} - S^a_s \tau \alpha \]
\[ = D^{b k} (A^k_b + S^a_s k i \nabla_k) D^{b k} - S^a_s \tau \alpha \]

which specializes to Eq. (B33) by inserting the gradient of Eq. (B18), in other words the local basis connection transforms as a general Berry connection matrix would.

Now, we introduce the basis change \( \Psi^k = w^k C^k \), which leads to the Berry connection matrix Eq. (54a). Then we can show that if \( A^k_b \) transforms as Eq. (B34) with \( \Phi \to w \), also \( A^k_b \) (Eq. (54a)) transforms like Eq. (B34) with \( \Phi \to \Psi \). Using the abbreviations \( a^b k \to b \), \( b^a k \to a \) and \( C^b k \to C \), one gets the transformation properties of \( \Psi \), \( w \) and \( C \).

\[ g w = w^a D^a \]
\[ g \Psi = \Psi^a D^a \]
\[ D^w C = C^a D^a \]

\[ \langle S^a w S^a \rangle_C = \langle S^a w S^a \rangle_C + \langle S^a w \rangle C \tau \]
\[ = \langle S^a w S^a \rangle_C + \langle S^a w \rangle C \tau \]
\[ = \langle S^a w \rangle C \tau \]
\[ = \langle S^a w \rangle C \tau \]

which leads to
\[ A^b = \langle A^a_b \rangle_D + \langle S^a w S^a \rangle_C - S^a \tau \]
\[ \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]
\[ = \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]
\[ = \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]
\[ = \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]

\[ = \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]

\[ = \langle A^a_b \rangle_D + \langle S^a w \rangle C \tau \]

Eq. (B36) explicitly shows that the basis change again leads to the generic transformation law Eq. (B34). All we need to show now is that this law leads to the proper transformation behavior of the curvature.

We start with the cross-product term, which we write in it’s most general form (including the correction for non-orthonormal \( \Psi \)) using Eq. (B36)
\[ i A^0 \frac{1}{S^0} \times A_\Psi \]
\[ = (a - S^0 \tau a)^+ \frac{1}{S^0} \times (a - S^0 \tau a) \]
(B37)
which by insertion gives
\[
\nabla \times (\mathbf{A}_\Psi) = D^\Psi \frac{1}{S^\Psi} \nabla D^\Psi
\]
which shows that the $A \times A$-term of the non-Abelian Berry curvature alone transforms as a matrix in orbital indices and as pseudo vector in vector indices. Subtracting $iPA \times PA$ from Eq. (B43) one finally gets for the full non-Abelian Berry curvature $P \mathbf{F}_{nA} P = P (\nabla \times \mathbf{A}_\Psi - i \mathbf{A}_\Psi P) \times PA \mathbf{F}$ the expression
\[
F_{nA}^k = D^\Psi k + F_{\Psi A}^k D^\Psi k - \pi
\]
Now, since both the full curvature as well as the $A \times A$-term transform the same proper way, also the difference $(\mathbf{f})_C - i \langle \mathbf{A} \rangle_C Q_i \times Q_i \langle \mathbf{A} \rangle_C$ in Eq. (54b) must transform this way.

The discussion of this section shows that the approximation Eq. (46) leads to proper behavior of the connection and hence curvature and consequently, that the leading term $\langle Si \nabla \times (S \nabla) \rangle$ alone cannot transform properly in the periodic gauge.

5. Bloch sum gauge invariance

We show the influence of the Bloch sum gauge choice (λ) on various expressions used in the main text. From Eq. (A4) with $\Phi \rightarrow w$ one gets
\[
w^{k,\lambda}_w = w^{k,\lambda}_s \Lambda^k
\]
\[
\Lambda^k = e^{i\kappa s} \lambda
\]
\[
0 = [\Lambda^k, s]_-
\]
\[
S^{k,\lambda} = \Lambda^{-k} s^{k,\lambda} \Lambda^k
\]
with $\lambda = 1 - \lambda$ and $(\lambda - \bar{\lambda})^2 = 1$. Together with $\Psi^k = w^k C^k = w^k \lambda C^k$ this leads to
\[
C^{k,\lambda} = C^k \lambda C^k
\]
\[
A^{k,\lambda}_w = \Lambda^{-k} \left( w^k \frac{\partial}{\partial k} \right) \Lambda^k
\]
\[
= \Lambda^{-k} \left( w^k \frac{\partial}{\partial k} \right) \Lambda^k
\]
\[
= \Lambda^{-k} \left( w^k \frac{\partial}{\partial k} \right) \Lambda^k
\]
and altogether to
\[
A^{k,\lambda}_w = \Lambda^{-k} \left( w^k \frac{\partial}{\partial k} \right) \Lambda^k
\]
\[
= \Lambda^{-k} \left( w^k \frac{\partial}{\partial k} \right) \Lambda^k
\]
so, the basis connection does not transform like a simple operator (as the overlap Eq. (B48)). Eqs. (B47,B49,B50) then give
\[
\langle A \rangle^\lambda_C = C^{k,\lambda} + A^{k,\lambda}_w C^{k,\lambda}
\]
\[
= C^{k,\lambda} + A^{k,\lambda}_w C^{k,\lambda}
\]
\[
= C^{k,\lambda} + A^{k,\lambda}_w C^{k,\lambda}
\]
and in a similar way
\[
(\hat{S}i\nabla)_C^\lambda = C^{k\lambda} + S^{k\lambda} \nabla_k C^{k\lambda} = C^{k\lambda} + \left( S^{k\lambda} \nabla_k \Lambda^{k+} + C^{k\bar{x}} \right) \\
= C^{k\lambda} + \left[ s^{k\lambda} \Lambda^{k+} i \nabla_k + S^{k\lambda} i \left( \nabla_k \Lambda^{k+} \right) \right] C^{k\bar{x}} \\
= \langle \hat{S}i\nabla C \rangle^\lambda_C + (\lambda - \bar{\lambda}) \langle S \rangle_C^\lambda 
\]

Consequently, Eqs. (49,50) are not invariant under gauge change of the Bloch sums, even though they are expressions in the eigenbasis. However, their sum and hence the Berry connection matrix in eigenbasis Eq. (54a) and the \( A \times A \)-term in Eq. (54b) are invariant. In the following we drop the \( k \)-index for simplification. The gradient of Eq. (B48) then reads
\[
\nabla S^\lambda = \Lambda^+ \left( \nabla S^\bar{x} + i (\lambda - \bar{\lambda}) \left[ S^\bar{x}, s \right] \right) \Lambda 
\] (B53)
The curl of the basis connection together with Eqs. (B50,35) and \( SS \times s = 0 \) then reads
\[
\nabla \times A_w^\lambda = \nabla \times \left( \Lambda^+ A_w^\bar{x} \Lambda \right) - (\lambda - \bar{\lambda}) \left( \nabla S^\lambda \right) \times s \\
= \Lambda^+ \left( \nabla \times A_w^\bar{x} \right) \Lambda \\
- i \Lambda^+ (\lambda - \bar{\lambda}) \left( s \times A_w^\bar{x} + A_w^\bar{x} \times s \right) \Lambda \\
+ \Lambda^+ (i s S^\bar{x} \times s) \Lambda 
\] (B54)
which bracketed between \( C_k \), by inserting \( CC^+S = 1 \) where needed and using Eq. (B49a), gives
\[
\langle \nabla \times A_w \rangle_C^\lambda = \langle \nabla \times A_w \rangle_C^\bar{x} \\
- i (\lambda - \bar{\lambda}) \left( \langle s \times A_w + A_w^\bar{x} \times s \rangle \right) \times C \\
+ i \langle s S \times s \rangle C
\] (B55)
Finally, \( \langle SS \rangle_C^\lambda = \langle S \rangle_C^\bar{x} \) and application of \( CC^+S = 1 \) and Eq. (B51) yields
\[
\langle A \rangle_C^{\lambda+} \times \langle A \rangle_C^\lambda = \langle A_w \rangle_C^{\bar{x}+} \times \langle A_w \rangle_C^{\bar{x}} \\
- (\lambda - \bar{\lambda}) \langle s \times A_w + A_w^\bar{x} \times s \rangle \times C \\
+ \langle s S \times s \rangle \times C
\] (B56)
from which follows
\[
\langle f \rangle_C^\lambda - i \langle A_w \rangle_C^{\lambda+} \times \langle A_w \rangle_C^\lambda \\
= \langle f \rangle_C^{\bar{x}} - i \langle A_w \rangle_C^{\bar{x}+} \times \langle A_w \rangle_C^{\bar{x}} \n\]
and hence the full Berry connection Eq. (54b) is invariant too.

### Appendix C: Crystal structures

1. **CaCuO2**

CaCuO2 forms a tetragonal lattice with spacegroup 123 (P4/mmm). We used lattice parameters \( a_0 = 7.29434 \) \( a_B \) and \( c_0 = 6.04712 \) \( a_B \) with Wyckoff: positions Ca at \( 1d = \left( \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \right) \), Cu at \( 1a = (000) \) and O at \( 2f = \left( \frac{1}{4}, 0, 0 \right) \). We performed a non spin polarized calculation in scalar relativistic mode within the local (spin) density approximation L(S)DA(33) (PW92) in FPLO version 19.00-60. The self-consistent \( k \)-mesh contains 12\(^3\) points in the primitive reciprocal unit cell.

2. **bcc Fe**

We performed a full relativistic spin polarized calculation for bcc iron with space group 220 (Im3m), Wyckoff position \( 2a = (000) \) and lattice parameter \( a_0 = 5.4049 \) in FPLO version 19.00-60. The exchange and correlation functional is LSDA (PW92) and the magnetization axis is (001) with resulting spin moment of 2.19\( \mu_B \). The self consistent \( k \)-mesh contains 16\(^3\) points in the primitive reciprocal unit cell.

3. **B2 FeAl**

We performed a full relativistic spin polarized calculation for B2 FeAl with space group 221 (Pn3m) with Fe at Wyckoff position \( 1a = (000) \) and Al at \( 1b = \left( \frac{1}{2}, \frac{1}{2}, \frac{1}{2} \right) \) and lattice parameter \( a_0 = 5.3640 \) in FPLO version 19.00-60. The exchange and correlation functional is LSDA (PW92) and the magnetization axis is (001) with resulting spin moment of 0.662\( \mu_B \). The self consistent \( k \)-mesh contains 12\(^3\) points in the primitive reciprocal unit cell.

4. **HgS**

We performed a full relativistic non spin polarized calculation for HgS with space group 216 (F43m) with Hg at Wyckoff position \( 4a = (000) \) and S at \( 4c = \left( \frac{1}{4}, \frac{1}{4}, \frac{1}{4} \right) \) and lattice parameter \( a_0 = 5.8586 \) in FPLO version 19.00-60. The exchange and correlation functional is LSDA (PW92). The self consistent \( k \)-mesh contains 12\(^3\) points in the primitive reciprocal unit cell.

5. **MgB\(_2\)**

We performed a scalar relativistic non spin polarized calculation for MgB\(_2\) with space group 191 (P6/mmm) with Mg at Wyckoff position \( 1a = (000) \) and B at \( 2d = \left( \frac{1}{3}, \frac{1}{3}, \frac{1}{2} \right) \) and lattice parameters \( a_0 = 3.078a_B, c_0 = 3.552a_B \) in FPLO version 19.00-60. The exchange and correlation functional is LSDA (PW92). The self consistent \( k \)-mesh contains 12\(^3\) points in the primitive reciprocal unit cell.
We performed a scalar relativistic non spin polarized calculation for H$_2$O in the group Pnnm2 with H at Wyckoff position 2c = (1.457, 0, 1.127)$a_B$ and O at 1a = (000) in FPLO version 19.00-60 within LDA (PW92).

For the interested reader we give an (incomplete) list of recent publications, which used the FPLO Wannier function module: [84] [85] [86] [87] [88] [89] [90] [91] [92] [93] [94] [95] [96] [97]
[96] M. Yao, K. Manna, Q. Yang, A. Fedorov, V. Voroshnin, B. V. Schwarze, J. Hornung, S. Chattopadhyay, Z. Sun, S. N. Guin, et al., Nature communications 11, 1 (2020).

[97] Q. Xu, G. Li, Y. Zhang, Q. Yang, Y. Sun, and C. Felser, ACS catalysis 10, 5042 (2020).