Supplementary material for "VASP2KP: $k \cdot p$ models and Landé g-factors from *ab-initio* calculations"

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A. Löwdin partitioning theory to obtain $k \cdot p$ Hamiltonian

The Löwdin partitioning theory, also called as quasi-degenerate perturbation, is really a useful and important method to make an approximation to simplify the Hamiltonian by reducing the dimension. The main idea of it is to introduce an anti-Hermitian matrix S, which can transform the origin Hamiltonian matrix H (such as $k \cdot p$ Hamiltonian H^{kp} defined by Eq. (3) in the main text to the optimal Hamiltonian matrix $\widetilde{H}$, which is expressed by

$$\widetilde{H} = e^{-S} H e^{S} \tag{A.1}$$

It is obvious that $\widetilde{H}$ has the same eigenvalues as H, thus making the corresponding bands all the same. Moreover, $\widetilde{H}$ is expected to be block diagonal, as shown in Fig. A1, where $\mathcal{A}$ corresponds to the subspace of the bands of interest and $\mathcal{B}$ corresponds to the subspace of other bands. After this transformation, we can directly use the matrix block corresponding to $\mathcal{A}$ to replace the original Hamiltonian matrix. However, it is difficult to get the analytic or accurate matrix S, so we must use the perturbation expansion method to find the series solution.

First, suppose that the Hamiltonian $H = H_0 + H'$, where H_0 is a diagonal matrix, which is the main part of the Hamiltonian while H' can be treated as a perturbation. For instance, the $k \cdot p$ Hamiltonian H^{kp} can be the sum of the diagonal matrix whose diagonal elements are the eigenvalues $\epsilon_n(\boldsymbol{k}_0)$

$$(H_0^{kp})_{mn} = \left(\epsilon_n(\boldsymbol{k}_0) + \frac{\hbar^2 k^2}{2m}\right)\delta_{mn} \tag{A.2}$$

and the perturbation terms

$$H'^{kp}_{mn} = \frac{\hbar}{m}\boldsymbol{\pi}_{mn} \cdot \boldsymbol{k} \ , \ (m \neq n) \tag{A.3}$$

with small $\boldsymbol{k}$. Furthermore, H' can be separated as the sum of H_1 and H_2 which only have nonzero elements in and between the subspaces $\mathcal{A}$ and $\mathcal{B}$, respectively, as shown in Fig. A2. Therefore, we can rewrite the origin Hamiltonian as

$$H = H_0 + H_1 + H_2 \tag{A.4}$$

We suppose that the matrix S is anti-Hermitian and in the same shape as H_2 (only has nonzero matrix elements between subspaces $\mathcal{A}$ and $\mathcal{B}$), making e^S a unitary matrix. It is simple to find that $[H_1, S]$ is in the same shape as H_2 and $[H_2, S]$ is in the same shape as H_1, no matter what H_1 and H_2 are. According to the Baker–Campbell–Hausdorff formula, Eq. (A.1) can be rewritten as

$$\begin{aligned}\widetilde{H} &= \sum_{i=0}^{+\infty} \frac{1}{i!}[H, S]^{(i)} \\ &= \sum_{i=0}^{+\infty} \frac{1}{(2i)!}[H_0 + H_1, S]^{(2i)} + \sum_{i=0}^{+\infty} \frac{1}{(2i+1)!}[H_2, S]^{(2i+1)} + \sum_{i=0}^{+\infty} \frac{1}{(2i+1)!}[H_0 + H_1, S]^{(2i+1)} + \sum_{i=0}^{+\infty} \frac{1}{(2i)!}[H_2, S]^{(2i)}\end{aligned} \tag{A.5}$$

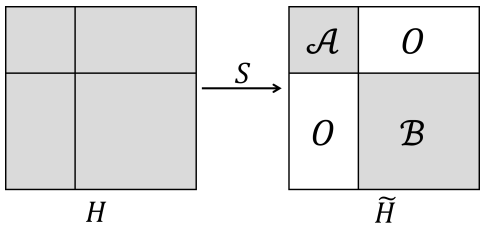

FIG. A1: Transform the original Hamiltonian H to the optimal Hamiltonian $\widetilde{H}$. The gray parts represent nontrivial matrix elements while the white parts represent trivial matrix elements (zero elements).

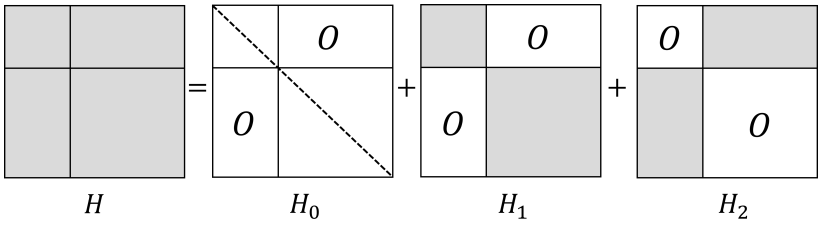

FIG. A2: The schematic of H, H_0, H_1 and H_2. The gray parts represent nontrivial matrix elements while the white parts represent trivial matrix elements.

The sum of first two terms of Eq. (A.5) is block diagonal in the same shape as H_1, which is denoted by

$$\widetilde{H}_D = \sum_{i=0}^{+\infty} \frac{1}{(2i)!}[H_0 + H_1, S]^{(2i)} + \sum_{i=0}^{+\infty} \frac{1}{(2i+1)!}[H_2, S]^{(2i+1)} \tag{A.6}$$

and the sum of last two terms of Eq. (A.5) is non-block diagonal in the same shape as H_2, which is denoted by

$$\widetilde{H}_N = \sum_{i=0}^{+\infty} \frac{1}{(2i+1)!}[H_0 + H_1, S]^{(2i+1)} + \sum_{i=0}^{+\infty} \frac{1}{(2i)!}[H_2, S]^{(2i)} \tag{A.7}$$

To make $\widetilde{H}$ block diagonal, we have $\widetilde{H}_N = 0$. Use the ansatz that S can be expanded as

$$S = S^{(1)} + S^{(2)} + S^{(3)} + \cdots \tag{A.8}$$

Extract the small quantities of each order in H_N and let them be 0:
1st order

$$[H_0, S^{(1)}] + H_2 = 0 \tag{A.9}$$

2nd order

$$[H_0, S^{(2)}] + [H_1, S^{(1)}] = 0 \tag{A.10}$$

3rd order

$$[H_0, S^{(3)}] + \frac{1}{6}[H_0, S^{(1)}]^{(3)} + [H_1, S^{(2)}] + \frac{1}{2}[H_2, S^{(1)}]^{(2)} = 0 \tag{A.11}$$

...

By solving Eqs. (A.9)-(A.11), the matrix elements of $S^{(i)}$ can be written as

$$\begin{cases} S^{(1)}_{\alpha l} = -\dfrac{H'_{\alpha l}}{E_\alpha - E_l} \\ S^{(2)}_{\alpha l} = \dfrac{1}{E_\alpha - E_l}\left[\sum\limits_{\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha\alpha'}H'_{\alpha' l}}{E_{\alpha'} - E_l} - \sum\limits_{l'\in\mathcal{B}} \dfrac{H'_{\alpha l'}H'_{l' l}}{E_\alpha - E_{l'}}\right] \\ S^{(3)}_{\alpha l} = \dfrac{1}{E_\alpha - E_l} \\ \quad \times\Bigg[-\sum\limits_{\alpha',\alpha''\in\mathcal{A}} \dfrac{H'_{\alpha\alpha''}H'_{\alpha''\alpha'}H'_{\alpha' l}}{(E_{\alpha''} - E_l)(E_{\alpha'} - E_l)} - \sum\limits_{l',l''\in\mathcal{B}} \dfrac{H'_{\alpha l'}H'_{l' l''}H'_{l'' l}}{(E_\alpha - E_{l''})(E_\alpha - E_{l'})} \\ \quad + \sum\limits_{l'\in\mathcal{B},\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha\alpha'}H'_{\alpha' l'}H'_{l' l}}{(E_{\alpha'} - E_l)(E_{\alpha'} - E_{l'})} + \sum\limits_{l'\in\mathcal{B},\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha\alpha'}H'_{\alpha' l'}H'_{l' l}}{(E_m - E_{l'})(E_{\alpha'} - E_{l'})} \\ \quad + \dfrac{1}{3}\sum\limits_{l'\in\mathcal{B},\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha l'}H'_{l'\alpha'}H'_{\alpha' l}}{(E_{\alpha'} - E_{l'})(E_{\alpha'} - E_l)} + \dfrac{1}{3}\sum\limits_{l'\in\mathcal{B},\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha l'}H'_{l'\alpha'}H'_{\alpha' l}}{(E_\alpha - E_{l'})(E_{\alpha'} - E_{l'})} + \dfrac{2}{3}\sum\limits_{l'\in\mathcal{B},\alpha'\in\mathcal{A}} \dfrac{H'_{\alpha l'}H'_{l'\alpha'}H'_{\alpha' l}}{(E_\alpha - E_{l'})(E_{\alpha'} - E_l)}\Bigg] \\ \cdots = \cdots \end{cases} \tag{A.12}$$

where $\alpha \in \mathcal{A}$ and $l \in \mathcal{B}$. This matrix S makes $\widetilde{H}_N = 0$ so that $\widetilde{H} = \widetilde{H}_D$. After obtaining the matrix S, we can directly obtain the optimal Hamiltonian by Eq. (A.6), which is expressed by

$$\widetilde{H} = \widetilde{H}^{(0)} + \widetilde{H}^{(1)} + \widetilde{H}^{(2)} + \widetilde{H}^{(3)} + \cdots \tag{A.13}$$

where

$$\begin{cases} H^{(0)}_{\alpha\alpha'} = H^0_{\alpha\alpha'} \\ H^{(1)}_{\alpha\alpha'} = H'_{\alpha\alpha'} \\ H^{(2)}_{\alpha\alpha'} = \dfrac{1}{2}\sum\limits_{l\in\mathcal{B}} H'_{\alpha l}H'_{l\alpha'}\left[\dfrac{1}{E_\alpha - E_l} + \dfrac{1}{E_{\alpha'} - E_l}\right] \\ H^{(3)}_{\alpha\alpha'} = -\dfrac{1}{2}\sum\limits_{l\in\mathcal{B},\alpha''\in\mathcal{A}}\left[\dfrac{H'_{\alpha l}H'_{l\alpha''}H'_{\alpha''\alpha'}}{(E_{\alpha'} - E_l)(E_{\alpha''} - E_l)} + \dfrac{H'_{\alpha\alpha''}H'_{\alpha'' l}H'_{l\alpha'}}{(E_\alpha - E_l)(E_{\alpha''} - E_l)}\right] \\ \qquad + \dfrac{1}{2}\sum\limits_{l,l'\in\mathcal{B}} H'_{\alpha l}H'_{ll'}H'_{l'\alpha'}\left[\dfrac{1}{(E_\alpha - E_l)(E_\alpha - E_{l'})} + \dfrac{1}{(E_{\alpha'} - E_l)(E_{\alpha'} - E_{l'})}\right] \\ \cdots = \cdots \end{cases} \tag{A.14}$$

Up to now, we have already obtained the optimal Hamiltonian. In the case of $k\cdot p$ Hamiltonian, by substituting Eq. (A.2) and Eq. (A.3) into Eq. (A.14), we can obtain

$$\begin{cases} H^{kp(0)}_{\alpha\beta} = \left(\epsilon_\alpha(\boldsymbol{k}_0) + \dfrac{\hbar^2 k^2}{2m}\right)\delta_{\alpha\beta} \\ H^{kp(1)}_{\alpha\beta} = \dfrac{\hbar}{m}\boldsymbol{\pi}_{\alpha\beta}\cdot\boldsymbol{k} \\ H^{kp(2)}_{\alpha\beta} = \dfrac{\hbar^2}{2m^2}\sum\limits_{l\in\mathcal{B}}\sum\limits_{ij}\left[\dfrac{1}{\epsilon_\alpha(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0)} + \dfrac{1}{\epsilon_\beta(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0)}\right]\pi^i_{\alpha l}\pi^j_{l\beta}k^i k^j \\ H^{kp(3)}_{\alpha\beta} = -\dfrac{\hbar^3}{2m^3}\sum\limits_{ijq}\Bigg\{\sum\limits_{l\in\mathcal{B},\gamma\in\mathcal{A}}\left[\dfrac{\pi^i_{\alpha l}\pi^j_{l\gamma}\pi^q_{\gamma\beta}}{(\epsilon_\beta(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))(\epsilon_\gamma(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))} + \dfrac{\pi^i_{\alpha\gamma}\pi^j_{\gamma l}\pi^q_{l\beta}}{(\epsilon_\alpha(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))(\epsilon_\gamma(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))}\right] \\ \quad - \sum\limits_{l,l'\in\mathcal{B}}\left[\dfrac{1}{(\epsilon_\alpha(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))(\epsilon_\alpha(\boldsymbol{k}_0) - \epsilon_{l'}(\boldsymbol{k}_0))} + \dfrac{1}{(\epsilon_\beta(\boldsymbol{k}_0) - \epsilon_l(\boldsymbol{k}_0))(\epsilon_\beta(\boldsymbol{k}_0) - \epsilon_{l'}(\boldsymbol{k}_0))}\right]\pi^i_{\alpha l}\pi^j_{ll'}\pi^q_{l'\beta}\Bigg\}k^i k^j k^q \end{cases} \tag{A.15}$$

Therefore, the $k\cdot p$ Hamiltonian of order 2 is the sum of $H^{kp(0)}$, $H^{kp(1)}$ and $H^{kp(2)}$, which is equal to Eq. (4) in the main text. Moreover, the $k\cdot p$ Hamiltonian of order 3 can be expressed by

$$\begin{aligned}
&H^{kp(\leq 3)}_{\alpha\beta}\\
&=\left(\epsilon_\alpha(\boldsymbol{k}_0)+\frac{\hbar^2k^2}{2m}\right)\delta_{\alpha\beta}+\frac{\hbar}{m}\boldsymbol{\pi}_{\alpha\beta}\cdot\boldsymbol{k}+\frac{\hbar^2}{2m^2}\sum_{l\in\mathcal{B}}\sum_{ij}\left[\frac{1}{\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}+\frac{1}{\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}\right]\pi^i_{\alpha l}\pi^j_{l\beta}k^ik^j\\
&\quad-\frac{\hbar^3}{2m^3}\sum_{ijq}\left\{\sum_{l\in\mathcal{B},\gamma\in\mathcal{A}}\left[\frac{\pi^i_{\alpha l}\pi^j_{l\gamma}\pi^q_{\gamma\beta}}{(\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))(\epsilon_\gamma(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))}+\frac{\pi^i_{\alpha\gamma}\pi^j_{\gamma l}\pi^q_{l\beta}}{(\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))(\epsilon_\gamma(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))}\right]\right.\\
&\qquad\left.-\sum_{l,l'\in\mathcal{B}}\left[\frac{1}{(\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))(\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_{l'}(\boldsymbol{k}_0))}+\frac{1}{(\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0))(\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_{l'}(\boldsymbol{k}_0))}\right]\pi^i_{\alpha l}\pi^j_{ll'}\pi^q_{l'\beta}\right\}k^ik^jk^q
\end{aligned}\tag{A.16}$$

B. Derivation of Zeeman's coupling

It is easy to compute the commutator $[\partial^i, A^j]$ as

$$[\partial^i,A^j]\phi=\partial^i(A^j\phi)-A^j\partial^i\phi=(\partial^iA^j)\phi+A^j\partial^i\phi-A^j\partial^i\phi=(\partial^iA^j)\phi\tag{B.1}$$

or in a simple form

$$[\partial^i,A^j]=\partial^iA^j.\tag{B.2}$$

In addition, the components of the magnetic field can be expressed by

$$B_k=(\nabla\times\boldsymbol{A})\cdot\boldsymbol{e}^k=\sum_{lmn}\epsilon_{lmn}\partial^mA^n\boldsymbol{e}^l\cdot\boldsymbol{e}^k=\sum_{lmn}\epsilon_{lmn}\partial^mA^n\delta_{lk}=\sum_{mn}\epsilon_{kmn}\partial^mA^n\tag{B.3}$$

where ϵ_{lmn} is the Levi-Civita symbol and A is the magnetic vector potential. Therefore, we can establish the relation that

$$\sum_k\epsilon^{ijk}B_k=\sum_{mnk}\epsilon^{ijk}\epsilon_{kmn}\partial^mA^n=\sum_{mn}\left(\sum_k\epsilon^{ijk}\epsilon_{kmn}\right)\partial^mA^n=\sum_{mn}(\delta_{im}\delta_{jn}-\delta_{in}\delta_{jm})\partial^mA^n=\partial^iA^j-\partial^jA^i\tag{B.4}$$

Therefore, we can obtain the relation in Sec. IIB that

$$\left[-i\hbar\partial^i+eA^i,-i\hbar\partial^j+eA^j\right]=-ie\hbar(\left[\partial^i,A^j\right]-\left[\partial^j,A^i\right])=-i\hbar e(\partial^iA^j-\partial^jA^i)=-i\hbar e\sum_k\epsilon^{ijk}B_k\tag{B.5}$$

Furthermore, after replacing k^i by $-i\hbar\partial^i+eA^i$ (Peierls substitution) in Eq. (4) in the main text, the last summation can be transformed as

$$\begin{aligned}
&\sum_{ij}\pi^i_{\alpha l}\pi^j_{l\beta}(-i\hbar\partial^i+eA^i)(-i\hbar\partial^j+eA^j)\\
&=\sum_{ij}\pi^i_{\alpha l}\pi^j_{l\beta}\left(\frac{1}{2}\left[-i\hbar\partial^i+eA^i,-i\hbar\partial^j+eA^j\right]+\frac{1}{2}\left\{-i\hbar\partial^i+eA^i,-i\hbar\partial^j+eA^j\right\}\right)\\
&=-\frac{i\hbar e}{2}\sum_{ijk}\pi^i_{\alpha l}\pi^j_{l\beta}\epsilon^{ijk}B_k+\frac{\hbar^2}{2}\sum_{ij}\pi^i_{\alpha l}\pi^j_{l\beta}\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\left(-i\partial^j+\frac{e}{\hbar}A^j\right)+\frac{\hbar^2}{2}\sum_{ij}\pi^i_{\alpha l}\pi^j_{l\beta}\left(-i\partial^j+\frac{e}{\hbar}A^j\right)\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\\
&=-\frac{i\hbar e}{2}\sum_{ijk}\pi^i_{\alpha l}\pi^j_{l\beta}\epsilon^{ijk}B_k+\frac{\hbar^2}{2}\sum_{ij}\pi^i_{\alpha l}\pi^j_{l\beta}\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\left(-i\partial^j+\frac{e}{\hbar}A^j\right)+\frac{\hbar^2}{2}\sum_{ij}\pi^j_{\alpha l}\pi^i_{l\beta}\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\left(-i\partial^j+\frac{e}{\hbar}A^j\right)\\
&=-\frac{i\hbar e}{2}\sum_{ijk}\pi^i_{\alpha l}\pi^j_{l\beta}\epsilon^{ijk}B_k+\hbar^2\sum_{ij}\frac{\pi^i_{\alpha l}\pi^j_{l\beta}+\pi^j_{\alpha l}\pi^i_{l\beta}}{2}\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\left(-i\partial^j+\frac{e}{\hbar}A^j\right)
\end{aligned}\tag{B.6}$$

The Hamiltonian of Zeeman's coupling is the gauge independent part in Eq. (4) (after Peierls substitution), which is expressed as

$$H^Z_{\alpha\beta} = \frac{\mu_B}{\hbar}\left(\boldsymbol{L}_{\alpha\beta} + 2\boldsymbol{s}_{\alpha\beta}\right)\cdot\boldsymbol{B} \tag{B.7}$$

where

$$L^k_{\alpha\beta} = -\frac{i\hbar}{2m}\sum_{l\in\mathcal{B}}\sum_{ij}\epsilon^{ijk}\pi^i_{\alpha l}\pi^j_{l\beta}\left(\frac{1}{\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}+\frac{1}{\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}\right) \tag{B.8}$$

and $\frac{2}{\hbar}\mu_B\boldsymbol{s}\cdot\boldsymbol{B}$ is the Zeemans's coupling of the bare electron. In addition, the gauge dependent part in Eq. (4) (after Peierls substitution) is expressed by

$$H^{kp}_{\alpha\beta} = \epsilon_\alpha(\boldsymbol{k}_0)\delta_{\alpha\beta} + \frac{\hbar}{m}\boldsymbol{\pi}_{\alpha\beta}\cdot\left(-i\nabla+\frac{e}{\hbar}\boldsymbol{A}\right) + \sum_{ij}M^{ij}_{\alpha\beta}\left(-i\partial^i+\frac{e}{\hbar}A^i\right)\left(-i\partial^j+\frac{e}{\hbar}A^j\right) \tag{B.9}$$

where

$$M^{ij}_{\alpha\beta} = \frac{\hbar^2}{2m}\delta_{\alpha\beta}\delta_{ij} + \frac{\hbar^2}{4m^2}\sum_{l\in\mathcal{B}}\left(\pi^i_{\alpha l}\pi^j_{l\beta}+\pi^j_{\alpha l}\pi^i_{l\beta}\right)\left(\frac{1}{\epsilon_\alpha(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}+\frac{1}{\epsilon_\beta(\boldsymbol{k}_0)-\epsilon_l(\boldsymbol{k}_0)}\right). \tag{B.10}$$

The Eqs. (B.7)-(B.10) are the same as Eqs. (5)-(8).

C. Construction of the coefficient matrix Q for finding the unitary transformation U

Only the generators of the group L should be taken into account when finding the unitary transformation matrix U. T is an anti-unitary generator, while S are the unitary generators. The Eq. (17) in the main text can be rewritten as

$$D^{\text{num}}(S)U - UD^{\text{std}}(S) = \mathcal{O} \tag{C.1}$$

$$D^{\text{num}}(T)U^* - UD^{\text{std}}(T) = \mathcal{O} \tag{C.2}$$

where $\mathcal{O}$ is a zero matrix. The matrices U, $D^{\text{std}}(R)$ and $D^{\text{num}}(R)$ are complex, so we can consider the real parts and the imaginary parts separately, thus transforming Eqs. (C.1)-(C.2) into

$$\begin{cases} D^{\text{num}}_r(S)U_r - U_rD^{\text{std}}_r(S) - D^{\text{num}}_i(S)U_i + U_iD^{\text{std}}_i(S) = \mathcal{O} \\ D^{\text{num}}_i(S)U_r - U_rD^{\text{std}}_i(S) + D^{\text{num}}_r(S)U_i - U_iD^{\text{std}}_r(S) = \mathcal{O} \end{cases} \tag{C.3}$$

and

$$\begin{cases} D^{\text{num}}_r(T)U_r - U_rD^{\text{std}}_r(T) + D^{\text{num}}_i(T)U_i + U_iD^{\text{std}}_i(T) = \mathcal{O} \\ -D^{\text{num}}_i(T)U_r + U_rD^{\text{std}}_i(T) + D^{\text{num}}_r(T)U_i + U_iD^{\text{std}}_r(S) = \mathcal{O} \end{cases} \tag{C.4}$$

where the subscripts r represent the real parts of U, $D^{\text{std}}(R)$ or $D^{\text{num}}(R)$ and the subscripts i represent the imaginary parts. Consider the real part and the imaginary part of each elements of U as independent variables, which are denoted as $U_{r11}, U_{r12}, \cdots U_{rnn}$ and $U_{i11}, U_{i12}, \cdots U_{inn}$, respectively. From Eqs. (C.3)-(C.4), it is clear to find that the matrix equations are all linear equations and all the parameters and variables are real. Introduced a column vector $\boldsymbol{u} = (U_{r11}, U_{r12}, \cdots, U_{rnn}, U_{i11}, U_{i12}, \cdots, U_{inn})^T$, which is comprised of all the independent variables to be solved, Eqs. (C.3)-(C.4) can be rewritten as

$$A(S)\boldsymbol{u} = \boldsymbol{0} \tag{C.5}$$

where

$$A(S) = \begin{pmatrix} D^{\text{num}}_r(S)\otimes I - I\otimes D^{\text{std}T}_r(S) & -D^{\text{num}}_i(S)\otimes I + I\otimes D^{\text{std}T}_i(S) \\ D^{\text{num}}_i(S)\otimes I - I\otimes D^{\text{std}T}_i(S) & D^{\text{num}}_r(S)\otimes I - I\otimes D^{\text{std}T}_r(S) \end{pmatrix} \tag{C.6}$$

and

$$B(T)\boldsymbol{u} = \mathbf{0} \tag{C.7}$$

where

$$B(T) = \begin{pmatrix} D_r^{\mathrm{num}}(T) \otimes I - I \otimes D_r^{\mathrm{std}T}(T) & D_i^{\mathrm{num}}(T) \otimes I + I \otimes D_i^{\mathrm{std}T}(T) \\ -D_i^{\mathrm{num}}(T) \otimes I + I \otimes D_i^{\mathrm{std}T}(T) & D_r^{\mathrm{num}}(T) \otimes I + I \otimes D_r^{\mathrm{std}T}(T) \end{pmatrix}. \tag{C.8}$$

The Eq. (C.5) holds for all unitary generators $S_1, S_2, \cdots, S_n$. Combined Eq. (C.5) with Eq. (C.6), we can construct the large parameter matrix Q so that the vector $\boldsymbol{u}$ corresponds to the transformation matrix U satisfies $Q\boldsymbol{u} = \mathbf{0}$, where

$$Q = (A^T(S_1), A^T(S_2), \cdots, A^T(S_n), B^T(T))^T. \tag{C.9}$$

Therefore, all vectors in the null space of the matrix Q is the solution to Eq. (16) in the main text.

D. Functions of `vasp2mat`

Except for the calculation of matrices of generalized momentum $\hat{\boldsymbol{\pi}} = \hat{\boldsymbol{p}} + \frac{1}{2mc^2}(\hat{\boldsymbol{s}} \times \nabla V(\boldsymbol{r}))$, spin $\hat{\boldsymbol{s}}$, time reversal operator $\hat{T}$ and crystalline symmetry operators $\hat{R}$, the patch `vasp2mat` can also do other calculations by setting the parameter `vmat` in `INCAR.mat`. All functions of `vasp2mat` are shown in TABLE D1, where $\hat{\boldsymbol{\sigma}}$ is Pauli operator. The matrix of time reversal operator $\hat{T}$ can be calculated with "`vmat=12; time_rev=.true.`".

TABLE D1: The function of different `vmat` of `vasp2mat`.

vmat	Functions
1	Calculate overlap matrix $\langle m(\boldsymbol{K})\|n(\boldsymbol{K})\rangle$
2	Calculate soft local potential matrix $\langle \widetilde{m}(\boldsymbol{K})\|\hat{V}_{\mathrm{eff}}\|\widetilde{n}(\boldsymbol{K})\rangle$
3	Calculate kinetic energy matrix of pseudo wavefunctions $\langle \widetilde{m}(\boldsymbol{K})\|\hat{T}_k\|\widetilde{n}(\boldsymbol{K})\rangle$
4	Calculate nonlocal potential matrix $\langle \widetilde{m}(\boldsymbol{K})\|\hat{V}_{NL}\|\widetilde{n}(\boldsymbol{K})\rangle$
5	Calculate Hamiltonian matrix $\langle m(\boldsymbol{K})\|\hat{H}\|n(\boldsymbol{K})\rangle$
7	Calculate momentum matrices $\langle m(\boldsymbol{K})\|\hat{\boldsymbol{p}}\|n(\boldsymbol{K})\rangle$
8	Calculate SOC Hamiltonian matrix $\langle m(\boldsymbol{K})\|\hat{H}_{SOC}\|n(\boldsymbol{K})\rangle$
10	Calculate spin matrices $\langle m(\boldsymbol{K})\|\hat{\boldsymbol{\sigma}}\|n(\boldsymbol{K})\rangle$
11	Calculate generalized momentum matrices $\langle m(\boldsymbol{K})\|\hat{\boldsymbol{\pi}}\|n(\boldsymbol{K})\rangle$
12	Calculate matrix representation of a symmetry operator $\langle m(\boldsymbol{K})\|\hat{R}\|n(\boldsymbol{K})\rangle$
13	Calculate Berry curvature, anomalous Hall conductance and spin Hall conductance
14	Calculate Wilson loops to obtain Berry phases

E. The standard matrix representations

TABLE E1: In Na_3Bi, the matrix representations of $\overline{\text{DT}}7$ and $\overline{\text{DT}}8$ irreps at $\boldsymbol{k}_D$ (0 0 w) are given on BCS server, https://www.cryst.ehu.es/cgi-bin/cryst/programs/corepresentations_out.pl?super=194.264&vecfinal=DT.

$D^{\text{std}}(R)$ \ irrep / R	$\overline{\text{DT}}7$	$\overline{\text{DT}}8$
$\{C_{3z}\|0,0,0\}$	$\begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}$	$\begin{pmatrix} e^{-\frac{\pi i}{3}} & 0 \\ 0 & e^{\frac{\pi i}{3}} \end{pmatrix}$
$\{C_{2z}\|0,0,\frac{1}{2}\}$	$\begin{pmatrix} -ie^{i\pi w} & 0 \\ 0 & ie^{i\pi w} \end{pmatrix}$	$\begin{pmatrix} -ie^{i\pi w} & 0 \\ 0 & ie^{i\pi w} \end{pmatrix}$
$\{M_x\|0,0,0\}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & e^{-\frac{2\pi i}{3}} \\ e^{-\frac{\pi i}{3}} & 0 \end{pmatrix}$
$\{TP\|0,0,0\}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}\mathcal{K}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}\mathcal{K}$

TABLE E2: In Te, the matrix representations of $\overline{\text{H}}4,5$ and $\overline{\text{H}}6$ irreps at H are given on BCS server, https://www.cryst.ehu.es/cgi-bin/cryst/programs/corepresentations_out.pl?super=152.34&vecfinal=H.

$D^{\text{std}}(R)$ \ irrep / R	$\overline{\text{H}}4$	$\overline{\text{H}}5$	$\overline{\text{H}}6$
$\{C_{3z}\|0,0,\frac{1}{3}\}$	1	1	$\begin{pmatrix} e^{-\frac{2\pi i}{3}} & 0 \\ 0 & e^{\frac{2\pi i}{3}} \end{pmatrix}$
$\{C_{2x}\|0,0,\frac{2}{3}\}$	i	$-i$	$\begin{pmatrix} 0 & e^{\frac{2\pi i}{3}} \\ e^{\frac{\pi i}{3}} & 0 \end{pmatrix}$

TABLE E3: In WZ InAs, the matrix representations of $\overline{\text{GM}}7$ and $\overline{\text{GM}}8$ irreps at Γ are given on BCS server, https://www.cryst.ehu.es/cgi-bin/cryst/programs/corepresentations_out.pl?super=186.204&vecfinal=GM.

$D^{\text{std}}(R)$ \ irrep / R	$\overline{\text{GM}}7$	$\overline{\text{GM}}8$
$\{C_{3z}\|0,0,0\}$	$\begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix}$	$\begin{pmatrix} e^{-\frac{\pi i}{3}} & 0 \\ 0 & e^{\frac{\pi i}{3}} \end{pmatrix}$
$\{C_{2z}\|0,0,\frac{1}{2}\}$	$\begin{pmatrix} -i & 0 \\ 0 & i \end{pmatrix}$	$\begin{pmatrix} -i & 0 \\ 0 & i \end{pmatrix}$
$\{M_x\|0,0,0\}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}$	$\begin{pmatrix} 0 & e^{-\frac{2\pi i}{3}} \\ e^{-\frac{\pi i}{3}} & 0 \end{pmatrix}$
$\{T\|0,0,0\}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}\mathcal{K}$	$\begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}\mathcal{K}$

TABLE E4: In 1H-TMD monolayers, the matrix representations of $\overline{\text{K}}8$ and $\overline{\text{K}}11$ irreps at K are given on BCS server, https://www.cryst.ehu.es/cgi-bin/cryst/programs/corepresentations_out.pl?super=187.210&vecfinal=K.

$D^{\text{std}}(R)$ \ irrep / R	$\overline{\text{K}}8$	$\overline{\text{K}}11$
$\{C_{3z}\|0,0,0\}$	-1	$e^{\frac{\pi i}{3}}$
$\{M_z\|0,0,0\}$	i	$-i$
$\{M_xT\|0,0,0\}$	$-1\cdot\mathcal{K}$	$e^{-\frac{\pi i}{3}}\cdot\mathcal{K}$

F. Sixband model at Γ in wurtzite InAs

At Γ in the Brillouin zone of InAs, we consider six valence bands for the hole doping sample. The band representations are $\overline{\text{GM}}8$, $\overline{\text{GM}}8$ and $\overline{\text{GM}}7$ in the ascending order. The representation matrices of generators are presented in TABLE E3. The second order $k\cdot p$ Hamiltonian of six valence bands of InAs at Γ is expressed by

$$
\begin{cases}
H_{11}^{kp} =H_{22}^{kp} = a_1 + \dfrac{4\sqrt{3}}{3}a_4 + d_1 k_+ k_- + d_2 k_z^2 \\
\qquad d_1 = c_1 + \dfrac{4\sqrt{3}}{3}c_5, d_2 = c_{10} + \dfrac{4\sqrt{3}}{3}c_{13}k_z^2 \\
H_{12}^{kp} =\xi_+(2b_1 + 4b_2 - 3b_3 - 2b_4)k_- \\
H_{13}^{kp} =H_{24}^{kp} = 2a_3 - 2ib_6 k_z + d_5 k_+ k_- + c_{12}k_z^2 \\
\qquad d_5 = 2c_3 + \dfrac{2\sqrt{3}}{3}c_4 \\
H_{14}^{kp} =2\xi_+ b_2 k_- + \dfrac{2\sqrt{3}}{3}i\xi_+ c_8 k_- k_z \\
H_{15}^{kp} =H_{26}^{kp*} = \left(-2c_3 - \dfrac{2\sqrt{3}}{3}c_4 + 2c_6\right)\xi_- k_-^2 \\
H_{16}^{kp} =2\xi_-(b_2 - b_4)k_+ - \dfrac{\sqrt{3}}{3}i\xi_-(2c_7 + 2c_8 + 3c_9)k_+ k_z \\
H_{23}^{kp} =2\xi_- b_2 k_+ + \dfrac{2\sqrt{3}}{3}i\xi_- c_8 k_+ k_z \\
H_{25}^{kp} =2\xi_+(b_2 - b_4)k_- - \dfrac{\sqrt{3}}{3}i\xi_+(2c_7 + 2c_8 + 3c_9)k_- k_z \\
H_{33}^{kp} =H_{44}^{kp} = a_1 + 2a_2 - \dfrac{2\sqrt{3}}{3}a_4 + d_3^+ k_+ k_- + d_4^+ k_z^2 \\
H_{34}^{kp} = -2\xi_+(b_2 - b_4 - b_5)k_- \\
H_{35}^{kp} =H_{46}^{kp*} = -\dfrac{2\sqrt{3}}{3}\xi_- c_4 k_-^2 \\
H_{36}^{kp} = -(4b_2 - 3b_3 - 2b_4)\xi_- k_+ - \dfrac{2\sqrt{3}}{3}i\xi_- c_7 k_+ k_z \\
H_{45}^{kp} = -(4b_2 - 3b_3 - 2b_4)\xi_+ k_- - \dfrac{2\sqrt{3}}{3}i\xi_+ c_7 k_- k_z \\
H_{55}^{kp} =H_{66}^{kp} = a_1 - 2a_2 - \dfrac{2\sqrt{3}}{3}a_4 + d_3^- k_+ k_- + d_4^- k_z^2 \\
\qquad d_3^\pm = c_1 \pm 2c_2 - \dfrac{2\sqrt{3}}{3}c_5, \\
\qquad d_4^\pm = c_{10} \pm 2c_{11} - \dfrac{2\sqrt{3}}{3}c_{13} \\
H_{56}^{kp} =0 \\
\qquad \text{with} \quad \xi_\pm = 1 \pm \sqrt{3}i
\end{cases}
\tag{F.1}
$$

The Zeeman's coupling of six valence bands of InAs at Γ can be expressed by

$$H^Z = \frac{\mu_B}{2}\begin{pmatrix} h_1B_z & h_3B_+ & 2g_7B_z & h_4B_- & 0 & h_5B_+ \\ & -h_1B_z & h_4^*B_+ & -2g_7B_z & h_5^*B_- & 0 \\ & & 3g_9B_z & h_6B_- & 0 & h_7B_+ \\ & & & -3g_9B_z & h_7^*B_- & 0 \\ & & \dagger & & h_2B_z & 0 \\ & & & & & -h_2B_z \end{pmatrix}$$

$$\begin{aligned} h_1 &= 2h_6 + \frac{2\sqrt{3}}{3}g_8 + 3g_9 \\ h_2 &= \frac{4\sqrt{3}}{3}g_8 + 3g_9 \\ h_3 &= -\frac{\sqrt{3}}{3}i\xi_+(2g_1 + 4g_2 - 3g_3 - 2g_4) \\ h_4 &= -\frac{2\sqrt{3}}{3}i\xi_+ g_2 \\ h_5 &= \frac{2\sqrt{3}}{3}i\xi_-(g_2 - g_4) \\ h_6 &= \frac{2\sqrt{3}}{3}i\xi_+(g_2 - g_4 - g_5) \\ h_7 &= -\frac{\sqrt{3}}{3}i\xi_-(4g_2 - 3g_3 - 2g_4) \\ &\text{with} \quad \xi_\pm = 1 \pm \sqrt{3}i \end{aligned} \tag{F.2}$$

The values of the parameters $\{a_i, b_i, c_i, g_i\}$ are presented in TABLE F1.

TABLE F1: The computed values of parameters $\{a_i, b_i, c_i, g_i\}$ for six valence states in InAs are obtained from the VASP calculations directly.

a (eV)	b (eV·Å)	c (eV·Å^2)	g
$a_1 = 3.91$	$b_1 = 0.13$	$c_1 = -45.78$	$g_1 = -3.28$
$a_2 = -0.09$	$b_2 = 0.08$	$c_2 = 12.94$	$g_2 = -2.44$
$a_3 = 0.02$	$b_3 = 0.11$	$c_3 = -1.54$	$g_3 = -3.16$
$a_4 = 0.03$	$b_4 = -0.09$	$c_4 = -20.14$	$g_4 = 5.50$
	$b_5 = 0.06$	$c_5 = 7.65$	$g_5 = -2.76$
	$b_6 = 0.21$	$c_6 = -2.23$	$g_6 = -11.20$
		$c_7 = 21.62$	$g_7 = -6.90$
		$c_8 = 32.86$	$g_8 = 19.82$
		$c_9 = -60.12$	$g_9 = -3.88$
		$c_{10} = -45.68$	
		$c_{11} = -7.45$	
		$c_{12} = 28.03$	
		$c_{13} = -23.43$	