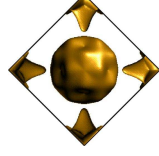


# FPLO documentation

## Band unfolding



Klaus Koepernik

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## 1 Citations

When a citation is need it would be [1, 2, 3]. The first is the idea by Wei Ku the second the first FPLO implementation/application.

### 1.1 Copy right note

The content of this report cannot be used in another publication without contacting the FPLO authors first.

## 2 Introduction

### 2.1 Original idea

The idea is due to Wei Ku [2]. Let's assume that we have a normal cell (NC) and a super cell (SC) derived from it. The NC contains  $N_s$  atoms  $\mathbf{s}_i \in [1, N_s]$  and has  $N_r$  lattice vectors  $\mathbf{r}$  in a defined Born von Karman torus. The SC contains  $N_S$  atoms  $\mathbf{S}_i \in [1, N_S]$  and has  $N_R$  lattice vectors  $\mathbf{R}$  in the **same** BvK torus. The SC is a "multiple" of NC, meaning  $N_S = fN_s$ ,  $f \in \mathbb{N}$  and because of the same BvK torus  $N_R = \frac{1}{f}N_r$ , such that both tori contain the same number of sites  $N_sN_r = N_SN_R$ . The KS states of both cells fulfill their respective Bloch theorem

$$\begin{aligned}\Psi_{\mathbf{k}N}(\mathbf{r} - \mathbf{p}) &= \Psi_{\mathbf{K}N}(\mathbf{r}) e^{-i\mathbf{k}\mathbf{p}} \\ \Psi_{\mathbf{K}N}(\mathbf{r} - \mathbf{P}) &= \Psi_{\mathbf{K}N}(\mathbf{r}) e^{-i\mathbf{K}\mathbf{P}}\end{aligned}$$

where we denote reciprocal lattice vectors in the NC Brillouin zone (bz) with  $\mathbf{k}$  and reciprocal lattice vectors in the SC Brillouin zone (BZ) with  $\mathbf{K}$ . The Bloch spectral density is defined as

$$\hat{A}(\omega) = -\frac{1}{\pi} \text{Im} \hat{G}(\omega)$$

with the single particle Green's function

$$\hat{G}(\omega) = \frac{1}{\omega^+ - \hat{H}}$$

and reads in the SC KS basis

$$\begin{aligned}A_{\overline{\mathbf{K}N}, \mathbf{K}N}(\omega) &= \langle \overline{\mathbf{K}N} | \hat{A} | \mathbf{K}N \rangle \\ &= \delta_{\overline{\mathbf{K}}\mathbf{K}} \delta_{\overline{N}N} \delta(\omega - \varepsilon_{\mathbf{K}N})\end{aligned}$$

It is diagonal in the SC quantum numbers. The full Green's function can be written as

$$\hat{G}^{-1} = \hat{G}_0^{-1} - \hat{V}$$

where  $G_0$  is the Green's function of the NC and  $V$  is the perturbing potential, which makes the difference between the exactly duplicated NC and the actual SC. Here, we already assume that only the potential is

different, meaning that all atoms are exactly the same, just moved around a bit. We will discuss this later. Of course, if  $V$  is weak, one can argue that the NC KS states  $\Psi_{\mathbf{k}n}$  are also a good basis to express the Bloch spectral density. When done so,  $A$  will no longer be diagonal in the  $\mathbf{k}$  indices, since the NC Bloch symmetry is broken in SC. However, focusing only on the diagonal elements one get's an approximation for  $A$ , which will have approximatly the Bloch symmetry of the NC and hence  $A_{\mathbf{k}n,\mathbf{k}n} = \langle \mathbf{k}n | \hat{A} | \mathbf{k}n \rangle$  will be the unfolded Bloch spectral density. Of course,  $\Psi_{\mathbf{k}n}$  can only be a basis of SC, if SC is a “multiple” of NC (no atom substitution, see discussion in Section 3).

## 2.2 The local orbital connection

Now, one can express the SC  $\Psi_{\mathbf{K}N}$  in terms of Wannier functions (WFs) and claim that the WF's are the same in NC. Then one can transfer the WF's from SC to NC and build the KS functions in both cells from the same basis. This allows to express everything in terms of these WF's. In FPLO the local basis forms a complete WF basis, if Löwdin orthogonalized. (This orthogonalization leads to a new basis  $\tilde{\Phi} = \Phi S^{-\frac{1}{2}}$ , which has the property of being the one “closest” to the non-orthogonal basis. Hence, an L-ortho FPLO basis is very localized and is a WF basis. To avoid unnessecary complications we work in our non-ortho FPLO basis and use gross-projection, when needed ( $\frac{1}{2}(S * + * S)$ ). Under the assumption that no atom substitution has taken place in SC we can do the algebra and calculate the following. The KS states are expressed via Bloch sums of WF's/local orbitals

$$\Phi_{S\mu}^{\mathbf{K}} = \frac{1}{\sqrt{N_R}} \sum_{\mathbf{R}} \Phi_{R S\mu} e^{i\mathbf{K}(\mathbf{R}+\mathbf{S})}$$

where  $\Phi_{R S\mu}$  is the  $\mu$  orbital at site  $\mathbf{S}$  in cell  $\mathbf{R}$ . The KS-state is a simple linear combination of these Bloch sums

$$\Psi_{\mathbf{K}N} = \sum_{S\mu} \Phi_{S\mu}^{\mathbf{K}} C_{S\mu,N}^{\mathbf{K}}$$

Hence,

$$\begin{aligned} \hat{A} &= \sum_{\mathbf{K}N} \Psi_{\mathbf{K}N} \delta(\omega - \varepsilon_{\mathbf{K}N}) \Psi_{\mathbf{K}N}^\dagger \\ &= \sum_{\mathbf{K}N} \sum_{S\mu, \bar{S}\bar{\mu}} \Phi_{S\mu}^{\mathbf{K}} n_{S\mu, \bar{S}\bar{\mu}}^{\mathbf{K}N}(\omega) \Phi_{\bar{S}\bar{\mu}}^{\mathbf{K}} \end{aligned}$$

which introduces the DOS weight matrix in the local basis

$$n_{S\mu, \bar{S}\bar{\mu}}^{\mathbf{K}N}(\omega) = C_{S\mu,N}^{\mathbf{K}} \delta(\omega - \varepsilon_{\mathbf{K}N}) C_{\bar{S}\bar{\mu},N}^{\mathbf{K}*}$$

Now, we map the sites of the SC onto the sites of NC. The SC consists of  $f$  “copies” of NC, translated by NC lattice vectors  $\mathbf{r}_j$ ,  $j \in [1, f]$ . For every site  $\mathbf{S}$  there is a site  $\mathbf{s}$  in NC, which is in the NC cell translated by  $\mathbf{r}_j$ . The SC lattice vector and the translation  $\mathbf{r}_j$  give a NC lattice vector

$$\mathbf{r} = \mathbf{R} + \mathbf{r}_j$$

and the site mapping can be written by labeling the SC sites according to their construction out of NC sites and cells (cf. Figure 1)

$$\mathbf{s} + \mathbf{r}_j = \mathbf{S}_{sj}.$$

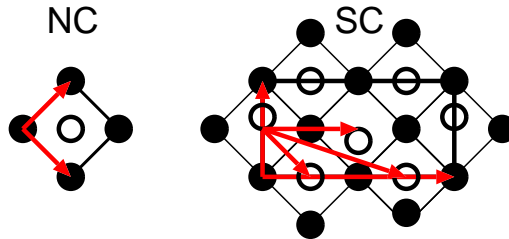


Figure 1: Cell mapping: The SC as the “fourfold” of the NC. Some of the open atoms are displaced against ideal NC positions. The small vectors show how the 4 SC open atoms are mapped onto their one NC equivalent.

If we now neglect eventual atom displacements we can form SC Bloch sums out of NC Bloch sums (but at the SC k-vector)

$$\begin{aligned}
\Phi_{[NC]s}^{\mathbf{K}} &= \frac{1}{\sqrt{N_r}} \sum_r \Phi_{rs\mu} e^{i\mathbf{K}(\mathbf{r}+\mathbf{s})} \\
&= \frac{\sqrt{N_R}}{\sqrt{N_r}} \left( \frac{1}{\sqrt{N_R}} \sum_{Rj} \Phi_{Rr_j, s\mu} e^{i\mathbf{K}(\mathbf{R}+\mathbf{r}_j+\mathbf{s})} \right) \\
&= \frac{1}{\sqrt{f}} \left( \frac{1}{\sqrt{N_R}} \sum_{Rj} \Phi_{RS_{sj}\mu} e^{i\mathbf{K}(\mathbf{R}+\mathbf{S}_{sj})} \right) \\
\Phi_{[NC]s}^{\mathbf{K}} &= \frac{1}{\sqrt{f}} \sum_j \Phi_{[SC]S_{sj}}^{\mathbf{K}}
\end{aligned} \tag{1}$$

The NC Bloch sum is a contraction of the SC Bloch sums. The KS states behave as

$$\begin{aligned}
\Psi_N^{\mathbf{K}} &= \sum_{S\mu} \Phi_{S\mu}^{\mathbf{K}} C_{S\mu, N}^{\mathbf{K}} \\
&= \sum_{sj\mu} \Phi_{S_{sj}\mu}^{\mathbf{K}} C_{S_{sj}\mu, N}^{\mathbf{K}}
\end{aligned}$$

Now, the coefficients would fulfill the translational symmetry

$$C_{S_{sj}} = C_{r_js} = C_s U$$

if  $\mathbf{r}_j$  is strictly a lattice vector. ( $U$  denotes a possible unitary transformation, which does not figure in the following, where we reverse the argumentation). Thus, if the NC Bloch symmetry were true, we could replace

$$C_{S_{sj}} = \frac{1}{f_s} \sum_{j=1}^{f_s} C_{S_{sj}}$$

Let's do that in the KS state and use Eq. (1)

$$\begin{aligned}
\Psi_N^{\mathbf{K}} &\approx \sum_{sj\mu} \Phi_{S_{sj}\mu}^{\mathbf{K}} \frac{1}{f_s} \sum_{i=1}^{f_s} C_{S_{si}\mu, N}^{\mathbf{K}} \\
&\approx \sqrt{f} \sum_{s\mu} \Phi_{[NC]s\mu}^{\mathbf{K}} \frac{1}{f_s} \sum_{i=1}^{f_s} C_{S_{si}\mu, N}^{\mathbf{K}} \\
&= \frac{\sqrt{f}}{f_s} \sum_{s\mu, i=1}^{f_s} \Phi_{[NC]s\mu}^{\mathbf{K}} C_{S_{si}\mu, N}^{\mathbf{K}}
\end{aligned}$$

Hence, we get the coefficients in the NC Bloch sum representation

$$C_{[NC]s\mu, N}^{\mathbf{K}} = \frac{\sqrt{f}}{f_s} \sum_{j=1}^{f_s} C_{S_{sj}\mu, N}^{\mathbf{K}} \tag{2}$$

and

$$\Psi_N^{\mathbf{K}} \approx \sum_{s\mu} \Phi_{[NC]s\mu}^{\mathbf{K}} C_{[NC]s\mu, N}^{\mathbf{K}}$$

Now, we can write the Bloch spectral density in terms of these approximate Bloch sums

$$\hat{A} \approx \sum_{KN} \sum_{s\mu, \bar{s}\bar{\mu}} \Phi_{[NC]s\mu}^{\mathbf{K}} \left[ \frac{f}{f_s f_{\bar{s}}} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_{\bar{s}}} n_{S_{sj}\mu, \bar{S}_{\bar{j}\bar{\mu}}}^{\mathbf{K}N}(\omega) \right] \Phi_{[NC]\bar{s}\bar{\mu}}^{\mathbf{K}} \tag{3}$$

The un-approximated Bloch spectral function fulfills

$$\int_{-\infty}^{\infty} A_{\mathbf{K}N}(\omega) d\omega = \int_{-\infty}^{\infty} \text{Tr} \hat{A}_{\mathbf{K}N}(\omega) d\omega = 1$$

with  $(\hat{A}_{\mathbf{KN}} = |\mathbf{KN}\rangle\delta(\omega - \varepsilon_{\mathbf{KN}})\langle\mathbf{KN}|)$ . The Bloch sums yield the overlap matrix via

$$S_{\bar{s}\mu, s\mu}^{\mathbf{K}} = \langle \Phi_{\bar{s}\mu}^{\mathbf{K}} | \Phi_{s\mu}^{\mathbf{K}} \rangle$$

So, we basically get  $\int A = \int \text{Tr} n^K(\omega) S^K$ , which is nothing but the normalization condition  $C^+ S C = 1$  for non-orthogonal eigenvalue problems. Using Eq. (2) it is clear that Eq. (3) is properly normalized if the NC Bloch symmetry is exact. (Note, that then  $f_s = f$ .) If we introduce a symmetrized gross projected method, where we use contragradient Bloch sums  $\tilde{\Phi}^K = \Phi^K (S^K)^{-1}$ ,  $\langle \Phi | \tilde{\Phi} \rangle = 1$  on one side of  $A$  we get the expression

$$\hat{A} = \frac{1}{2} (\Phi n S \tilde{\Phi}^+ + \tilde{\Phi} S n \Phi^+)$$

and the trace give

$$\text{Tr} \underline{A} = \frac{1}{2} \text{Tr} (nS + Sn)$$

Hence, the diagonal part of  $\underline{A} = \frac{1}{2} (nS + Sn)$  gives the unfolded weights and is normalized ( $\text{Tr} \underline{A} = 1$ ), if the NC Bloch symmetry is strictly true. For cases, where atoms got substituted, we have to make a modification to Eq. (1). The  $f$  should be the actual multiplicity of the NC site  $s$  under consideration  $f = f_s$ . This is a definition, which is consistent with any possible choice of NC and also with the cases of partial unfolding due to substitutions. In any case the normalization is proper. Consider the case of perfect NC Bloch symmetry. Then  $n_{[SC]S_{sj}} = \frac{1}{f} n_{[NC]s}$ . Hence (omitting  $S$  for simplicity) the diagonal part of Eq. (3) reads

$$\begin{aligned} A_s &= \frac{f}{f_s f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} n_{[SC]S_{sj} S_{s\bar{j}}} \\ &= \frac{f}{f_s f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} \frac{1}{f} n_{[NC]ss} \\ &= n_{[NC]ss} \end{aligned}$$

And normalization becomes

$$\sum_s \text{Tr}_\mu A_{s\mu} = 1$$

which is fulfilled since it is for the NC expression. If we now have to partially unfold because out of  $f$  sites, which would backfold to a single NC site, several (let's say  $m_s$ ) are occupied by a different atom then the  $f_s = f - m_s$  other sites, we cannot fully contract over all  $f$  sites. But we can contract over  $f_s = f - m_s$  sites and leave the  $m_s$  other atoms uncontracted (means not unfolded). This would restrict the contraction sum to  $f_s \neq f$ . Let's assume that still approximately  $n_{[SC]S_{sj}} = \frac{1}{f} n_{[NC]s}$  (just to discuss the normalization) then we get, letting the first parameter  $f = F$  be choosen later

$$\begin{aligned} A_s &= \frac{F}{f_s f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} n_{[SC]S_{sj} S_{s\bar{j}}} \\ &= \frac{F}{f} n_{[NC]ss} \end{aligned}$$

Besides this we have the  $m_s$  un-unfolded weights of the sites  $s_\lambda$ , which have substituted atoms

$$\begin{aligned} A_{s_\lambda} &= \frac{1}{f} n_{[NC]s_\lambda s_\lambda}, \quad \lambda \in [1, m_s] \\ &\approx \frac{1}{f} n_{[NC]ss}, \quad (\text{if perfect}) \end{aligned}$$

The normalization reads

$$A_s + \sum_{\lambda=1}^{m_s} A_{s_\lambda} = \frac{F + m_s}{f} n_{[NC]ss}$$

and it becomes clear that  $F = f_s$  gives normalization when summed over all sites. Hence our final definition is

$$w_{s\mu}^{\mathbf{KN}}(\omega) = \frac{1}{f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} n_{S_{sj}\mu S_{s\bar{j}}\mu}^{\mathbf{KN}}(\omega) \quad (4)$$

Or in gross projection and dropping the delta function, which was a placeholder for the energy dependence

$$\begin{aligned}
w_{s\mu}^{KN} &= \frac{1}{f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} \frac{1}{2} \left( C_{S_{sj}\mu, N}^{\mathbf{K}} [C^+ S]_{N, S_{s\bar{j}}\mu}^{\mathbf{K}} + [SC]_{S_{sj}\mu, N}^{\mathbf{K}} C_{S_{s\bar{j}}\mu, N}^{\mathbf{K}*} \right) \\
&= \frac{1}{f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} \frac{1}{2} \left( C_{S_{sj}\mu, N}^{\mathbf{K}} [C^+ S]_{N, S_{s\bar{j}}\mu}^{\mathbf{K}} + [C^+ S]_{N, S_{s\bar{j}}\mu}^{\mathbf{K}*} C_{S_{sj}\mu, N}^{\mathbf{K}*} \right), \quad j \leftrightarrow \bar{j}, \text{ in second term} \\
&= \frac{1}{f_s} \sum_{j=1}^{f_s} \sum_{\bar{j}=1}^{f_s} \text{Re} \left( C_{S_{sj}\mu, N}^{\mathbf{K}} [C^+ S]_{N, S_{s\bar{j}}\mu}^{\mathbf{K}} \right)
\end{aligned}$$

Net projection will be discussed below.

## 2.3 Alternative considerations

First let's defined the normal cell problem and derive some symmetries. The orbitals  $\Phi_{rs\mu}$  form Bloch sums

$$\Phi_{s\mu}^{\mathbf{k}} = \frac{1}{\sqrt{N_r}} \sum_r \Phi_{rs\mu} e^{i\mathbf{k}(\mathbf{r}+\mathbf{s})}$$

and wave functions

$$\Psi_n^{\mathbf{k}} = \sum_{s\mu} \Phi_{s\mu}^{\mathbf{k}} C_{s\mu, n}^{\mathbf{k}}$$

where  $C$  is determined by (overlap ignored, since it does not change the results derived here)

$$\begin{aligned}
H^{\mathbf{k}} &= (\Phi^{\mathbf{k}} | H | \Phi^{\mathbf{k}}) \\
H^{\mathbf{k}} C^{\mathbf{k}} &= C^{\mathbf{k}} \varepsilon^{\mathbf{k}}
\end{aligned}$$

At a shifted  $\mathbf{k}$ -vector we get

$$\begin{aligned}
\Phi_s^{\mathbf{k}+\mathbf{g}} &= \Phi_{s\mu}^{\mathbf{k}} e^{i\mathbf{g}(\mathbf{r}+\mathbf{s})} \\
&= \Phi_{s\mu}^{\mathbf{k}} e^{i\mathbf{g}\mathbf{s}}
\end{aligned}$$

and hence

$$\begin{aligned}
H_{ss}^{\mathbf{k}+\mathbf{g}} &= e^{-i\mathbf{g}\bar{\mathbf{s}}} H_{\bar{s}\bar{s}}^{\mathbf{k}} e^{i\mathbf{g}\mathbf{s}} \\
H^{\mathbf{k}+\mathbf{g}} C^{\mathbf{k}+\mathbf{g}} &= C^{\mathbf{k}+\mathbf{g}} \varepsilon^{\mathbf{k}+\mathbf{g}} \\
H^{\mathbf{k}} (e^{i\mathbf{g}\mathbf{s}} C^{\mathbf{k}+\mathbf{g}}) &= (e^{i\mathbf{g}\mathbf{s}} C^{\mathbf{k}+\mathbf{g}}) \varepsilon^{\mathbf{k}+\mathbf{g}}
\end{aligned}$$

which says that the expression in parenthese is also an eigenvector for  $\mathbf{k}$  and hence

$$\varepsilon^{\mathbf{k}+\mathbf{g}} = \varepsilon^{\mathbf{k}}$$

$$\boxed{C^{\mathbf{k}+\mathbf{g}} = e^{-i\mathbf{g}\mathbf{s}} C^{\mathbf{k}} U^{\mathbf{k}}} \quad (5)$$

where  $U$  mixes only states in degenerate subspaces (degenerate bands along symmetry lines/planes or at band crossings). Now, one can argue that there is always a way to adjust the phases (gauge freedom) of an actually calculated  $C^{\mathbf{k}+\mathbf{g}}$  (solution to the eigenvalue problem) such that the result of a shift by  $\mathbf{g}$  is at most a reordering of band indices, which allows to set  $U = 1$  in our considerations. In essence we are saying that the set over all band indices  $\bigcup_n \Psi_n^{\mathbf{k}+\mathbf{g}}$  equals  $\bigcup_n \Psi_n^{\mathbf{k}}$ . We have periodicity of the sets of bands with respect to reciprocal translations. This is strictly only true for the sets, which happen to form continuous and periodic functions of  $\mathbf{k}$  (yes functions of sets!). If an individual band is followed smoothly (e.g. by the  $\mathbf{k} \cdot \mathbf{p}$ -method) the resulting  $\Psi_n^{\mathbf{k}+\mathbf{g}}$  can become another band  $\Psi_{n'}^{\mathbf{k}}$ .

Now, we describe the same system by introducing a super cell with  $f$  copies of the original cell. This leads to new lattice vectors  $\mathbf{R}$  and sites  $\mathbf{S}_{sj} = \mathbf{r}_j + \mathbf{s}$ , which are formed by shifting the original sites  $\mathbf{s}$  via a set of  $f$  original lattice vectors  $\mathbf{r}_j$ ,  $j \in [1, f]$ . Of course, the set of normal cell lattice vectors is obtained from  $\{\mathbf{r}\} = \bigcup_{j=1}^f \{\mathbf{R} + \mathbf{r}_j\}$ , which also leads to  $\sum_{Rj} F(\mathbf{R} + \mathbf{r}_j) = \sum_r F(\mathbf{r})$ . We also have to require that the BvK torus in both descriptions has the same volume or that the number of normal cell lattice vectors is  $f$ -times

the number of super cell lattice vectors:  $N_r = fN_R$ . Now, we can reformulate the problem in this super cell description by forming Bloch sums with proper super cell translational symmetry

$$\Phi_{[SC]S_{sj}\mu}^{\mathbf{k}} = \frac{1}{\sqrt{N_R}} \sum_R \Phi_{RS_{js}\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{S}_{js})}$$

The corresponding coefficients are  $C_{[SC]S_{sj}\mu,n}^{\mathbf{k}}$ .

This description cannot lead to a different result compared to the normal cell description, since we only changed the artificial choice of a supercell. However, by increasing the matrix size (number of sites per unit cell) we increased the number of eigen solutions or bands per  $\mathbf{k}$ -point. In order to get the same number of physically different solutions the size of the reciprocal unit cell must be  $f$ -times smaller than the NC reciprocal unit cell. Well, as everyone knows the excess solutions are bands, which got backfolded into this smaller reciprocal cell. To understand this mathematically it suffices to note that we already showed that in every unit cell the sets of bands form periodic and continuous functions. Hence, the SC wave functions are periodic with respect to translations by reciprocal lattice vectors  $\mathbf{G}$ :  $\bigcup_n \Psi_n^{\mathbf{k}+\mathbf{G}} = \bigcup_n \Psi_n^{\mathbf{k}}$ . Let's construct the  $f \cdot m$  SC solutions out of the  $m$  NC solutions. The first  $m$  solutions are just identical to the NC solutions  $\bigcup_{n=1}^m \Psi_{[SC]n}^{\mathbf{k}} = \bigcup_{n=1}^m \Psi_{[NC]n}^{\mathbf{k}}$ . Now, for every reciprocal vector  $\mathbf{G}$  the set  $\bigcup_{n=1}^m \Psi_{[NC]n}^{\mathbf{k}+\mathbf{G}}$  is either equivalent to  $\bigcup_{n=1}^m \Psi_{[NC]n}^{\mathbf{k}}$  ( $\mathbf{G}$  is a NC vector  $\mathbf{g}$ ) or forms a new backfolded set in which case there are exactly  $f - 1$  different additional sets. We identify  $f - 1$  representative vectors  $\mathbf{G}_l \notin \{\mathbf{g}\}$ ,  $l \in [1, f - 1]$  and  $\mathbf{G}_0 = 0$  and define the set of  $f \cdot m$  SC solutions via  $\bigcup_{n=1}^m \Psi_{[SC]n+lm}^{\mathbf{k}} = \bigcup_{n=1}^m \Psi_{[NC]n}^{\mathbf{k}+\mathbf{G}_l}$ ,  $\forall l \in [0, f - 1]$ . This is just fancy talk for all the backfolding. Beware, that in a real calculations all coefficients  $C$  contain random phase factors from the eigenvalue solver, which means that the construction above deviates in practice from the actual solutions by phases and unitary mixing of degenerate bands.

However, now we can go a step further by identifying equivalences between coefficients. Using the construction of the SC sites discussed above we can write

$$\begin{aligned} \sum_S \Phi_{[SC]S_{sj}\mu}^{\mathbf{k}} &= \frac{1}{\sqrt{N_R}} \sum_{RS} \Phi_{RS_{js}\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{S}_{js})} \\ &= \frac{1}{\sqrt{N_R}} \sum_{Rjs} \Phi_{Rr_js\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{s}+\mathbf{r}_j)} \\ &= \frac{\sqrt{f}}{\sqrt{N_r}} \sum_{rs} \Phi_{rs\mu} e^{i\mathbf{k}(\mathbf{r}+\mathbf{s})} \end{aligned}$$

which leads to the SC expression

$$\begin{aligned} \Psi_{[SC]n+lm}^{\mathbf{k}} &= \sum_{S\mu} \Phi_{[SC]S\mu}^{\mathbf{k}} C_{[SC]S\mu,n+lm}^{\mathbf{k}} \\ &= \frac{1}{\sqrt{N_R}} \sum_{RS\mu} \Phi_{RS\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{S})} C_{[SC]S\mu,n+lm}^{\mathbf{k}} \\ &= \frac{\sqrt{f}}{\sqrt{N_r}} \sum_{Rjs\mu} \Phi_{Rr_js\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{r}_j+\mathbf{s})} C_{[SC]S_{js}\mu,n+lm}^{\mathbf{k}} \end{aligned}$$

and the NC expression, whose equivalence via backfolding we established in the argument above.

$$\begin{aligned} \Psi_{[SC]n+lm}^{\mathbf{k}} &= \Psi_{[NC]n}^{\mathbf{k}+\mathbf{G}_l} \\ &= \sum_{s\mu} \Phi_{[NC]s\mu}^{\mathbf{k}+\mathbf{G}_l} C_{[NC]s\mu,n}^{\mathbf{k}+\mathbf{G}_l} \\ &= \frac{1}{\sqrt{N_r}} \sum_{rs\mu} \Phi_{rs\mu} e^{i(\mathbf{k}+\mathbf{G}_l)(\mathbf{r}+\mathbf{s})} C_{[NC]s\mu,n}^{\mathbf{k}+\mathbf{G}_l} \\ &= \frac{1}{\sqrt{N_r}} \sum_{Rjs\mu} \Phi_{Rr_js\mu} e^{i(\mathbf{k}+\mathbf{G}_l)(\mathbf{R}+\mathbf{r}_j+\mathbf{s})} C_{[NC]s\mu,n}^{\mathbf{k}+\mathbf{G}_l} \\ &= \frac{1}{\sqrt{N_r}} \sum_{Rjs\mu} \Phi_{Rr_js\mu} e^{i\mathbf{k}(\mathbf{R}+\mathbf{r}_j+\mathbf{s})} e^{i\mathbf{G}_l(\mathbf{r}_j+\mathbf{s})} C_{[NC]s\mu,n}^{\mathbf{k}+\mathbf{G}_l} \end{aligned}$$

By comparison with the SC expression we identify

$$\begin{aligned}
C_{[SC]S_{js}\mu, n+lm}^{\mathbf{k}} &= \frac{1}{\sqrt{f}} e^{i\mathbf{G}_l \mathbf{S}_{sj}} C_{[NC]s\mu, n}^{\mathbf{k}+\mathbf{G}_l} \\
&= \frac{1}{\sqrt{f}} e^{i\mathbf{G}_l \mathbf{r}_j} e^{i\mathbf{G}_l \mathbf{s}} C_{[NC]s\mu, n}^{\mathbf{k}+\mathbf{G}_l}
\end{aligned} \tag{6}$$

which is (up to unitary mixing, which we neglected) the exact mapping between the solutions of the two equivalent descriptions. Now, we average the SC coefficients over the equivalent sites with respect to NC periodicity ( $j$ -sum with  $f_s$  terms).

$$\frac{1}{f_s} \sum_j C_{[SC]S_{js}\mu, n+lm}^{\mathbf{k}} = \frac{1}{\sqrt{f}} \frac{1}{f_s} \left( \sum_j e^{i\mathbf{G}_l \mathbf{r}_j} \right) e^{i\mathbf{G}_l \mathbf{s}} C_{[NC]s\mu, n}^{\mathbf{k}+\mathbf{G}_l}$$

The sum in parentheses runs over all NC lattice vectors  $\mathbf{r}_j$ , which are needed to make the whole NC lattice from the SC lattice vectors  $\mathbf{R}$ . We can introduce a basis in the real space lattice via

$$\begin{aligned}
\mathbf{R} &= \sum_{I=1}^3 R_I \mathbf{A}_I \\
\mathbf{r} &= \sum_I r_I \mathbf{a}_I
\end{aligned}$$

where in order for the SC to be a commensurate multiple of the NC the relation  $\mathbf{A}_I = \mathbf{a}_J M_{JI}$ ,  $M_{JI} \in \mathbb{N}$  must hold. The reciprocal lattice then also has a basis

$$\begin{aligned}
\mathbf{G} &= \sum_I G_I \mathbf{B}_I \\
\mathbf{g} &= \sum_I g_I \mathbf{b}_I
\end{aligned}$$

with the defining relation of the reciprocal basis  $\mathbf{B}_I \mathbf{A}_J = \mathbf{b}_I \mathbf{a}_J = 2\pi \delta_{IJ}$ . Hence  $\mathbf{B}_I \mathbf{a}_K M_{KJ} = 2\pi \delta_{IJ}$  or  $\mathbf{B}_I \mathbf{a}_J = 2\pi (M^{-1})_{IJ}$

$$\begin{aligned}
\mathbf{G}_l \mathbf{r}_j &= \sum_I G_{l,I} \mathbf{B}_I \cdot \sum_J r_{j,J} \mathbf{a}_J \\
&= \sum_{IJ} G_{l,I} r_{j,J} \mathbf{B}_I \cdot \mathbf{a}_J \\
&= 2\pi \sum_{IJ} G_{l,I} r_{j,J} (M^{-1})_{IJ}
\end{aligned}$$

The inverse of an integer matrix  $M$  is a matrix of rational numbers. The coefficients  $G_{l,I} r_{j,J}$  in the equation above are integer in such a way that if  $\mathbf{G}_l$  were a reciprocal lattice vector  $\mathbf{g}$  of the NC  $G_{l,I} (M^{-1})_{IJ} \in \mathbb{N}$  must be an integer because  $\mathbf{g}\mathbf{r} = 2\pi h$ ,  $h \in \mathbb{N}$  always holds for dual lattices. Hence for the non trivial  $\mathbf{G}_l$ , which actually backfold the original bands onto new bands  $G_{l,I} (M^{-1})_{IJ}$  must be rationals. On the other hand  $r_{j,J}$  are also integer but chosen such that they do not represent SC lattice vectors, since otherwise  $r_{j,J} (M^{-1})_{IJ}$  must be integer for  $\mathbf{G}\mathbf{R} = 2\pi H$ ,  $H \in \mathbb{N}$ . Altogether, when thinking hard one realizes that  $\mathbf{G}_l \mathbf{r}_j$  runs over  $f$  fractionals such that the resulting  $f$  complex numbers  $e^{i\mathbf{G}_l \mathbf{r}_j}$ ,  $j \in [1, f]$  are equally spaced on the unit circle including the number 1 (for  $\mathbf{r}_j = 0$ ). But then a general sum of unity theorem tells us  $\sum_j e^{i\mathbf{G}_l \mathbf{r}_j} = f_s \delta_{\mathbf{G}_l \mathbf{g}}$ , where any NC reciprocal vector  $\mathbf{g}$  will work. If we do partial backfolding, where  $f_s < f$  the sum of unity is no longer correct, but one can argue that we get something with  $f_s$  terms of order one and an approximate delta function.

Hence, the average becomes (using Eq. (5))

$$\frac{1}{f_s} \sum_j C_{[SC]S_{js}\mu, n+lm}^{\mathbf{k}} = \frac{1}{\sqrt{f}} \delta_{\mathbf{G}_l \mathbf{g}} C_{[NC]s\mu, n}^{\mathbf{k}} U^{\mathbf{k}} \tag{7}$$

The unitary mixing only happens in degenerate subspaces and drops out of the final weight expressions if an average over the degenerate bands is taken.

In words: if the bands are from the non backfolded set,  $l = 0$ ,  $\mathbf{G}_l = \mathbf{g}$  we get  $C_{[SC]} = \frac{1}{\sqrt{f}} C_{[NC]}$ . If it is a backfolded set  $l \neq 0$  and  $\mathbf{G}_l \neq \mathbf{g}$  we get exactly (approximately) zero. Hence, the average defined above is shown to differentiate between backfolded and original bands. Of course one can continue this relation to  $\mathbf{k}$ -points outside of the first SC reciprocal unit cell and hence recover the full band structure of the larger NC unit cell.

What is left is the normalization condition. We will discuss net weights in order to stay consistent with the FPLO scheme of things. The sum over all standard SC weights must equal one. Using Eq. (6) we can write

$$\begin{aligned}
1 = \sum_{S\mu} w_{S\mu}^{k,n+lm} &= \frac{1}{\Omega} \sum_{S\mu} \left\langle \left| C_{[SC]S\mu,n+lm}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n} \\
&= \frac{1}{\Omega f} \sum_{S\mu} \left\langle \left| C_{[NC]S\mu,n}^{\mathbf{k}+\mathbf{G}_l} \right|^2 \right\rangle_{\deg n} \\
&= \frac{1}{\Omega f} \sum_{jS\mu} \left\langle \left| C_{[NC]S\mu,n}^{\mathbf{k}+\mathbf{G}_l} \right|^2 \right\rangle_{\deg n} \\
&= \frac{1}{f\Omega} \sum_{s\mu} f_s \left\langle \left| C_{[NC]s\mu,n}^{\mathbf{k}+\mathbf{G}_l} \right|^2 \right\rangle_{\deg n}
\end{aligned}$$

The proposed unfolded weights using the  $r_j$  averages read (with an undetermined factor  $X_s$ )

$$\begin{aligned}
w_{s\mu}^{kn+lm} &= X_s \left\langle \left| \frac{1}{f_s} \sum_j C_{[SC]S_{js\mu},n+lm}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n} \\
&= X_s \frac{1}{f} \left\langle \left| C_{[NC]s\mu,n}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n} \delta_{\mathbf{G}_l, \mathbf{g}}
\end{aligned}$$

The normalization sum reads

$$1 = \sum_{s\mu} w_{s\mu}^{kn+lm} = \frac{1}{f} \sum_{s\mu} X_s \left\langle \left| C_{[NC]s\mu,n}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n} \delta_{\mathbf{G}_l, \mathbf{g}}$$

Comparing to the sum of SC net weights above assuming that we have an original band ( $\delta = 1$ ) we get  $X_s = \frac{f_s}{\Omega}$  and the final normalized unfolded net weight result is

$$\boxed{w_{s\mu}^{kn} = \frac{\frac{1}{f_s} \left\langle \left| \sum_j C_{[SC]S_{js\mu},n}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n}}{\sum_{S\mu} \left\langle \left| C_{[SC]S\mu,n}^{\mathbf{k}} \right|^2 \right\rangle_{\deg n}}} \quad (8)$$

Finally, after having discussed everything for the exact supercell, we postulate that the same procedure is also applicable for slightly distorted supercells, which gives the unfolding procedure.

## 2.4 Summary:

Band unfolding is a fat-band method. By projecting the Bloch spectral density operator onto Bloch sums of the NC periodicity, the resulting weights will be strongest for the bands, which belong to the original NC band structure. The other bands of the SC, which are obtained by backfolding NC bands, will have smaller or zero weight, depending on the amount of perturbation, which differentiates the SC from a perfect duplication of NC cells. The philosophy of unfolding contradicts atom substitution. However, atom substitution can be handled too at least formally.

## 3 Perturbations

There are two kinds of perturbations, moving atoms and replacing atoms. Moving atoms means that the overlap matrix between the NC and the SC orbitals/WFs gets approximated in this unfolding technique and that the phase factors of the NC Bloch states are only correct on average, which usually is a small thing, if atoms are moved only slightly. However, one should keep in mind that the projections used assume perfect matching (which only exists in perfect multiples of NC). Moving physically unimportant atoms (no contribution to the energy window under consideration, e.g. Fermi level) is off course a potential-only perturbation for the important atoms (which contribute to the energy window) and hence unfolding for the important atoms makes sense.

Replacing atoms comes in two modes. Replacing unimportant atoms (charge donors, buffer atoms) really just changes the potential of the important (other) atoms and the unfolding for the important atoms is rather

meaningful. Example: replacing cations in the pnictides while leaving the FeAs planes intact, gives meaningful unfolding for the Fe bands around the Fermi level. Replacing the important atoms gives problems for the following reason. Assume the NC and SC as shown in Figure 2a.

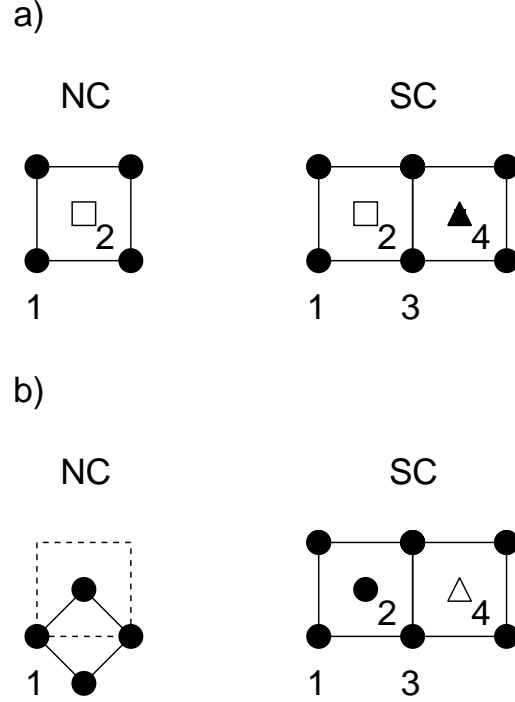


Figure 2: Example for replacing atoms

The sites  $s = 1$  gets unfolded by equating sites  $S = 1, 3$ . This will lead to nicely unfolded bands. The unfolding of sites  $S = 2, 4$  cannot be done in the technique used here, since the atoms and hence the basis of these atoms are different and there is no way of forming approximate Bloch sums of sites  $S = 2$  and  $S = 4$ . Instead one has to request unfolding with site list  $S = 2$  and  $S = 4$  separately. This just means that the unfolded band weights of these two sites are identical to the not-unfolded weights and hence show the SC periodicity (backfolding in the BZ). This makes some sense, since there really is no approximate NC symmetry for these atoms. The total unfolded weight (sum over all orbitals) for unfolded  $S = 2$  is half as big as the total unfolded weight of  $S = 1, 3$  due to the site count. In that sense the unfolding is still visible to a certain extend. In Figure 2b the situation is a bit better. Now, unfolded site weights can be defined from  $S = 1, 2, 3$  and  $S = 4$ . The weights will now play out better, since only one out of four sites, which form a full NC Bloch sum is missing. So we can plot the  $S = 1, 2, 3$  unfolded weights and the unfolded=not-unfolded  $S = 4$  bands.

## 4 Brillouine zones

The NC bz is larger than the SC BZ. Example from Figure 2a: the bz is shown in Figure 3.

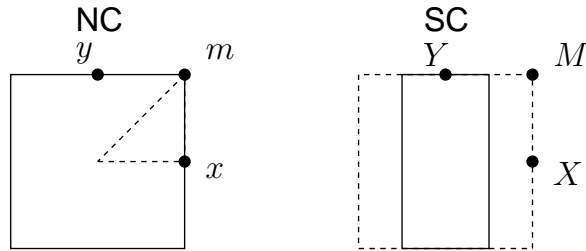


Figure 3: bz and BZ of Figure 2

In the NC we have the high symmetry points

$$\begin{aligned} x &= \left( \frac{\pi}{a_{[NC]}}, 0, 0 \right) = \frac{2\pi}{a_{[NC]}} \left( \frac{1}{2}, 0, 0 \right) \\ m &= \left( \frac{\pi}{a_{[NC]}}, \frac{\pi}{a_{[NC]}}, 0 \right) = \frac{2\pi}{a_{[NC]}} \left( \frac{1}{2}, \frac{1}{2}, 0 \right) \\ y &= \left( 0, \frac{\pi}{a_{[NC]}}, 0 \right) = \frac{2\pi}{a_{[NC]}} \left( 0, \frac{1}{2}, 0 \right) \end{aligned}$$

In the SC the equivalent points are at the same cartesian coordinates, but using  $a_{[SC]} = 2a_{[NC]}$  we get

$$\begin{aligned} X &= \left( \frac{2\pi}{a_{[SC]}}, 0, 0 \right) = \frac{2\pi}{a_{[SC]}} (1, 0, 0) \\ M &= \left( \frac{2\pi}{a_{[SC]}}, \frac{2\pi}{a_{[SC]}}, 0 \right) = \frac{2\pi}{a_{[SC]}} (1, 1, 0) \\ y &= \left( 0, \frac{2\pi}{a_{[SC]}}, 0 \right) = \frac{2\pi}{a_{[SC]}} (0, 1, 0) \end{aligned}$$

Although, only the  $x$ -direction changes in the new BZ, all fedit coordinates change. This is because the cartesian coordinates are the same in NC and SC, but the unit  $\frac{2\pi}{a_{[SC]}}$  scales for all directions.

## 5 Caveats

- The unfolded weights might look much smaller than the weights in the NC along some directions. This can happen, when the bands are degenerate in the NC and is due to a splitting in the SC combined with the way of counting. So, a width 1 band becomes, let's say, 2 width 0.5 bands shifted slightly against each other. Now, they appear to be half as broad, when the splitting is small and hence the band are more or less plotted on top of each other.
- Take care, when determining the NC bz in the SC setup.
- Unfolded weights do not show the full information, because the Bloch spectral density is not diagonal in the approximate NC Bloch states.

## 6 User input/output

The user defines unfolding by creating the file `=.unfold`. If the file is detected the fatbands (`+bweights_unfolded/`  
`+bweights_kp_unfolded`) are created according to formula 8.

### 6.1 `=.unfold`

The file looks like this

```
# NCsite SCsites
1          1 2 3
# some comment
2          4
```

There can be any number of comment lines starting with '#', or lines only containing whitespace. They are ignored.

Each line, which is not a comment line and not whitespace-only is an unfolding definition.

An unfolding definition contains

**the NC site number:** which has no other use than labeling the contracted atoms in the label information in `+bweights_unfold`. Maybe it's best to use the actual site number in the NC.

**the SC site list:** which is the list of sites in the SC, which get contracted onto the NC site.

The number of SC sites, which are contracted to this NC site (in the formulas above it is  $f_s$ ) is determined from the input. Usually it equals the number of times the NC fits into the SC  $f_s = f$ . In cases where atom substitutions took place  $f_s < f$  as in the example file above: 4 NC cells form one SC, hence  $f = 4$ . Atom 4 got substituted and has a different atom than sites 1...3. So, we contract/unfold sites 1...3 and leave site 4 by itself. Note, that one can contract different atoms, as long as they have the same basis, i.e. in most VCA cases and when substituting similar elements (e.g. Fe with Ni).

## 6.2 choosing k-points

Beware of choosing the correct  $k$ -points. The unfolded fatbands are calculated in the SC BZ. So, first you determine the cartesian representation of the special points in the NC bz. Then you find the relation between bz and BZ and finally transform the bz points into BZ points and put them into the fedit menu in units of  $\frac{2\pi}{a_{SC}}$ . That means that a  $k$ -point in cartesian coordinates  $\vec{k}$  is entered as  $\tilde{k}$  in fedit with  $\vec{k} = \frac{2\pi}{a}\tilde{k}$ , where  $a$  is the first lattice constant.

## 6.3 Fermi surfaces

Fermi surfaces can be created with xfsf. Xfsf uses the symmetry to create an irreducible mesh and hence to save calculational time. The NC bz is larger than the SC BZ. Therefore, it is impossible to create enough NC  $k$ -points by using the SC BZ. There is a menu input->handmade symmetry, which allows the user to enter the NC cell and symmetry operations (only some generators are needed). This overwrites the default mesh symmetry. After this the process is straight forward, except for the file suffix, of the unfolded fatbands, which requires to set a non-default filename in input->files. Note that the new convention is to name the band file **+band\_kp** for the Fermi surface (in general if **=.kp** was used), in order to not overwrite **+band**. So, the unfolded file is **+bweights\_kp\_unfold**.

The bandweights can be used for coloring: plot->coloring: extern, state. The extern checkbox switches from Fermi velocity coloring to extern coloring from the file given in input->files. State selects the data column from the file. (Transparent coloring as in Ref. [3] is not officially available, since it was a hard coded hack.)

## References

- [1] Technical report, <http://www.fplo.de>.
- [2] Wei Ku, Tom Berlijn, and Chi-Cheng Lee. Unfolding first-principles band structures. *Phys. Rev. Lett.*, 104(21):216401, May 2010.
- [3] Erik van Heumen, Johannes Vuorinen, Klaus Koepernik, Freek Massee, Yingkai Huang, Ming Shi, Jesse Klei, Jeroen Goedkoop, Matti Lindroos, Jeroen van den Brink, and Mark S. Golden. Existence, character, and origin of surface-related bands in the high temperature iron pnictide superconductor *bafe2 - xcozas2*. *Phys. Rev. Lett.*, 106(2):027002, Jan 2011.