Supplementary material

Bowen Hou, Jinyuan Wu, Xingzhi Sun, Smita Krishnaswamy, and Diana Y. Qiu

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$$M_{nn'}(\mathbf{k},\mathbf{q},\mathbf{G}) = \langle n\mathbf{k}+\mathbf{q}|\mathrm{e}^{\mathrm{i}(\mathbf{q}+\mathbf{G})\cdot\mathbf{r}}|n'\mathbf{k}\rangle \tag{1}$$

$$\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega=0) = \sum_{\mathbf{k}}^{\text{occ}}\sum_{n}^{\text{emp}}\sum_{n'} M_{nn'}(\mathbf{k},\mathbf{q},\mathbf{G})M^*_{nn'}(\mathbf{k},\mathbf{q},\mathbf{G}')\frac{2}{E_{n\mathbf{k}+\mathbf{q}}-E_{n'\mathbf{k}}} \tag{2}$$

In the conventional pseudobands technique, the empty states are divided into one low-energy protected subspace and a series of pseudobands blocks; each of the blocks contain bands with comparable energies. The high-energy terms in $\chi_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega=0)$ are therefore replaced by

$$\chi^{\text{P.B. blocks}}_{\mathbf{G}\mathbf{G}'}(\mathbf{q},\omega=0) = \sum_{\mathbf{k}}^{\text{P.B. terms}}\sum_{S}\frac{2}{E_{n\mathbf{k}+\mathbf{q}}-\bar{E}_S}\sum_{n_1',n_2'}^{S}\sum_{n}^{\text{occ}} M_{nn_1'}(\mathbf{k},\mathbf{q},\mathbf{G})M^*_{nn_2'}(\mathbf{k},\mathbf{q},\mathbf{G}'). \tag{3}$$

Because the energies of states in each pseudobands block are comparable, in each pseudobands block, the states share the same set of predominant $\mathbf{G}$ vectors. Thus the states in pseudobands block S can be written as

$$\langle\mathbf{r}|n'\mathbf{k}\rangle = \sum_{i}^{N} c^{(i)}_{n'\mathbf{k}}\mathrm{e}^{\mathrm{i}(\mathbf{k}+\mathbf{G}^{(i)}_{S\mathbf{k}})\cdot\mathbf{r}}, \quad \sum_{i}^{N} c^{(i)}_{n_1'\mathbf{k}}c^{(i)*}_{n_2'\mathbf{k}} = \delta_{n_1'n_2'}, \tag{4}$$

and therefore

$$M_{nn'}(\mathbf{k},\mathbf{q},\mathbf{G}) = \sum_{i}^{N} c^{(i)}_{n'\mathbf{k}}c^{*}_{n\mathbf{k}+\mathbf{q}}(\mathbf{G}+\mathbf{G}^{(i)}_{S\mathbf{k}}). \tag{5}$$

The pseudobands approximation of the diagonal $\chi_{\mathbf{G}\mathbf{G}}$ components of the polarizability is proportional to

$$\begin{aligned}
&\sum_{n}^{\text{occ}} M_{nn_1'}(\mathbf{k},\mathbf{q},\mathbf{G})M^*_{nn_2'}(\mathbf{k},\mathbf{q},\mathbf{G}) \\
&= \sum_{i,j}^{N} c^{(i)}_{n_1'\mathbf{k}}c^{(j)*}_{n_2'\mathbf{k}}\sum_{n}^{\text{occ}} c^{*}_{n\mathbf{k}+\mathbf{q}}(\mathbf{G}+\mathbf{G}^{(i)}_{S\mathbf{k}})c_{n\mathbf{k}+\mathbf{q}}(\mathbf{G}+\mathbf{G}^{(j)}_{S\mathbf{k}}) \\
&\approx \sum_{i,j}^{N} c^{(i)}_{n_1'\mathbf{k}}c^{(j)*}_{n_2'\mathbf{k}}\delta_{ij} = \delta_{n_1'n_2'}.
\end{aligned} \tag{6}$$

In the third step we argue that the summation over the occupied states leads to an approximate orthogonal relation, because the subspace spanned by the dominant $\mathbf{G}$ components of the occupied states is expected to largely overlap with the subspace of the occupied states. Therefore, the unwanted non-diagonal terms in (3) can be ignored when $\mathbf{G}=\mathbf{G}'$. The

which justifies the validity of the pseudobands technique for high-energy bands.