Detailed comments on manuscript by Esteve-Paredes and Placios

I list below some comments and criticsms, some of which are simple matters of presentation, while others go a little deeper.

1. I don't really understand why the manuscript puts so much emphasis on the result of Gu et al., Eq. (1). I was unfamiliar with this result, and had a hard time understanding at first what it was about.

   (a) The sentence below Eq. (1) suggested to my mind a finite crystallite of volume v with physical surfaces, so I had trouble understanding why the eigenstates were not "decaying at the boundaries" (vacuum tails). The authors should explain at the outset that the volume in question is just a mathematical construction of an $N\times N\times N$ supercell inscribed in the bulk. But just below Eq. (1) and later in Sec. II.D, it is clear that this expression is not true for general wave functions, or even for arbitrary functions of Bloch form; it is only true for Bloch states (eigenstates of H). Also, it seems only to be true for $H'=0$ (absence of nonlocal terms in the potential). The authors also point out that it involves an awkward set of surface integrals. I wonder if this entire discussion could be deemphasized, or moved to an Appendix.

   (b) I have a few comments about Eqs. (11-12) and the surrounding discussion. First, isn't it true that $\langle n\boldsymbol{k}|\boldsymbol{v}|n'\boldsymbol{k}'\rangle_v$ in Eq. (1) contains a $\delta_{\boldsymbol{k}\boldsymbol{k}'}$? Why don't I see this reflected in Eqs. (10-12)? Shouldn't we be writing expressions for quantities like $\langle n\boldsymbol{k}|\boldsymbol{v}|n'\boldsymbol{k}\rangle_v$ instead? Are the two terms in Eq. (11) individually nonzero in Eq. (11), but they cancel for $\boldsymbol{k}\neq\boldsymbol{k}'$? I find it confusing.

   (c) Also, can any intuitive physical interpretation be given to Eqs. (11) and (12)? The latter looks like a current flow across the boundary, doesn't it? I suppose that for diagonal elements $\langle n\boldsymbol{k}|\boldsymbol{v}|n\boldsymbol{k}\rangle_v$, the first term in (11) vanishes, right?

   (d) I suspect Eqs. (11-12) can be recast in terms of integrals over a the interior and the boundary of a single primitive cell, with wave functions normalized to the unit cell, and with an average over $\boldsymbol{k}$, where $\boldsymbol{k}$ runs (as now) over the $N\times N\times N$ k-space mesh of the primitive Brillouin zone. It seems to me that this would be a more natural way to present the result.

   (e) Minor additional comment about Eq. (12): the final $\boldsymbol{r}$ is difficult to notice, but is crucial. Maybe there could be a way to call the reader's attention to its presence.

2. I found Eq. (7) and its discussion to be confusing. It seems to me that Eq. (7) is simply true without the last correction term. I think the authors have to find

a way to be more explicit about what they really mean. For example, let P_b be the projection operator onto the basis ("almost unity"), and define objects like $\hat{H}' = P_\mathrm{b}\,\hat{H}P_b$, $\hat{p}' = P_\mathrm{b}\,\hat{p}P_b$, $\hat{r}' = P_\mathrm{b}\,\hat{r}P_b$, $\hat{v}' = P_\mathrm{b}\,\hat{v}P_b$, etc. In this notation, I think what the authors are trying to say is that if the operators are replaced by their primed versions in Eq. (7), the Δ correction term is needed. Is that the idea? It needs to be clearer. By the way, I think there is a typo 7 lines below Eq. (7), where it should be $\mathrm{Tr}(r_\alpha, p_\beta)$=$\mathrm{Tr}(p_\beta, r_\alpha)$ (when using primed operators in my notation).

3. There is some confusing notation in Eq. (5). Putting $O(\boldsymbol{r})$ inside the integrand seems to suggest the O is a local operator, but clearly this is not the case, or else $\hat{O}_{\boldsymbol{k}}$ would be the same as $\hat{O}$. I suppose what is meant would correspond to replacing $\hat{O}(\boldsymbol{r})\psi_{n'k'}(\boldsymbol{r})$ by $[\hat{O}\psi_{n'k'}](\boldsymbol{r})$, or more pedantically, by $\langle \boldsymbol{r}|\hat{O}|\psi_{n'k'}\rangle$.

4. Shortly after Eq. (5), the wording "... such that $O(\boldsymbol{r})\psi_{nk}(\boldsymbol{r})$ still satisfies the Bloch theorem" is misleading. There was at least one more similar misuse later in the manuscript. Bloch's theorem is a theorem about the eigenstates of a Hamiltonian, which this wave function is not. The correct wording would instead be something like "of Bloch form". Even better would be to discuss whether $\hat{O}$ is a periodic operator, i.e., one that commutes with crystal translations; if so, then the product of such an operator with a wave function of Bloch form is automatically a wave function of Bloch form.

5. Another awkward detail of notation is the appearance of superscripts (v) and (n) in Eq. (19) and elsewhere. It looks as though the notation is parallel, whereas in fact the meanings of these superscripts are completely different.

6. I think the LHS of the first Eq. (25) should be $\boldsymbol{A}_{\alpha\alpha'}(\boldsymbol{k})$. Also, the asymmetry between the two Eqs. (25) "looks wrong" although I think it is actually correct. It may be worth a few words of explanation. Perhaps it may be worth emphasizing that in the case of a nonorthogonal basis, $\boldsymbol{A}$ is not Hermitian, as it is in the orthonormal case. It also may be a good idea to explicitly write an expression for the last term in Eq. (44) in a language parallel to the first term; I guess it comes to something like $\sum_{\boldsymbol{R}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}}\boldsymbol{R}\langle\alpha 0|\alpha'\boldsymbol{R}\rangle$, doesn't it?

7. The choice of the acronym CREN is not explained; what does it stand for?