Dear Dr. Attaccalite,

We kindly thank the referee for their additional comments. Please find below a series of responses to the questions posed, as well as direction to locations in the manuscript where changes have been made. We hope that our manuscript can now be published.

Best,
PTM and JES

Dear editor,

I thanks the authors for their answers to my questions and comments and for the revised manuscript. I think the manuscript can be accepted as an article in SciPost Physics if the authors take into account the following questions and comments

1)

In their answer the authors write

"We do agree with the referee that model calculations implementing our formalism would provide some physical insight. However, because ELWFs are central to our formalism, and because such functions are not provided in simple tight-binding models appearing in the literature (except, e.g., approximating them as delta functions), there is no simple calculation we can do at this time. "

However in the Introduction they write

"And a fifth reason is that, with its emphasis on ELWFs and the interest in those functions for electronic structure and response calculations in general, we can hope that the approach here will be useful in numerical calculations."

These the remarks seem to be in contradiction.

Either ELWF are simple to implement and then the authors could have done so to add some numerical results to this work or they are not simple to implement and the usefulness of the authors' approach is in doubt.
It would be good if the authors clearly state in the manuscript how their approach can be used in practice and what would be the difficulties for a practical implementation in a computer code.

We thank the referee for this comment. What we mean is that, for a practitioner of DFT-type computer codes, which we ourselves are not, the implementation of our formalism is typically straightforward. In particular, with a set of ELWFs and some Bloch energy eigenvectors and eigenvalues in-hand, the numerical evaluation of, for example, Eq. (24)-(27), (28), and (32) (of

the modified manuscript, which were previously numbered Eq. (26)-(29), (30), and (34)) can be made. Depending on the quantity of interest, it may be easier to evaluate the expressions written as multipole moments of ELWFs or doing the evaluation entirely in k-space. For example, in Eq. (24) diagonal matrix elements of the Berry connection appear, which can be difficult to obtain numerically (in contrast to off-diagonal elements which can be found from velocity matrix elements). But, when written in terms of ELWFs, the computation should be possible. We have added the "mixed" form of (24) above it as an unnumbered equation as well as the text beginning "Although the matrix elements appearing in …" just before the start of Sec. III A to illustrate this point. We mention again that while we feel the numerical evaluation of these quantities would be interesting, and that we plan to undertake such computation in future works, the actual values that result do not hold precise physical meaning as they are gauge dependent (i.e., depend on the choice of ELWFs). In general, the physically meaningful quantities appearing here are Eq. (33), (34), (35), which do not require ELWFs to calculate and agree with known results.

2)

In the Introduction the authors write

"Here, polarization and magnetization fields, and free charge and current densities serve as intermediary quantities that aid calculation and provide physical insight, but in general only the appropriate combinations that lead to the charge and current densities have direct physical significance."

Could the authors please write explicitly what this physical insight is ?

We thank the referee for this comment. To expound on this point, we have added the footnote starting as "For example, in the linear response of a "topologically trivial" insulator, we have previously shown…" to the end of the sentence the referee refers to.

3)

The authors write

"One reason is that usual calculations made in minimal coupling can require the identification of sum rules to show properly behaved results at low frequencies [23, 24]"

I think Refs 23 and 24 should (at least) be complemented with PRB 82 035104 and PRB 95 155203

We thank the referee for reminding us of these references, which we have now included.

4) In their answer to my question 7 the authors write

“The physical significance here is that, in an ordinary insulator, the entire electronic response to an electric field is due to a modification of the electric polarization about each lattice site, and this is gauge invariant."

The electric polarisation around a lattice site is not a measurable quantity.
So what does “gauge invariance” mean ?

The point is that in the particular case of a trivial insulator, the electronic current density that is linearly induced by a uniform electric field can be entirely associated with an induced electric dipole moment. Since the current response is gauge invariant (in that it is independent of the choice of Bloch energy eigenfunctions, or indeed of the choice of ELWFs employed in our framework assuming that they are taken completely “occupied” or “unoccupied”), so too is that of the electric dipole moment in this case, and thus it is physically significant. In fact, starting from our general definitions, we have previously shown this to be the case for such insulators. Indeed, in order for a general induced multipole moment to be physically measurable, the corresponding susceptibility tensor must be gauge invariant in that it must be insensitive to how the set of ELWFs - with respect to which it is identified – are chosen. This is not typically the case.

5) In their answer to my question 8 the authors write

“We agree that such corrections can be important. In this first work, we neglect such corrections to give room to fully analyze this more simple case, which is indeed not so very simple.”

I think the authors should mention this in the manuscript.

Per the referees suggestion, we have modified a sentence, which now reads “To simplify these initial considerations we here neglect local field corrections, which can be important \cite{Debernardi2012}, and take the applied electric field be the macroscopic Maxwell field.”

6) In their answer to my question 10 the authors write

"We do not claim this feature is unique to our method, only that it is a supporting aspect of it. We emphasize that consistency with works such as those referenced by the referee add support to the definitions we employ, as we arrive at the expected result via a new method. We have added text to the Introduction and refined the Conclusion to make this point more clear. For example, at the end of the Introduction we add the text beginning with “Ultimately, when implemented in a metallic crystal, that our general definitions agree with past work…”."

I think the authors should (at least) mention PRB 86 125139

We thank the referee for reminding us of this reference, which we have now included in the modified sentence listed in response to point 5).

7) In their answer to my question 11 the authors write

"In a subspace of the Hilbert space of square-integrable functions, the position operator is well-defined (e.g., the usual position expectation value in a space of atomic wavefunctions is well-defined). A basis of ELWFs spans such a subspace, while the Bloch functions are not even square-integrable. This allows the position matrix elements in the basis of ELWFs to be well-defined, in particular since they have compact support; See, e.g., Ref [43]."

Do I then correctly understand that the authors' approach does not use PBC?
How then do they propose to model a solid?
Could the authors clarify?

We do apologize for any confusion. To avoid any further issues, including potential confusion of a reader uninitiated to these considerations, we here summarize some past results, and illustrate how they motivate our approach. We also slightly modify the wording in that area of Appendix B in the hope of avoiding such confusion. We stress that our results are valid *only* for a bulk crystalline solid (with periodic boundary conditions) and that in finite metallic systems additional considerations are necessary (for example, those raised in arguments of the "modern theory of polarization.").

The question of the existence of a position operator in an infinite crystalline solid is essentially looking for a linear map $\mathfrak{X}: \mathcal{H} \to \mathcal{H}$, for $\mathcal{H}$ an "electronic Hilbert space." Typically, one thinks of $\mathcal{H}$ as the Hilbert space spanned by the set of (cell-periodic parts of) Bloch energy eigenfunctions $u_{nk}(x)$ associated with some set of isolated energy bands (possibly even all of the energy bands). In any such space, the usual position operator is ill-defined. This follows from the argument (see Resta [9]) that, even in one spatial dimension, the basis vectors of this space satisfy $u_{nk}(x) = u_{nk}(x+R)$ for any Bravais lattice vector R and therefore so does any element of that space $f: \mathbb{R} \to \mathbb{C}$. For any such f, define the usual operator via $(\mathfrak{X}f)(x) \coloneqq xf(x)$. This $\mathfrak{X}$ is linear on that Hilbert space. Then, $(\mathfrak{X}f)(x+R) \coloneqq (x+R)f(x+R) = (x+R)f(x) \neq xf(x) =: (\mathfrak{X}f)(x)$ so $\mathfrak{X}$ is not an operator in that space. However, depending on the topology of the Hilbert bundle constructed using a set of Bloch energy eigenvectors that are associated with an isolated set of energy bands, there may exist a space isomorphic to the above Hilbert space that is spanned by a set of exponentially localized Wannier functions (when all energy bands are included, such a space always exists). In this space, the above definition of $\mathfrak{X}$ results in a genuine operator. First, note that a set of ELWFs are a subset of the set of square integrable functions, and indeed so too is $(\mathfrak{X}W_{\alpha R})(x) \coloneqq xW_{\alpha R}(x)$; $\int |xW_{\alpha R}(x)|^2 \mathrm{d}x = 2R\int W_{\alpha 0}(x)^* x W_{\alpha 0}(x)\mathrm{d}x + \int W_{\alpha 0}(x)^* x^2 W_{\alpha 0}(x)\mathrm{d}x =: 2R\bar{x}_\alpha + \langle x^2 \rangle_\alpha < \infty$, where the inequality follows from the fact that in the construction of ELWFs $W_{\alpha R}(x)$, $\bar{x}_\alpha$ and $\langle x^2 \rangle_\alpha$ are ingredients in the spread functional (see, for example, Eq. (18) of Marzari *et al.* [30]) and are always finite for *exponentially localized* or *almost exponentially localized* Wannier functions. Moreover, $\mathfrak{X}W_{\alpha R}$ remains in the space spanned by the set of ELWFs since one can write

$(\mathfrak{x}W_{\alpha R})(x) = \sum_{\beta R'} c_{\alpha R,\beta R'} W_{\beta R'}(x)$, where, using the orthogonality of the ELWFs, $c_{\alpha R,\beta R'} = \int W_{\beta R'}(x)^* x W_{\alpha R}(x)\mathrm{d}x = \int W_{\beta 0}(x)^* x W_{\alpha R-R'}(x)\mathrm{d}x + R'\delta_{\alpha\beta}\delta_{RR'} = \int_{\mathrm{BZ}} e^{-ik(R-R')}\tilde{\xi}_{\alpha\beta}(k)\mathrm{d}k + R'\delta_{\alpha\beta}\delta_{RR'}$.

For a topologically trivial insulator, the energy eigenvectors associated with the set of occupied energy bands can be mapped to a set of ELWFs (by definition of being "topologically trivial"). In the "modern theory of polarization" – which applies to such bulk insulators – a definition for the electric polarization is given as an integral of the valence band trace of the Berry connection over BZ (Eq. 4.44 of Vanderbilt [2]) or equivalently as the sum of the first moments of a set of ELWFs constructed from completely occupied eigenvectors over all of coordinate space (Eq. 3.95, 3.96 of Vanderbilt [2]). The question addressed by Resta [9] as well as others in more recent works (for example, in Phys. Rev. B 96, 245115), pertains to the definition of multipole operators on the electronic Hilbert space of Bloch functions, *not* on the space of ELWFs where the usual operators work OK. Our work essentially makes use of the fact that in the space of ELWFs such multipole moments are defined, hence we decompose all quantities with respect to a set of ELWFs to identify polarization, magnetization, etc. Once we have these expressions in hand, only then can we map into the Hilbert space of Bloch functions to express our results as a single BZ integral.