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Microscopic polarization and magnetization fields in extended systems

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We introduce microscopic polarization and magnetization fields at each site of an extended system, as well as free charge and current density fields associated with charge movement from site to site, by employing a lattice gauge approach based on a set of orthogonal orbitals associated with each site. These microscopic fields are defined using a single-particle electron Green function, and the equations governing its evolution under excitation by an electromagnetic field at arbitrary frequency involve the electric and magnetic fields rather than the scalar and vector potentials. If the sites are taken to be far from each other, we recover the limit of isolated atoms. For an infinite crystal, we choose the orbitals to be maximally localized Wannier functions, and in the long-wavelength limit we recover the expected linear response of an insulator, including the zero frequency transverse conductivity of a topologically nontrivial insulator. For a topologically trivial insulator, we recover the expected expressions for the macroscopic polarization and magnetization in the ground state and find that the linear response to excitation at arbitrary frequency is described solely by the microscopic polarization and magnetization fields. For very general optical response calculations, the microscopic fields necessarily satisfy charge conservation, even under basis truncation, and do not suffer from the false divergences at zero frequency that can plague response calculations using other approaches.

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I. INTRODUCTION

The optical properties of materials were first related to the microscopic structure and properties of matter near the start of the twentieth century [1]. Macroscopic polarization and magnetization fields, $\boldsymbol{P}(\boldsymbol{x},t)$ and $\boldsymbol{M}(\boldsymbol{x},t)$, which appear in the description of the macroscopic charge and current densities,

$$\begin{aligned}\varrho(\boldsymbol{x},t) &= -\nabla\cdot\boldsymbol{P}(\boldsymbol{x},t)+\varrho_F(\boldsymbol{x},t),\\ \boldsymbol{J}(\boldsymbol{x},t) &= \frac{\partial\boldsymbol{P}(\boldsymbol{x},t)}{\partial t}+c\nabla\times\boldsymbol{M}(\boldsymbol{x},t)+\boldsymbol{J}_F(\boldsymbol{x},t),\end{aligned} \qquad (1)$$

were associated with charges "bound" in molecules and attributed to the electric and magnetic dipole moments of those molecules. The "free" charge and current densities, $\varrho_F(\boldsymbol{x},t)$ and $\boldsymbol{J}_F(\boldsymbol{x},t)$, were associated with charged carriers free to move through the medium if driven by applied fields, as in a conductor. Later developments extended the definitions of $\boldsymbol{P}(\boldsymbol{x},t)$ and $\boldsymbol{M}(\boldsymbol{x},t)$ to include contributions from higher multipole moments of constituent molecules [2,3]. The work of Power, Zienau, Wooley,[1] and Healy [5] established a framework in which the interaction of a molecule with the electromagnetic field, fundamentally described by the minimal coupling Hamiltonian, could be written involving microscopic polarization and magnetization fields associated with each molecule and the microscopic electromagnetic field in the neighborhood of that molecule. An expansion of the electromagnetic field about a point in the molecule then leads to a Hamiltonian involving the multipole moments of the molecule. For fluid systems, this was combined with the definition of the macroscopic polarization and magnetization as ensemble averages of the densities of multipoles and their derivatives [6], establishing the basis of the modern theory of the optical properties of fluids.

The approach is based on the identification of molecules as stable units, with charges moving within units but not moving from unit to unit. Sometimes this can be a good approximation even for solids, as in the treatment of molecular crystals. Yet at a fundamental level it appears problematic: valence electrons in a periodic solid are typically associated with Bloch waves extending throughout the crystal, and as the electron motion is perturbed by an electromagnetic field it is not clear how a multipole expansion about any point would make sense. Of course, one can always work directly with the minimal coupling Hamiltonian and simply calculate the charge and current densities induced by any applied field, bypassing a description in terms of polarization and magnetization fields. However, the physical insight that such a description would provide is lacking, and when the basis states used in calculations are truncated, as they inevitably must be, the use of the electromagnetic potentials in a perturbation calculation can lead to unphysical divergences due to the violation of certain sum rules [7,8]. A strategy developed by Adams, Blount, and others [9] in the 1960s relied on the introduction of a macroscopic polarization associated with the position operator, but that operator is poorly defined in a periodic solid and the calculations must be treated with care [10,11].

A different strategy is that of the "modern theory of polarization" [12,13], which has focused on gapped systems

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[1] For a review and references to original work see Ref. [4].

where physical intuition suggests that to lowest order none of the response to slowly varying fields should be identified with "free" currents. Then, under the application of a uniform applied electric field, a change in polarization is associated with the induced current density $\boldsymbol{J}$ via the equation $d\boldsymbol{P}/dt \equiv \boldsymbol{J}$. Since $\boldsymbol{J}$ can be calculated for an adiabatically applied uniform electric field, at least the change in $\boldsymbol{P}$ can be identified. Magnetizations $\boldsymbol{M}$ associated with unperturbed systems can also be identified [14,15], and for topologically trivial insulators, the ground-state polarization and magnetization are associated with the electric and magnetic dipole moments of the filled Wannier functions, respectively. For the polarization, however, there is a "quantum of ambiguity" that arises and is related to the way one associates Wannier functions with lattice sites; this is related to the gauge choice made in the definition of the Bloch eigenstates. There are also subtleties arising in the definition of the magnetization that are associated with the itinerant motion of charges between lattice sites [14]. Recently the effects of electron-electron interactions on these quantities have been addressed [16–19]. Yet treatments following this strategy have generally been aimed only at describing the ground state, or the response to uniform, adiabatically varying applied fields.[2]

A more general approach would be to seek *microscopic* polarization and magnetization fields $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$, and *microscopic* "free" charge and current densities $\rho_F(\boldsymbol{x},t)$ and $\boldsymbol{j}_F(\boldsymbol{x},t)$, such that the expectation value of the microscopic charge and current density operators would be given by

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle &= -\nabla\cdot\boldsymbol{p}(\boldsymbol{x},t)+\rho_F(\boldsymbol{x},t),\\ \langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle &= \frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t}+c\nabla\times\boldsymbol{m}(\boldsymbol{x},t)+\boldsymbol{j}_F(\boldsymbol{x},t).\end{aligned}\tag{2}$$

For a specified $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$, a set of such quantities $(\boldsymbol{p}(\boldsymbol{x},t),\boldsymbol{m}(\boldsymbol{x},t),\rho_F(\boldsymbol{x},t),\boldsymbol{j}_F(\boldsymbol{x},t))$ required only to satisfy (2) is far from unique.[3] Yet to have clear physical significance we could seek polarization and magnetization fields that consist of contributions associated with different sites, labeled by $\boldsymbol{R}$,

$$\begin{aligned}\boldsymbol{p}(\boldsymbol{x},t) &= \sum_{\boldsymbol{R}}\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t),\\ \boldsymbol{m}(\boldsymbol{x},t) &= \sum_{\boldsymbol{R}}\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t),\end{aligned}\tag{3}$$

with $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$ explicitly related to the microscopic charge and current densities in the neighborhood of the site $\boldsymbol{R}$.

Although there is no unique choice for the site(s), there may be a natural one. For an isolated atom, if the nucleus is taken to be fixed in space, the position of the nucleus can be taken to identify the single site $\boldsymbol{R}$. For a crystal with a single atom per unit cell, if we neglect the motion of the ion cores, we can take the sites to be the positions of the ions, such that the $\{\boldsymbol{R}\}$ form a Bravais lattice. For a crystal with multiple atoms per unit cell, there may not be such a natural choice for the sites. However, we can always choose there to be one site in each unit cell such that the $\{\boldsymbol{R}\}$ again form a Bravais lattice. The positions of the ions composing the crystal lattice are then described by a Bravais lattice, consisting of the sites $\{\boldsymbol{R}\}$, with a basis; in this case, there will necessarily be positive charge located away from each site. We refer to a collection of sites that form a Bravais lattice as the "lattice sites"; dealing with the periodic structures that host such lattice sites is the primary focus of this paper. The total charge and current densities will be unaffected by the choice of the lattice sites, although the unit cell quantities $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$ will depend on that choice. In most of the paper, we focus only on the charge and current densities associated with the valence electrons. In Conclusion, we indicate how the charge density of the ions in the unit cell can be included in our unit cell quantities, in the limit that the ions are considered fixed; in a future publication, we plan to generalize to include the motion of the ions.

The macroscopic fields corresponding to $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$ would be obtained by taking spatial averages of these microscopic fields, and a moment expansion of the $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$ about their lattice sites would generate a moment expansion in the resulting macroscopic fields $\boldsymbol{P}(\boldsymbol{x},t)$ and $\boldsymbol{M}(\boldsymbol{x},t)$. Such an approach could describe the effects of variations of the electromagnetic field over the unit cell on the optical response, and could treat the optical response at arbitrary frequency; it would not be restricted to treating excitation by adiabatically applied uniform optical fields. The microscopic polarization and magnetization fields, which could vary significantly over the unit cell, would give a physical picture of the effects of optical excitation.

This is the approach we develop in this paper. Overall, the framework of the dynamical equations that arise provides the microscopic underpinning of a lattice gauge theory, involving matrices that include labels for basis functions associated with each lattice site. The site charges and link currents are identified with the free charge and current densities, respectively, and the polarization and magnetization fields (3) arise from the matrices that are associated with the lattice sites. And since we find that the free charge and current densities themselves satisfy continuity by construction, regardless of the approximations made in the calculation of $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$, the values of $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$ determined from (2) necessarily satisfy continuity. Therefore, in this approach, charge conservation is completely robust against any approximations.

In constructing the description of the dynamics of the fields $\boldsymbol{p}(\boldsymbol{x},t)$, $\boldsymbol{m}(\boldsymbol{x},t)$, $\rho_F(\boldsymbol{x},t)$, and $\boldsymbol{j}_F(\boldsymbol{x},t)$ we are naturally led to describe the effects of the electromagnetic field by the electric and magnetic fields themselves, rather than the scalar and vector potentials. Thus, calculations following this strategy should be free of the kind of unphysical divergences that can plague the use of the minimal coupling Hamiltonian [7,8]. The transformation from an interaction involving the scalar and vector potentials to one involving the electric and magnetic

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[2] For a treatment of the magnetic response, see, e.g., Ref. [20].

[3] As a trivial example, consider $(\boldsymbol{p}(\boldsymbol{x},t),\boldsymbol{m}(\boldsymbol{x},t),\rho_F(\boldsymbol{x},t),\boldsymbol{j}_F(\boldsymbol{x},t))=(-\boldsymbol{e}(\boldsymbol{x},t)/4\pi,\boldsymbol{b}(\boldsymbol{x},t)/4\pi,0,0)$, where $\boldsymbol{e}(\boldsymbol{x},t)$ and $\boldsymbol{b}(\boldsymbol{x},t)$ are the total microscopic electric and magnetic fields, including contributions from the charge and current densities themselves. Then Maxwell's equations guarantee that (2) are satisfied. Indeed, one could even just choose $\boldsymbol{e}(\boldsymbol{x},t)$ and $\boldsymbol{b}(\boldsymbol{x},t)$ to be just the electrostatic and magnetostatic fields generated by the charge current distribution. See Ref. [22].

fields arises because we borrow some of our strategy from the theory of the optical response of molecules [21]. Yet there are important differences between that approach and ours. Because charges can move from site to site, we do not attempt to construct a Hamiltonian involving operators for $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$, and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$, as is usually done for atoms and molecules, since there is no reasonable protocol by which charges that would be associated with each site could be identified [22]. Rather, we construct expressions for $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$ in terms of electron Green functions and their expansions in terms of localized basis functions associated with each site $\boldsymbol{R}$. In treating a large molecule these localized orbitals could be convenient orthonormal molecular orbitals [23], but for the problem of a periodic solid, that is, the focus of this paper, we choose the maximally localized Wannier functions that can be constructed from each set of bands that is topologically trivial [24–27]; in general, we make no assumption about the initial occupation of any of these bands. In the process of constructing $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$, we can confirm that they are indeed related to the microscopic charge and current densities near $\boldsymbol{R}$. Thus, for lattice constants much less than the wavelength of light, a multipole expansion about each lattice site is justified.

In this first paper, we consider an incident classical electromagnetic field and neglect interactions between electrons except as could be captured in a simple mean-field treatment. While the formalism does not require it, the restriction to a single-particle Hamiltonian eases our initial formulation, for it is sufficient to consider only the lesser, equal time single-particle Green function. We omit the spin contribution to the magnetization; it could be easily included and does not affect the kind of issues that arise here. We also assume the ions are fixed, but do allow for a general dependence of the unperturbed Hamiltonian on position and momentum. Our goal here is to present the basic formalism, and so we will only be able to allude to some of the physical points made above, and some made below. We plan to return to many of the issues raised here in future publications and to present a treatment of electron-electron interactions that will rely on a more general Green function framework.

After deriving the basic equations for an arbitrary applied electromagnetic field here, however, we consider four important limits that can be reached from the general results; these limits serve as a form of validation for the formalism we develop, in addition to being helpful examples of its use. We first confirm that for isolated atoms on a lattice our result reduces to what would be expected from the usual treatment of atoms, to all orders in the multipole moments. We then consider the limit that is often of interest in optical response, where the applied electric field is approximated as uniform and the applied magnetic field as vanishing; this is the so-called "long-wavelength limit." We show how the current density in that limit consists of the time derivative of the polarization and a free current density. For a topologically nontrivial insulator, the linear response of the free current density to an applied field includes the current perpendicular to the applied field described by a transverse conductivity, as expected [28,29].

For the other two limits under investigation, we restrict ourselves to topologically trivial insulators. In the first, we show that the usual expressions for the bulk polarization and magnetization from the "modern theory" are reproduced in our treatment of the ground state. We then show that in the linear response to an electromagnetic field of arbitrary wavelength there is no free current or change in the free charge density induced; the first-order response is completely described by the polarization and magnetization. This is what one would physically expect of the kind of approach we develop here; it is only to higher order, when injected electrons and holes can be driven by the electromagnetic field, that one would expect the appearance of free charges and currents. We note that this would hold even for excitation by x rays. In future publications, we will extend these investigations to systems that are topologically nontrivial, as well as consider the description of linear and nonlinear response of a range of materials.

In Sec. II, we present the derivation, in Sec. III, we present the limits mentioned above, and in Sec. IV, we conclude. Many details of the derivation have been relegated to Appendices.

II. MICROSCOPIC POLARIZATION AND MAGNETIZATION FIELDS

We work in the Heisenberg picture with the lesser, equal time single-particle Green function

$$G_{mc}(\boldsymbol{x},\boldsymbol{y};t) = i\langle\psi^\dagger(\boldsymbol{y},t)\psi(\boldsymbol{x},t)\rangle, \tag{4}$$

where $\psi(\boldsymbol{x},t)$ is the fermionic electron field operator, and the subscript mc denotes the usual minimal coupling procedure to include the effect of an external, classical electromagnetic field specified by a scalar potential $\phi(\boldsymbol{x},t)$ and a vector potential $\boldsymbol{A}(\boldsymbol{x},t)$; interactions between the electrons, except as they might be included within the use of a self-consistent electromagnetic field, are neglected. The Green function satisfies the dynamical equation

$$i\hbar\frac{\partial G_{mc}(\boldsymbol{x},\boldsymbol{y};t)}{\partial t} = \mathcal{K}_{mc}(\boldsymbol{x},\boldsymbol{y};t)G_{mc}(\boldsymbol{x},\boldsymbol{y};t), \tag{5}$$

where

$$\mathcal{K}_{mc}(\boldsymbol{x},\boldsymbol{y};t) = \mathcal{H}_{mc}(\boldsymbol{x},t) - \mathcal{H}^*_{mc}(\boldsymbol{y},t), \tag{6}$$

with the "script" fonts (such as $\mathcal{K}_{mc}$ and $\mathcal{H}_{mc}$) denoting differential operators acting on the functions that follow, at and in the neighborhood of the spatial variable(s) indicated. We have

$$\mathcal{H}_{mc}(\boldsymbol{x},t) = H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + e\phi(\boldsymbol{x},t), \tag{7}$$

where

$$\mathfrak{p}_{mc}(\boldsymbol{x},t) = \mathfrak{p}(\boldsymbol{x}) - \frac{e}{c}\boldsymbol{A}(\boldsymbol{x},t), \tag{8}$$

with

$$\mathfrak{p}(\boldsymbol{x}) = \frac{\hbar}{i}\frac{\partial}{\partial\boldsymbol{x}} - \frac{e}{c}\boldsymbol{A}_{\text{static}}(\boldsymbol{x}). \tag{9}$$

In (9), we have included the possibility of the presence of a static, periodic magnetic field described by a vector potential

$\boldsymbol{A}_{\text{static}}(\boldsymbol{x})$, where $\boldsymbol{A}_{\text{static}}(\boldsymbol{x}) = \boldsymbol{A}_{\text{static}}(\boldsymbol{x}+\boldsymbol{R})$ for any lattice site position $\boldsymbol{R}$; note that we distinguish such a static, periodic vector potential from the "external" electromagnetic field associated with $\phi(\boldsymbol{x},t)$ and $\boldsymbol{A}(\boldsymbol{x},t)$. The use of "normal" fonts (such as H_0) denotes a function of the quantities indicated as variables, some of which may be differential operators. For the usual Schrödinger Hamiltonian, for example, in the absence of an external electromagnetic field, we have

$$H_0^{\text{Sch}}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x})) = \frac{(\mathfrak{p}(\boldsymbol{x}))^2}{2m} + V(\boldsymbol{x}), \tag{10}$$

but more general forms can be considered. In a system that is periodic before the application of an external electromagnetic field, we have $H_0(\boldsymbol{x}+\boldsymbol{R},\mathfrak{p}(\boldsymbol{x})) = H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))$, but for much of the following this assumption is not necessary; a large part of the formalism developed here could be used to introduce microscopic polarization and magnetization fields to describe the optical response of large molecules. The dynamical Eq. (5) was obtained from the simpler equation governing the dynamics of the Green function in the absence of an external electromagnetic field by the usual minimal coupling prescription, which in our notation is written

$$H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x})) \to H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + e\phi(\boldsymbol{x},t).$$

The expectation value of the electronic charge density operator, $\hat{\rho}(\boldsymbol{x},t)$, and the electronic current density operator, $\hat{\boldsymbol{j}}(\boldsymbol{x},t)$, are given by

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle &= -ie\big[G_{mc}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}},\\ \langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle &= \big[\mathcal{J}_{mc}(\boldsymbol{x},\boldsymbol{y};t)G_{mc}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}},\end{aligned} \tag{11}$$

where

$$\mathcal{J}_{mc}(\boldsymbol{x},\boldsymbol{y};t) = -\frac{i}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + \boldsymbol{J}(\boldsymbol{y},\mathfrak{p}^*_{mc}(\boldsymbol{y},t))\big],$$

and the function $\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))$ follows from $H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))$ in the usual fashion, see, e.g., Ref. [30]. For the Schrödinger Hamiltonian, for example, we have

$$\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) = \frac{e}{m}\mathfrak{p}_{mc}(\boldsymbol{x},t).$$

Many of the expression above involve the scalar and vector potentials explicitly. It is possible to replace that set of expressions by a corresponding set of expressions that involve only the electric and magnetic fields associated with these potentials,

$$\begin{aligned}\boldsymbol{E}(\boldsymbol{x},t) &= -\nabla\phi(\boldsymbol{x},t) - \frac{1}{c}\frac{\partial\boldsymbol{A}(\boldsymbol{x},t)}{\partial t},\\ \boldsymbol{B}(\boldsymbol{x},t) &= \nabla\times\boldsymbol{A}(\boldsymbol{x},t).\end{aligned}$$

The strategy for doing this was introduced many years ago for atoms and molecules where a "special point," such as the center of mass of the atom or molecule, or the position of a nucleus assumed fixed, is identified [21]. We implement an analogous strategy in the paragraph below without identifying such a special point, but still employing quantities used in problems in atomic and molecular physics. Those quantities, or "relators," are defined [31,32] as

$$\begin{aligned}s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) &= \int_{C(\boldsymbol{x},\boldsymbol{y})} dz^i\,\delta(\boldsymbol{w}-\boldsymbol{z}),\\ \alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) &= \epsilon^{jmn}\int_{C(\boldsymbol{x},\boldsymbol{y})} dz^m\,\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z}),\\ \beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) &= \epsilon^{jmn}\int_{C(\boldsymbol{x},\boldsymbol{y})} dz^m\,\frac{\partial z^n}{\partial y^k}\delta(\boldsymbol{w}-\boldsymbol{z}),\end{aligned} \tag{12}$$

where for each two points $\boldsymbol{x}$ and $\boldsymbol{y}$ we use $C(\boldsymbol{x},\boldsymbol{y})$ to indicate some path from $\boldsymbol{y}$ to $\boldsymbol{x}$. Here and below superscripts indicate Cartesian components, ϵ^{jmn} is the Levi-Civita symbol, and repeated Cartesian components are summed over.[4] Regardless of the path chosen, the quantities (12) satisfy

$$\begin{aligned}\frac{\partial s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^i} &= -\delta(\boldsymbol{w}-\boldsymbol{x}) + \delta(\boldsymbol{w}-\boldsymbol{y}),\\ \frac{\partial s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial x^k} &= \delta^{ik}\delta(\boldsymbol{w}-\boldsymbol{x}) - \epsilon^{ipj}\frac{\partial\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p},\\ \frac{\partial s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial y^k} &= -\delta^{ik}\delta(\boldsymbol{w}-\boldsymbol{y}) - \epsilon^{ipj}\frac{\partial\beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}\end{aligned} \tag{13}$$

(see Appendix A). It is useful to choose a "symmetric" set of paths, by which we mean that for each and every $\boldsymbol{x}$ and $\boldsymbol{y}$ the path $C(\boldsymbol{x},\boldsymbol{y})$ is the "reverse" of the path $C(\boldsymbol{y},\boldsymbol{x})$. For a symmetric set of paths, we call the resulting sets of relators "symmetric," and they satisfy

$$\begin{aligned}s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) + s^i(\boldsymbol{w};\boldsymbol{y},\boldsymbol{x}) &= 0,\\ \alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) + \beta^{jk}(\boldsymbol{w};\boldsymbol{y},\boldsymbol{x}) &= 0.\end{aligned} \tag{14}$$

(See Appendix A). The second of these means that $\beta^{jk}(\boldsymbol{w};\boldsymbol{y},\boldsymbol{x})$ can be eliminated in favor of $\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})$, which we generally do in the formulas below.

A. The global Green function

We now use these relators to introduce a new Green function, $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$.[5] The subscript gl denotes "global," in that no special point is introduced, and yet the dynamical equation for $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ and the expressions for the electronic charge and current densities in terms of it are gauge invariant in that they depend directly on the electric and magnetic fields, and not explicitly on the scalar and vector potentials. The gauge freedom in choosing the scalar and vector potentials for a given electromagnetic field has in a sense been replaced by a similar freedom in choosing the paths $C(\boldsymbol{x},\boldsymbol{y})$, or in choosing sets of relators satisfying (13) that are even more general than (12). While straight lines between $\boldsymbol{y}$ and $\boldsymbol{x}$ are almost always chosen for the paths $C(\boldsymbol{x},\boldsymbol{y})$ in applications (see Appendix A), the freedom to choose different sets of relators is still there. To avoid confusion, we use the term "gauge invariant" here to refer specifically to quantities that do not depend on the scalar and vector potentials explicitly, but only on the electric and magnetic fields.

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[4] As the metric for spatial components is identity, covariant and contravariant objects transform trivially into one another.

[5] This extends earlier works [31,32,37].

To obtain the global Green function, $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$, we introduce

$$\Phi(\boldsymbol{x},\boldsymbol{y};t) \equiv \frac{e}{\hbar c}\int s^{i}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})A^{i}(\boldsymbol{w},t)d\boldsymbol{w}, \tag{15}$$

for a general path—note that if a straight-line path is chosen this is just the standard Peierls phase—and put

$$G_{gl}(\boldsymbol{x},\boldsymbol{y};t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{y};t)}G_{mc}(\boldsymbol{x},\boldsymbol{y};t). \tag{16}$$

As we always use symmetric relators, we obtain

$$G^{*}_{gl}(\boldsymbol{x},\boldsymbol{y};t) = -G_{gl}(\boldsymbol{y},\boldsymbol{x};t),$$

following from the fact that $G_{mc}(\boldsymbol{x},\boldsymbol{y};t)$ trivially satisfies this relation. While each factor on the right-hand side of (16) is gauge dependent, their product is not. The dynamical equation for $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ is found to be

$$i\hbar\frac{\partial G_{gl}(\boldsymbol{x},\boldsymbol{y};t)}{\partial t} = \mathcal{K}_{gl}(\boldsymbol{x},\boldsymbol{y};t)G_{gl}(\boldsymbol{x},\boldsymbol{y};t), \tag{17}$$

where

$$\begin{aligned}\mathcal{K}_{gl}(\boldsymbol{x},\boldsymbol{y};t) = {} & H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{y};t)) - H^{*}_{0}(\boldsymbol{y},\mathfrak{p}(\boldsymbol{y},\boldsymbol{x};t)) \\ & - e\Omega^{0}_{\boldsymbol{y}}(\boldsymbol{x},t),\end{aligned} \tag{18}$$

with

$$\mathfrak{p}^{k}(\boldsymbol{x},\boldsymbol{y};t) \equiv \mathfrak{p}^{k}(\boldsymbol{x}) - \frac{e}{c}\Omega^{k}_{\boldsymbol{y}}(\boldsymbol{x},t), \tag{19}$$

and

$$\begin{aligned}\Omega^{0}_{\boldsymbol{y}}(\boldsymbol{x},t) &\equiv \int s^{i}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})E^{i}(\boldsymbol{w},t)d\boldsymbol{w}, \\ \Omega^{k}_{\boldsymbol{y}}(\boldsymbol{x},t) &\equiv \int \alpha^{lk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})B^{l}(\boldsymbol{w},t)d\boldsymbol{w}.\end{aligned} \tag{20}$$

Now using (11,16), we can write the expectation value of the electronic charge and current densities as

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle &= -ie\big[G_{gl}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}}, \\ \langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle &= \big[\mathcal{J}_{gl}(\boldsymbol{x},\boldsymbol{y};t)G_{gl}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}},\end{aligned} \tag{21}$$

where

$$\mathcal{J}_{gl}(\boldsymbol{x},\boldsymbol{y};t) = -\frac{i}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{y};t)) + \boldsymbol{J}^{*}(\boldsymbol{y},\mathfrak{p}(\boldsymbol{y},\boldsymbol{x};t))\big].$$

Each these quantities is clearly gauge invariant.

B. Wannier functions and adjusted Wannier functions

While the global gauge invariant Green function $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ is interesting in its own right, our goal here is to use it to associate charge and current densities with each lattice site, and then associate microscopic polarizations and magnetizations with those sites in a way similar, as much as possible, with what one would do to treat a model consisting of "isolated atoms" where charges could not move from atom to atom. To do this, we introduce a set of localized basis functions $\{W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\}$, or "orbitals," labeled by the lattice site $\boldsymbol{R}$ and a "type index" α. We take the functions in the set $\{W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\}$ to be orthogonal,

$$\int W^{*}_{\beta\boldsymbol{R}'}(\boldsymbol{x})W_{\alpha\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x} = \delta_{\beta\alpha}\delta_{\boldsymbol{R}'\boldsymbol{R}}. \tag{22}$$

In the special case of a periodic system, which is our main focus here, we have

$$W_{\alpha\boldsymbol{R}}(\boldsymbol{x}) = W_{\alpha}(\boldsymbol{x}-\boldsymbol{R}), \tag{23}$$

where we identify $W_{\alpha}(\boldsymbol{x})$ as a function localized near the origin—in general the functions will be centered at different points for different α—and $\{W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\}$ is a set of Wannier functions. We make a particular choice for these Wannier functions by requiring that they are maximally localized functions; then each N element subset of the Wannier functions considered will be constructed from a subset of N bands that do not intersect in energy with elements of other subsets of bands, and where the subset of bands used is, as a whole, topologically trivial[6] [24–27]; at the moment we make no assumption about the initial occupation of any of these bands.

In the presence of a *uniform* vector potential, $\boldsymbol{A}(\boldsymbol{x},t)\to\boldsymbol{A}(t)$, the set $\{W'_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$ of modified orbitals

$$W'_{\alpha\boldsymbol{R}}(\boldsymbol{x},t) = e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}W_{\alpha\boldsymbol{R}}(\boldsymbol{x}) \tag{24}$$

are orthonormal and often useful in calculations. Yet for *nonuniform* vector potentials the functions in the set $\{W'_{\alpha\boldsymbol{R}}(\boldsymbol{x})\}$ are not orthonormal, nor even are their overlap integrals gauge invariant. However, Löwdin's method of symmetric orthogonalization [23] can be used to construct an orthonormal set of functions $\{\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$,

$$\int \bar{W}^{*}_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)d\boldsymbol{x} = \delta_{\beta\alpha}\delta_{\boldsymbol{R}'\boldsymbol{R}}, \tag{25}$$

which are as close as possible to the functions $\{W'_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$, in the sense that

$$\sum_{\alpha,\boldsymbol{R}}\int\big|\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t) - W'_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\big|^{2}d\boldsymbol{x} \tag{26}$$

is a minimum at each time t. We refer to these new functions as "adjusted Wannier functions." For a finite system, the set $\{\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$ can be found numerically if the sum over α is truncated. In any case, it always follows that the $\{\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$ are of the form

$$\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t) = e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t), \tag{27}$$

where the functions in the set $\{\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\}$ are generally *not* orthonormal, but can be written in a gauge invariant way, and in fact depend only on the magnetic field and not on the electric field (see Appendix B). In the limit of a weak applied magnetic field, one can construct a perturbative expansion for $\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$; the first two terms are

$$\begin{aligned}\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t) = {} & W_{\alpha\boldsymbol{R}}(\boldsymbol{x}) - \frac{1}{2}i\sum_{\beta,\boldsymbol{R}'}W_{\beta\boldsymbol{R}'}(\boldsymbol{x}) \\ & \times\left[\int W^{*}_{\beta\boldsymbol{R}'}(\boldsymbol{y})\Delta(\boldsymbol{R}',\boldsymbol{y},\boldsymbol{R};t)W_{\alpha\boldsymbol{R}}(\boldsymbol{y})d\boldsymbol{y}\right] + \dots\end{aligned} \tag{28}$$

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[6] In 2D, this condition implies the net Chern number associated with the bands in the subset used in constructing Wannier functions is zero.

(see Appendix B), where very generally

$$\Delta(\boldsymbol{x},\boldsymbol{y},\boldsymbol{z};t) \equiv \Phi(\boldsymbol{z},\boldsymbol{x};t) + \Phi(\boldsymbol{x},\boldsymbol{y};t) + \Phi(\boldsymbol{y},\boldsymbol{z};t) \tag{29}$$

is a gauge invariant quantity, since it involves only the flux of the magnetic field through the surface identified by $\boldsymbol{x}, \boldsymbol{y}, \boldsymbol{z}$ and the paths connecting them.

Neglecting the spin degree of freedom, which could be included in a straight-forward way, we expand the fermionic electron field operator,

$$\psi(\boldsymbol{x},t) = \sum_{\alpha,\boldsymbol{R}} a_{\alpha\boldsymbol{R}}(t)\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t), \tag{30}$$

where

$$\begin{aligned}\left\{a_{\alpha\boldsymbol{R}}(t), a_{\beta\boldsymbol{R}'}(t)\right\} &= 0,\\ \left\{a_{\alpha\boldsymbol{R}}(t), a^{\dagger}_{\beta\boldsymbol{R}'}(t)\right\} &= \delta_{\alpha\beta}\delta_{\boldsymbol{R}\boldsymbol{R}'}.\end{aligned} \tag{31}$$

Formally, of course we can take the number of type indices to be infinite, so we have a complete set of basis functions in the expansion (30). However, we will derive expressions for the microscopic charge and current density involving intermediate quantities such that charge is explicitly conserved, and so in evaluating those intermediate quantities it will be possible to truncate the basis without violating continuity. In this basis, the Green function $G_{mc}(\boldsymbol{x},\boldsymbol{y};t)$ takes the form

$$G_{mc}(\boldsymbol{x},\boldsymbol{y};t) = i\sum_{\alpha,\beta,\boldsymbol{R},\boldsymbol{R}'} \breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)\bar{W}^{*}_{\beta\boldsymbol{R}'}(\boldsymbol{y},t)\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t), \tag{32}$$

where

$$\breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) = \langle a^{\dagger}_{\beta\boldsymbol{R}'}(t)a_{\alpha\boldsymbol{R}}(t)\rangle. \tag{33}$$

From the dynamical Eq. (5) for $G_{mc}(\boldsymbol{x},\boldsymbol{y};t)$, we immediately find the equations of motion for the $\breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$,

$$i\hbar\frac{\partial\breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t} = \sum_{\lambda,\boldsymbol{R}''}\left(e^{i\Phi(\boldsymbol{R},\boldsymbol{R}'';t)}\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\breve{\eta}_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t) - \breve{\eta}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\bar{H}_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)e^{i\Phi(\boldsymbol{R}'',\boldsymbol{R}';t)}\right) - e\Omega^{\phi}_{\boldsymbol{R}'}(\boldsymbol{R},t)\breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t), \tag{34}$$

where

$$\Omega^{\phi}_{\boldsymbol{y}}(\boldsymbol{x},t) \equiv -\int s^{i}(\boldsymbol{z};\boldsymbol{x},\boldsymbol{y})\frac{\partial\phi(\boldsymbol{z},t)}{\partial z^{i}}d\boldsymbol{z},$$

and the matrix $\bar{H}$ is Hermitian, $\bar{H}^{*}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t) = \bar{H}_{\lambda\boldsymbol{R}'';\alpha\boldsymbol{R}}(t)$, with elements that are gauge invariant,

$$\begin{aligned}\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t) = {} & \frac{1}{2}\int \chi^{*}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}'';t)}H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R}'';t))\chi_{\lambda\boldsymbol{R}''}(\boldsymbol{x},t)d\boldsymbol{x}\\ & + \frac{1}{2}\int \left(H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\right)^{*}\chi_{\lambda\boldsymbol{R}''}(\boldsymbol{x},t)e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}'';t)}d\boldsymbol{x}\\ & - \frac{e}{2}\int e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}'';t)}\chi^{*}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\left(\Omega^{0}_{\boldsymbol{R}''}(\boldsymbol{x},t) + \Omega^{0}_{\boldsymbol{R}}(\boldsymbol{x},t)\right)\chi_{\lambda\boldsymbol{R}''}(\boldsymbol{x},t)d\boldsymbol{x}\\ & - \frac{1}{2}i\hbar\int e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}'';t)}\left(\chi^{*}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\frac{\partial\chi_{\lambda\boldsymbol{R}''}(\boldsymbol{x},t)}{\partial t} - \chi_{\lambda\boldsymbol{R}''}(\boldsymbol{x},t)\frac{\partial\chi^{*}_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)}{\partial t}\right)d\boldsymbol{x}.\end{aligned} \tag{35}$$

Equations (34) and (35) can be understood as the microscopic underpinnings of the equations of a lattice gauge theory. These expressions display five main features: (a) the functions $\Phi(\boldsymbol{R},\boldsymbol{R}'';t)$ and $\Omega^{\phi}_{\boldsymbol{R}''}(\boldsymbol{R},t)$ allow for an arbitrary variation of the scalar and vector potential as one moves between neighboring lattice sites; (b) the "hopping" matrix elements $\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)$ involve a set of states (labeled by Greek letters) at each site, and thus are matrices even for fixed $\boldsymbol{R}$ and $\boldsymbol{R}''$; (c) in principle the hopping matrix elements connect each site to every other site, although in practice any site will only be coupled to sites close to it; (d) the matrix elements are also more complicated than in tight-binding models, containing the electric and magnetic fields, and are therefore time dependent; note, however, that they are gauge invariant; (e) since the complete electronic charge and current densities can be constructed once the $\breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ are found and $G_{mc}(\boldsymbol{x},\boldsymbol{y};t)$ identified from (32), the solution of (34) in fact allows for a determination of the full densities within the model identified by our original orbitals, and not just site charges and link currents. Indeed, with the use of the matrices $\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)$ we will construct the microscopic polarization and magnetization fields associated with each lattice site, and the site charges and link currents identified from the lattice gauge perspective will be associated with the microscopic version of the "free charges" and "free currents" of elementary electrodynamics.

C. Density operator dynamics

It is convenient to move to a fully gauge-invariant description by defining a single-particle density matrix

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) \equiv \breve{\eta}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)e^{i\Phi(\boldsymbol{R}',\boldsymbol{R};t)}, \tag{36}$$

from which it follows that the dynamics of $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ are specified by

$$\begin{aligned} & i\hbar\frac{\partial\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t}\\ & \quad = \sum_{\lambda,\boldsymbol{R}''} e^{i\Delta(\boldsymbol{R},\boldsymbol{R}'',\boldsymbol{R}';t)}\big(\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\eta_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\\ & \qquad - \eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\bar{H}_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\big) - e\Omega^{0}_{\boldsymbol{R}'}(\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t).\end{aligned} \tag{37}$$

Although the form of (37) is a bit unusual because of the factor $\exp(i\Delta(\boldsymbol{R},\boldsymbol{R}'',\boldsymbol{R}';t))$, note that all terms appearing in this equation for $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ are gauge invariant. Also, since any initial $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ before any fields are applied is trivially gauge invariant, then $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ itself will be gauge-invariant even as the fields are introduced. We can write $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ in terms of the $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ via (16,32,36), and from the orthonormality relations (25), we can write the $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ in terms of $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$; writing these results in terms of the $\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$ of (27), we have

$$\begin{aligned} G_{gl}(\boldsymbol{x},\boldsymbol{y};t) &= i\sum_{\alpha,\beta,\boldsymbol{R},\boldsymbol{R}'}\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)e^{i\Delta(\boldsymbol{x},\boldsymbol{R},\boldsymbol{R}',\boldsymbol{y};t)} \\ &\quad\times\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{y},t)\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t), \\ \eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) &= -i\int\chi_{\beta\boldsymbol{R}'}(\boldsymbol{y},t)\chi^*_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)e^{-i\Delta(\boldsymbol{x},\boldsymbol{R},\boldsymbol{R}',\boldsymbol{y};t)} \\ &\quad\times G_{gl}(\boldsymbol{x},\boldsymbol{y};t)d\boldsymbol{x}d\boldsymbol{y}, \end{aligned} \tag{38}$$

where in general

$$\begin{aligned}\Delta(\boldsymbol{x},\boldsymbol{u},\boldsymbol{v},\boldsymbol{y};t) &= \Phi(\boldsymbol{y},\boldsymbol{x};t)+\Phi(\boldsymbol{x},\boldsymbol{u};t)\\ &\quad+\Phi(\boldsymbol{u},\boldsymbol{v};t)+\Phi(\boldsymbol{v},\boldsymbol{y};t)\end{aligned}$$

is gauge-invariant. While the first equation of (38) involves a double sum over lattice sites, we now introduce a Green function $G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)$ associated with each lattice site $\boldsymbol{R}$, with the goal of writing $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ involving a sum over these. To guarantee that each $G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)$ satisfies the property

$$G^*_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t) = -G_{\boldsymbol{R}}(\boldsymbol{y},\boldsymbol{x};t),$$

as do $G_{mc}(\boldsymbol{x},\boldsymbol{y};t)$ and $G_{gl}(\boldsymbol{x},\boldsymbol{y};t)$ [see (4) and (16)], we take

$$\begin{aligned} G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t) &= \frac{i}{2}\sum_{\alpha,\beta,\boldsymbol{R}'}\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)e^{i\Delta(\boldsymbol{R}',\boldsymbol{y},\boldsymbol{R};t)}\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{y},t)\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t) \\ &\quad+\frac{i}{2}\sum_{\alpha,\beta,\boldsymbol{R}'}\eta_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}}(t)e^{i\Delta(\boldsymbol{R},\boldsymbol{x},\boldsymbol{R}';t)}\chi^*_{\alpha\boldsymbol{R}}(\boldsymbol{y},t)\chi_{\beta\boldsymbol{R}'}(\boldsymbol{x},t) \end{aligned} \tag{39}$$

and then have

$$G_{gl}(\boldsymbol{x},\boldsymbol{y};t) = \sum_{\boldsymbol{R}} e^{-i\Delta(\boldsymbol{x},\boldsymbol{y},\boldsymbol{R};t)}G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t). \tag{40}$$

The quantities (39) are chosen so that the gauge invariant phase factors $\exp(-i\Delta(\boldsymbol{x},\boldsymbol{y},\boldsymbol{R};t))$ appear in the decomposition (40) for reasons that will later become clear.[7]

We use the $G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)$ to identify components of the full electronic charge and current densities that we associate with each lattice site, $\rho_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$. From (21) and (40), we have

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle &= \sum_{\boldsymbol{R}}\rho_{\boldsymbol{R}}(\boldsymbol{x},t),\\ \langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle &= \sum_{\boldsymbol{R}}\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t),\end{aligned} \tag{41}$$

where

$$\begin{aligned}\rho_{\boldsymbol{R}}(\boldsymbol{x},t) &= -ie\big[G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}},\\ \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t) &= \big[\mathcal{J}_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)\big]_{\boldsymbol{y}\to\boldsymbol{x}},\end{aligned} \tag{42}$$

with

$$\mathcal{J}_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t) = -\frac{i}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))+\boldsymbol{J}^*(\boldsymbol{y},\mathfrak{p}(\boldsymbol{y},\boldsymbol{R};t))\big]. \tag{43}$$

In particular, we obtain

$$\begin{aligned}\rho_{\boldsymbol{R}}(\boldsymbol{x},t) &= \sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t),\\ \boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t) &= \sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\boldsymbol{j}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t),\end{aligned} \tag{44}$$

where

$$\begin{aligned}\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t) &= \frac{e}{2}(\delta_{\boldsymbol{R}\boldsymbol{R}'}+\delta_{\boldsymbol{R}\boldsymbol{R}''})e^{i\Delta(\boldsymbol{R}',\boldsymbol{x},\boldsymbol{R}'';t)}\\ &\quad\times\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\chi_{\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\end{aligned} \tag{45}$$

and

$$\begin{aligned}&\boldsymbol{j}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)\\ &=\tfrac{1}{4}\delta_{\boldsymbol{R}\boldsymbol{R}''}e^{i\Delta(\boldsymbol{R}',\boldsymbol{x},\boldsymbol{R}'';t)}\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\big(\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))\chi_{\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\big)\\ &\quad+\tfrac{1}{4}\delta_{\boldsymbol{R}\boldsymbol{R}'}\big(\boldsymbol{J}^*(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\big)e^{i\Delta(\boldsymbol{R}',\boldsymbol{x},\boldsymbol{R}'';t)}\chi_{\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\\ &\quad+\tfrac{1}{4}\delta_{\boldsymbol{R}\boldsymbol{R}''}\big(\boldsymbol{J}^*(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))e^{i\Delta(\boldsymbol{R}',\boldsymbol{x},\boldsymbol{R}'';t)}\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\big)\chi_{\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\\ &\quad+\tfrac{1}{4}\delta_{\boldsymbol{R}\boldsymbol{R}'}\chi^*_{\beta\boldsymbol{R}'}(\boldsymbol{x},t)\big(\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))e^{i\Delta(\boldsymbol{R}',\boldsymbol{x},\boldsymbol{R}'';t)}\chi_{\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\big).\end{aligned} \tag{46}$$

Note that in the expression (39) for $G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)$ the lattice site $\boldsymbol{R}$ always appears as one of the indices of the single-particle density matrix. Then, since $\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)$ can be reasonably expected to be nonzero only for $\boldsymbol{x}$ close to $\boldsymbol{R}$ [see the expansion (28)], we can expect the $\rho_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$ of (42) to be nonzero only for $\boldsymbol{x}$ close to $\boldsymbol{R}$ as well.

D. Site polarizations and magnetizations

Equations (42) and (43) are precisely the ones that would be written down in a model for isolated atoms, where there $G_{\boldsymbol{R}}(\boldsymbol{x},\boldsymbol{y};t)$ would be the Green function for the atom at $\boldsymbol{R}$. The "special point" $\boldsymbol{R}$ has been identified for the charge-current distribution about lattice site $\boldsymbol{R}$, and appears in the expression for the current density in precisely the way that a "special point" is identified in the treatment of an atom; it appears in (42) in that way because the phase factors $\exp(-i\Delta(\boldsymbol{x},\boldsymbol{y},\boldsymbol{R};t))$ were introduced in (40). A difference between our problem and that of isolated atoms, of course, is that in general

$$K_{\boldsymbol{R}}(\boldsymbol{x},t) \equiv \nabla\cdot\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)+\frac{\partial\rho_{\boldsymbol{R}}(\boldsymbol{x},t)}{\partial t}\neq 0, \tag{47}$$

since electrons can move from the region nearest one lattice site to regions nearest others. Thus generally the site charges

$$Q_{\boldsymbol{R}}(t) \equiv \int\rho_{\boldsymbol{R}}(\boldsymbol{x},t)d\boldsymbol{x} = e\sum_{\alpha}\eta_{\alpha\boldsymbol{R};\alpha\boldsymbol{R}}(t) \tag{48}$$

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[7] This is a further development of a strategy introduced earlier [32].

are time dependent. From the dynamical Eq. (37), we find that we can write

$$\frac{dQ_{\boldsymbol{R}}(t)}{dt}=\sum_{\boldsymbol{R}'}I(\boldsymbol{R},\boldsymbol{R}';t), \tag{49}$$

where

$$\begin{aligned}I(\boldsymbol{R},\boldsymbol{R}';t)=\frac{e}{i\hbar}\sum_{\alpha,\lambda}\big(&\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)\eta_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\\&-\eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)\bar{H}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\big).\end{aligned} \tag{50}$$

Since

$$I(\boldsymbol{R}',\boldsymbol{R};t)=-I(\boldsymbol{R},\boldsymbol{R}';t), \tag{51}$$

$I(\boldsymbol{R},\boldsymbol{R}';t)$ can be interpreted as the net current flowing from site $\boldsymbol{R}'$ to site $\boldsymbol{R}$, and thus as a link current.

With the site charges and link currents identified, we can now define microscopic "free" charge and current densities associated with them,

$$\rho_F(\boldsymbol{x},t)\equiv\sum_{\boldsymbol{R}}Q_{\boldsymbol{R}}(t)\delta(\boldsymbol{x}-\boldsymbol{R}), \tag{52}$$

$$\boldsymbol{j}_F(\boldsymbol{x},t)\equiv\frac{1}{2}\sum_{\boldsymbol{R},\boldsymbol{R}'}\boldsymbol{s}(\boldsymbol{x};\boldsymbol{R},\boldsymbol{R}')I(\boldsymbol{R},\boldsymbol{R}';t). \tag{53}$$

The first simply takes the free charge density to be the sum of the charges associated with each lattice site placed at that lattice site. The second introduces a microscopic current density by distributing the net current from site $\boldsymbol{R}'$ to $\boldsymbol{R}$ along the path from $\boldsymbol{R}'$ to $\boldsymbol{R}$ defined by $C(\boldsymbol{R},\boldsymbol{R}')$. From (49,51) and the second of the properties (13), we immediately find

$$\nabla\cdot\boldsymbol{j}_F(\boldsymbol{x},t)+\frac{\partial\rho_F(\boldsymbol{x},t)}{\partial t}=0. \tag{54}$$

That is, the microscopic free charge and current densities themselves satisfy continuity. We shall write the remaining contributions to the total charge and current densities in terms of microscopic polarization and magnetization fields associated with each site. We begin by introducing these terms as we would were we dealing with isolated atoms. We define site polarization fields $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ as

$$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)\equiv\int\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho_{\boldsymbol{R}}(\boldsymbol{y},t)d\boldsymbol{y}, \tag{55}$$

and define preliminary site magnetization fields as

$$\bar{m}^j_{\boldsymbol{R}}(\boldsymbol{x},t)\equiv\frac{1}{c}\int\alpha^{jk}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})j^k_{\boldsymbol{R}}(\boldsymbol{y},t)d\boldsymbol{y}. \tag{56}$$

Since $\rho_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x},t)$ are nonzero only for $\boldsymbol{x}$ close to $\boldsymbol{R}$, we can expect $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\bar{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$ to share that property as well. Introducing associated total microscopic polarization and magnetization fields,

$$\begin{aligned}\boldsymbol{p}(\boldsymbol{x},t)&=\sum_{\boldsymbol{R}}\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t),\\\bar{\boldsymbol{m}}(\boldsymbol{x},t)&=\sum_{\boldsymbol{R}}\bar{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t),\end{aligned} \tag{57}$$

with the use of (13) we obtain

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle&=-\nabla\cdot\boldsymbol{p}(\boldsymbol{x},t)+\rho_F(\boldsymbol{x},t),\\\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle&=\frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t}+c\nabla\times\bar{\boldsymbol{m}}(\boldsymbol{x},t)+\tilde{\boldsymbol{j}}(\boldsymbol{x},t)+\boldsymbol{j}_F(\boldsymbol{x},t),\end{aligned} \tag{58}$$

where

$$\tilde{\boldsymbol{j}}(\boldsymbol{x},t)=-\sum_{\boldsymbol{R}}\int\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})K_{\boldsymbol{R}}(\boldsymbol{y},t)d\boldsymbol{y}-\boldsymbol{j}_F(\boldsymbol{x},t). \tag{59}$$

In contrast with the problem of isolated atoms, here in general we have a time dependent $\rho_F(\boldsymbol{x},t)$, a nonzero $\boldsymbol{j}_F(\boldsymbol{x},t)$, and a nonzero $\tilde{\boldsymbol{j}}(\boldsymbol{x},t)$, all arising because the functions in $\{K_{\boldsymbol{R}}(\boldsymbol{x},t)\}$ are generally nonzero as charge moves from site to site. Nonetheless, since total charge is conserved, we have

$$\sum_{\boldsymbol{R}}K_{\boldsymbol{R}}(\boldsymbol{x},t)=0,$$

and it is easy to confirm that

$$\nabla\cdot\tilde{\boldsymbol{j}}(\boldsymbol{x},t)=0.$$

To complete our treatment of the site polarizations and magnetizations, we express the divergenceless $\tilde{\boldsymbol{j}}(\boldsymbol{x},t)$ in terms of the curls of magnetizations associated with the different lattice sites. We begin by writing

$$\tilde{\boldsymbol{j}}(\boldsymbol{x},t)=\sum_{\alpha,\beta,\boldsymbol{R}'',\boldsymbol{R}'''}\tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}''';\alpha\boldsymbol{R}''}(\boldsymbol{x},t)\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'''}(t),$$

where the expressions (47) and (53) for $K_{\boldsymbol{R}}(\boldsymbol{y},t)$ and $\boldsymbol{j}_F(\boldsymbol{x},t)$, respectively, are used to identify $\tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}''';\alpha\boldsymbol{R}''}(\boldsymbol{x},t)$,

$$\begin{aligned}\tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}''';\alpha\boldsymbol{R}''}(\boldsymbol{x},t)=&-\sum_{\boldsymbol{R}}\int\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\Gamma^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'''}_{\boldsymbol{R}}(\boldsymbol{y},t)d\boldsymbol{y}\\&-\frac{1}{2}\sum_{\boldsymbol{R},\boldsymbol{R}'}\boldsymbol{s}(\boldsymbol{x};\boldsymbol{R},\boldsymbol{R}')\varsigma^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'''}_{\boldsymbol{R}\boldsymbol{R}'}(t),\end{aligned} \tag{60}$$

where

$$\begin{aligned}\varsigma^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'''}_{\boldsymbol{R}\boldsymbol{R}'}(t)=\frac{e}{i\hbar}\big(&\delta_{\boldsymbol{R}'''\boldsymbol{R}}\delta_{\boldsymbol{R}''\boldsymbol{R}'}\bar{H}_{\beta\boldsymbol{R};\alpha\boldsymbol{R}'}(t)\\&-\delta_{\boldsymbol{R}''\boldsymbol{R}}\delta_{\boldsymbol{R}'''\boldsymbol{R}'}\bar{H}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\big)\end{aligned} \tag{61}$$

and

$$\begin{aligned}\Gamma^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}_{\boldsymbol{R}}(\boldsymbol{x},t)=&\nabla\cdot\boldsymbol{j}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)+\frac{\partial\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)}{\partial t}\\&+\frac{1}{i\hbar}\sum_{\mu,\nu,\boldsymbol{R}_1,\boldsymbol{R}_2}\rho_{\nu\boldsymbol{R}_2;\mu\boldsymbol{R}_1}(\boldsymbol{x},\boldsymbol{R};t)\mathfrak{F}^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}_{\mu\boldsymbol{R}_1;\nu\boldsymbol{R}_2}(t),\end{aligned} \tag{62}$$

with

$$\begin{aligned}\mathfrak{F}^{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}_{\mu\boldsymbol{R}_1;\nu\boldsymbol{R}_2}(t)=&\delta_{\beta\nu}\delta_{\boldsymbol{R}_2\boldsymbol{R}'}e^{i\Delta(\boldsymbol{R}_1,\boldsymbol{R}'',\boldsymbol{R}_2;t)}\bar{H}_{\mu\boldsymbol{R}_1;\alpha\boldsymbol{R}''}(t)\\&-\delta_{\alpha\mu}\delta_{\boldsymbol{R}''\boldsymbol{R}_1}e^{i\Delta(\boldsymbol{R}_1,\boldsymbol{R}',\boldsymbol{R}_2;t)}\bar{H}_{\beta\boldsymbol{R}';\nu\boldsymbol{R}_2}(t)\\&-e\delta_{\beta\nu}\delta_{\alpha\mu}\delta_{\boldsymbol{R}_2\boldsymbol{R}'}\delta_{\boldsymbol{R}_1\boldsymbol{R}''}\Omega^0_{\boldsymbol{R}_2}(\boldsymbol{R}_1;t).\end{aligned} \tag{63}$$

We associate portions of $\tilde{\boldsymbol{j}}(\boldsymbol{x},t)$ with each lattice site in an obvious way,

$$\tilde{\boldsymbol{j}}(\boldsymbol{x},t)=\sum_{\boldsymbol{R}}\tilde{\boldsymbol{j}}_{\boldsymbol{R}}(\boldsymbol{x},t),$$

where

$$\tilde{\boldsymbol{j}}_{\boldsymbol{R}}(\boldsymbol{x},t) = \sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''} \tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t)\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t), \tag{64}$$

with

$$\tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},\boldsymbol{R};t) = \tfrac{1}{2}(\delta_{\boldsymbol{R}\boldsymbol{R}''}+\delta_{\boldsymbol{R}\boldsymbol{R}'})\tilde{\boldsymbol{j}}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x},t). \tag{65}$$

We then define

$$\tilde{m}^{j}_{\boldsymbol{R}}(\boldsymbol{x},t) \equiv \frac{1}{c}\int \alpha^{jk}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\tilde{j}^{k}_{\boldsymbol{R}}(\boldsymbol{y},t)d\boldsymbol{y}, \tag{66}$$

which, following the arguments used above for $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$, can be expected to be nonzero only for $\boldsymbol{x}$ close to $\boldsymbol{R}$. Using the relator properties (13), we obtain

$$\tilde{\boldsymbol{j}}_{\boldsymbol{R}}(\boldsymbol{x},t) = c\nabla\times\tilde{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t),$$

as desired. We can now write

$$\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t) \equiv \bar{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t) + \tilde{\boldsymbol{m}}_{\boldsymbol{R}}(\boldsymbol{x},t)$$

and, with

$$\boldsymbol{m}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}}\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t), \tag{67}$$

we can write the second of (58) as

$$\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle = \frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{m}(\boldsymbol{x},t) + \boldsymbol{j}_F(\boldsymbol{x},t). \tag{68}$$

E. Summary

We can now separate the "free" and "bound" charge and current densities, writing the total expectation values of the microscopic electronic charge and current density operators as

$$\langle\hat{\rho}(\boldsymbol{x},t)\rangle = \rho_B(\boldsymbol{x},t) + \rho_F(\boldsymbol{x},t),$$
$$\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle = \boldsymbol{j}_B(\boldsymbol{x},t) + \boldsymbol{j}_F(\boldsymbol{x},t). \tag{69}$$

The free charge density is given in terms of site charges $Q_{\boldsymbol{R}}(t)$ by (52) and the free current density in terms of link currents $I(\boldsymbol{R},\boldsymbol{R}';t)$ by (53); the site charges evolve via the link currents according to (49), and the link currents evolve according to (50). This guarantees that the free charge and current densities satisfy continuity (54). The bound charge and current densities are given by

$$\rho_B(\boldsymbol{x},t) = -\nabla\cdot\boldsymbol{p}(\boldsymbol{x},t),$$
$$\boldsymbol{j}_B(\boldsymbol{x},t) = \frac{\partial\boldsymbol{p}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{m}(\boldsymbol{x},t), \tag{70}$$

which guarantee that the bound charge and current densities satisfy continuity as well,

$$\nabla\cdot\boldsymbol{j}_B(\boldsymbol{x},t) + \frac{\partial\rho_B(\boldsymbol{x},t)}{\partial t} = 0. \tag{71}$$

The microscopic polarization and magnetization fields can be broken up into site contributions,

$$\boldsymbol{p}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}}\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t),$$
$$\boldsymbol{m}(\boldsymbol{x},t) = \sum_{\boldsymbol{R}}\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t). \tag{72}$$

The site polarizations are given by (55), or

$$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t) = \sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\left[\int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t)d\boldsymbol{y}\right] \times \eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t), \tag{73}$$

where $\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t)$ is given by (45), and we have used the first of (44). The site magnetizations are given by the sums of (56) and (66), or

$$m^{j}_{\boldsymbol{R}}(\boldsymbol{x},t) = \frac{1}{c}\sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\left[\int \alpha^{jk}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\big(j^{k}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t) + \tilde{j}^{k}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t)\big)d\boldsymbol{y}\right]\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t), \tag{74}$$

where $j^{k}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t)$ and $\tilde{j}^{k}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{y},\boldsymbol{R};t)$ are given by (46) and (65) respectively, and we have used the second of (44) and (64). Each of these quantities involve the single particle density matrix $\eta_{\alpha\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)$, the dynamics of which is governed by (37). We have omitted the spin degree of freedom and its contribution to the magnetization, but that could be easily included.

We emphasize that as long as in any approximations made the quantities $I(\boldsymbol{R},\boldsymbol{R}';t)$ that result still satisfy (51), and the evolution of the site charges is governed by (49), the approximate free charge and current densities will satisfy continuity. Moreover, for *any* approximations made in evaluating $\{\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)\}$ and $\{\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)\}$, the bound charge and current densities (70) will satisfy continuity (71). Thus, in this description, full charge conservation at the microscopic level is extremely robust against approximations.

III. SOME LIMITS OF INTEREST

A. The isolated atom limit

We first consider the limit where the Wannier functions $W_{\alpha\boldsymbol{R}}(\boldsymbol{x})$ and $W_{\beta\boldsymbol{R}'}(\boldsymbol{x})$ are assumed to have no common support if $\boldsymbol{R}\neq\boldsymbol{R}'$. Then we would expect our treatment of the solid to reduce to that of isolated atoms positioned at the lattice sites. That is, we would expect to find

$$\langle\hat{\rho}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}}\big(-\nabla\cdot\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t) + Q_{\boldsymbol{R}}\delta(\boldsymbol{x}-\boldsymbol{R})\big),$$
$$\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle = \sum_{\boldsymbol{R}}\left(\frac{\partial\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)}{\partial t} + \nabla\times\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)\right), \tag{75}$$

where $Q_{\boldsymbol{R}}$ is the fixed electronic charge associated with lattice site $\boldsymbol{R}$, and $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$ are the polarization and magnetization expressions we would expect from isolated atoms placed at the indicated lattice sites. In Appendix C, we review the usual results for an isolated atom, and in Appendix D, we show that indeed in the limit of no common support of Wannier functions at different sites our expressions do reduce to (75). Note that this holds even if the electromagnetic field varies strongly over the extension of the Wannier functions.

B. The long-wavelength limit

We next consider our equations in the long-wavelength limit, taking $\boldsymbol{E}(\boldsymbol{x},t)\rightarrow\boldsymbol{E}(t)$, and restrict ourselves to an infinite, periodic crystal. The magnetic field must then be time-independent, and we take any applied constant magnetic field to vanish; microscopic magnetic fields that have the periodicity of the lattice can be taken into account in the unperturbed Hamiltonian, $H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))$, using the expression (9). This is a standard model often used to calculate the optical response of crystals [10]. For the paths $C(\boldsymbol{x},\boldsymbol{y})$ in (12), we take straight lines.

With $\boldsymbol{B}(\boldsymbol{x},t)=0$ and $\boldsymbol{E}(t)$ uniform we have $\Delta(\boldsymbol{x},\boldsymbol{y},\boldsymbol{z};t)\rightarrow 0$ from (29), $\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x},t)\rightarrow W_{\alpha\boldsymbol{R}}(\boldsymbol{x})$ from (28) and the discussion preceding it, $\Omega^0_{\boldsymbol{x}}(\boldsymbol{y};t)\rightarrow(\boldsymbol{y}-\boldsymbol{x})\cdot\boldsymbol{E}(t)$ from (20) and the choice of a straight-line path, and $\mathfrak{p}^k(\boldsymbol{x},\boldsymbol{y};t)\rightarrow\mathfrak{p}^k(\boldsymbol{x})$ from (19). The dynamical equation (37) then simplifies to

$$\begin{aligned}i\hbar\frac{\partial\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t}&=\sum_{\lambda,\boldsymbol{R}''}\big(H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\eta_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)-\eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)H_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)\big)\\&\quad-e(\boldsymbol{R}-\boldsymbol{R}')\cdot\boldsymbol{E}(t)\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t),\end{aligned}\tag{76}$$

where from (35) we have used

$$\begin{aligned}\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)&\rightarrow H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)\\&=\int W^*_{\alpha\boldsymbol{R}}(\boldsymbol{x})H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_{\lambda\boldsymbol{R}''}(\boldsymbol{x})d\boldsymbol{x}\\&\quad-\frac{e}{2}\boldsymbol{E}(t)\cdot\int\big(W^*_{\alpha\boldsymbol{R}}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{R}'')W_{\lambda\boldsymbol{R}''}(\boldsymbol{x})\\&\quad+W^*_{\alpha\boldsymbol{R}}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{R})W_{\lambda\boldsymbol{R}''}(\boldsymbol{x})\big)d\boldsymbol{x},\end{aligned}\tag{77}$$

as well as the Hermiticity of $H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))$. Using the orthogonality of the Wannier functions (22) and translational invariance (23) we can manipulate this into the form

$$H_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)=H_{\alpha\lambda}(\boldsymbol{R}-\boldsymbol{R}'';t),$$

where

$$\begin{aligned}H_{\alpha\lambda}(\boldsymbol{R};t)&\equiv\int W^*_{\alpha}(\boldsymbol{x}-\boldsymbol{R})H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_{\lambda}(\boldsymbol{x})d\boldsymbol{x}\\&\quad-e\boldsymbol{E}(t)\cdot\int W^*_{\alpha}(\boldsymbol{x}-\boldsymbol{R})\boldsymbol{x}W_{\lambda}(\boldsymbol{x})d\boldsymbol{x}.\end{aligned}\tag{78}$$

We now look at the transform of $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ into crystal momentum space. Using the identities

$$\begin{aligned}\frac{\Omega_{uc}}{(2\pi)^3}\int_{BZ}e^{i\boldsymbol{k}\cdot(\boldsymbol{R}-\boldsymbol{R}')}d\boldsymbol{k}&=\delta_{\boldsymbol{R}\boldsymbol{R}'},\\\frac{\Omega_{uc}}{(2\pi)^3}\sum_{\boldsymbol{R}}e^{i(\boldsymbol{k}-\boldsymbol{k}')\cdot\boldsymbol{R}}&=\delta(\boldsymbol{k}-\boldsymbol{k}'),\end{aligned}\tag{79}$$

where we restrict $\boldsymbol{k}$ and $\boldsymbol{k}'$ to the first Brillouin zone, and $\boldsymbol{R}$ and $\boldsymbol{R}'$ are lattice sites, we introduce the Fourier transform

$$\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)\equiv\frac{\Omega_{uc}}{(2\pi)^3}\sum_{\boldsymbol{R},\boldsymbol{R}'}e^{i(\boldsymbol{k}'\cdot\boldsymbol{R}'-\boldsymbol{k}\cdot\boldsymbol{R})}\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t),\tag{80}$$

and from the dynamical Eq. (76) for $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ we obtain the corresponding equations for $\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)$,

$$\begin{aligned}i\hbar\frac{\partial\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)}{\partial t}&=\sum_{\gamma}\big(H_{\alpha\gamma}(\boldsymbol{k};t)\eta_{\gamma\boldsymbol{k};\beta\boldsymbol{k}'}(t)-\eta_{\alpha\boldsymbol{k};\gamma\boldsymbol{k}'}(t)H_{\gamma\beta}(\boldsymbol{k}';t)\big)\\&\quad-ie\boldsymbol{E}(t)\cdot\left(\frac{\partial}{\partial\boldsymbol{k}}+\frac{\partial}{\partial\boldsymbol{k}'}\right)\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t),\end{aligned}\tag{81}$$

where

$$H_{\alpha\gamma}(\boldsymbol{k};t)\equiv\sum_{\boldsymbol{R}}e^{-i\boldsymbol{k}\cdot\boldsymbol{R}}H_{\alpha\gamma}(\boldsymbol{R};t).\tag{82}$$

From the inverse Fourier transform of (80),

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)=\frac{\Omega_{uc}}{(2\pi)^3}\iint e^{-i(\boldsymbol{k}'\cdot\boldsymbol{R}'-\boldsymbol{k}\cdot\boldsymbol{R})}\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)d\boldsymbol{k}d\boldsymbol{k}',\tag{83}$$

where we have used (79), we see that if we have a state that shares the translational symmetry of the lattice, for which $\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ depends only on $(\boldsymbol{R}-\boldsymbol{R}')$, we have $\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)$ of the form

$$\eta_{\alpha\boldsymbol{k};\beta\boldsymbol{k}'}(t)=\eta_{\alpha\beta}(\boldsymbol{k};t)\delta(\boldsymbol{k}-\boldsymbol{k}').\tag{84}$$

If this holds initially then the dynamical Eq. (81) guarantees that it will hold at all later times, with

$$\begin{aligned}i\hbar\frac{\partial\eta_{\alpha\beta}(\boldsymbol{k};t)}{\partial t}&=\sum_{\gamma}\big(H_{\alpha\gamma}(\boldsymbol{k};t)\eta_{\gamma\beta}(\boldsymbol{k};t)-\eta_{\alpha\gamma}(\boldsymbol{k};t)H_{\gamma\beta}(\boldsymbol{k};t)\big)\\&\quad-ie\boldsymbol{E}(t)\cdot\frac{\partial\eta_{\alpha\beta}(\boldsymbol{k};t)}{\partial\boldsymbol{k}}.\end{aligned}\tag{85}$$

We can write (82) in a more familiar form by introduce a Bloch function associated with each Wannier function,

$$\phi_{\alpha\boldsymbol{k}}(\boldsymbol{x})=\sqrt{\frac{\Omega_{uc}}{(2\pi)^3}}\sum_{\boldsymbol{R}}e^{i\boldsymbol{k}\cdot\boldsymbol{R}}W_{\alpha\boldsymbol{R}}(\boldsymbol{x})=\frac{1}{\sqrt{(2\pi)^3}}e^{i\boldsymbol{k}\cdot\boldsymbol{x}}u_{\alpha\boldsymbol{k}}(\boldsymbol{x}).\tag{86}$$

They are orthonormal according to

$$\int\phi^*_{\alpha\boldsymbol{k}}(\boldsymbol{x})\phi_{\beta\boldsymbol{k}'}(\boldsymbol{x})d\boldsymbol{x}=\delta_{\alpha\beta}\delta(\boldsymbol{k}-\boldsymbol{k}'),$$

and in the second line of (86) we have introduced the periodic function $u_{\alpha\boldsymbol{k}}(\boldsymbol{x})$, where $u_{\alpha\boldsymbol{k}}(\boldsymbol{x})=u_{\alpha\boldsymbol{k}}(\boldsymbol{x}+\boldsymbol{R})$ for any lattice constant $\boldsymbol{R}$. Using the inverse relation of (86),

$$W_{\alpha\boldsymbol{R}}(\boldsymbol{x}) = \sqrt{\frac{\Omega_{uc}}{(2\pi)^3}}\int e^{-i\boldsymbol{k}\cdot\boldsymbol{R}}\phi_{\alpha\boldsymbol{k}}(\boldsymbol{x})d\boldsymbol{k}$$

in (78), we find we can write (82) as

$$H_{\alpha\gamma}(\boldsymbol{k};t) = H^0_{\alpha\gamma}(\boldsymbol{k}) - e\boldsymbol{\xi}_{\alpha\gamma}(\boldsymbol{k})\cdot\boldsymbol{E}(t), \tag{87}$$

where

$$\begin{aligned} H^0_{\alpha\gamma}(\boldsymbol{k}) &= \sum_{\boldsymbol{R}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}}\int W^*_\alpha(\boldsymbol{x}-\boldsymbol{R})H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_\gamma(\boldsymbol{x})d\boldsymbol{x} \\ &= \frac{1}{\Omega_{uc}}\int_{uc} u^*_{\alpha\boldsymbol{k}}(\boldsymbol{x})H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x})+\hbar\boldsymbol{k})u_{\gamma\boldsymbol{k}}(\boldsymbol{x})d\boldsymbol{x}, \end{aligned}$$

with the second integral ranging over any unit cell, and

$$\begin{aligned} \boldsymbol{\xi}_{\alpha\gamma}(\boldsymbol{k}) &= \sum_{\boldsymbol{R}} e^{-i\boldsymbol{k}\cdot\boldsymbol{R}}\int W^*_\alpha(\boldsymbol{x}-\boldsymbol{R})\boldsymbol{x}W_\gamma(\boldsymbol{x})d\boldsymbol{x} \\ &= \frac{i}{\Omega_{uc}}\int_{uc} u^*_{\alpha\boldsymbol{k}}(\boldsymbol{x})\frac{\partial u_{\gamma\boldsymbol{k}}(\boldsymbol{x})}{\partial\boldsymbol{k}}d\boldsymbol{x}, \end{aligned} \tag{88}$$

where $\boldsymbol{\xi}_{\alpha\gamma}(\boldsymbol{k})$ is a non-Abelian connection associated with the polarization; this object is discussed at length in earlier work, including, for example, Aversa [10] and Resta [12]. Dropping the matrix indices and writing, for example, $\eta(\boldsymbol{k};t)$ for the matrix with elements $\eta_{\alpha\beta}(\boldsymbol{k};t)$, we can then write (85) in the standard matrix form,

$$\begin{aligned} &i\hbar\frac{\partial\eta(\boldsymbol{k};t)}{\partial t} \\ &\quad = \left[H^0(\boldsymbol{k}) - e\boldsymbol{\xi}(\boldsymbol{k})\cdot\boldsymbol{E}(t), \eta(\boldsymbol{k};t)\right] - ie\boldsymbol{E}(t)\cdot\frac{\partial\eta(\boldsymbol{k},t)}{\partial\boldsymbol{k}}. \end{aligned} \tag{89}$$

When considering optical response, with the electric field treated as uniform, one is usually interested in the spatially averaged current density $\boldsymbol{J}(t)$. Returning to the expression (68) for $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$, since for excitation by a uniform electric field we expect this quantity to share the translational symmetry of the lattice, the spatial average of the magnetization term will vanish, and we will only recover contributions from the polarization $\boldsymbol{p}(\boldsymbol{x},t)$ and the free current density $\boldsymbol{j}_F(\boldsymbol{x},t)$. Using the decomposition (72) of $\boldsymbol{p}(\boldsymbol{x},t)$ into contributions from different lattice sites, and noting that the $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ for different $\boldsymbol{R}$ will be the same except for a translation associated with the difference in the lattice sites, we can introduce a spatially averaged polarization

$$\boldsymbol{P}(t) = \frac{1}{\Omega_{uc}}\int\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)d\boldsymbol{x}, \tag{90}$$

which will be the same regardless of the $\boldsymbol{R}$ chosen to evaluate it. Similarly decomposing the free current density (53) into contributions from different lattice sites,

$$\boldsymbol{j}_F(\boldsymbol{x},t) = \sum_{\boldsymbol{R}}\boldsymbol{j}_{F;\boldsymbol{R}}(\boldsymbol{x},t), \tag{91}$$

where

$$\boldsymbol{j}_{F;\boldsymbol{R}}(\boldsymbol{x},t) = \frac{1}{2}\sum_{\boldsymbol{R}'}\boldsymbol{s}(\boldsymbol{x};\boldsymbol{R},\boldsymbol{R}')I(\boldsymbol{R},\boldsymbol{R}';t), \tag{92}$$

we can introduce a free current density,

$$\boldsymbol{J}_F(t) = \frac{1}{\Omega_{uc}}\int\boldsymbol{j}_{F;\boldsymbol{R}}(\boldsymbol{x},t)d\boldsymbol{x}, \tag{93}$$

which will be independent of the $\boldsymbol{R}$ chosen to evaluate it. From (68), we then have

$$\boldsymbol{J}(t) = \frac{d\boldsymbol{P}(t)}{dt} + \boldsymbol{J}_F(t). \tag{94}$$

Using the expression (55,44) for $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\rho_{\boldsymbol{R}}(\boldsymbol{y},t)$, together with the form (12) for $\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})$, the use of the Fourier decomposition (83) yields

$$\boldsymbol{P}(t) = e\sum_{\alpha,\beta}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\boldsymbol{\xi}_{\beta\alpha}(\boldsymbol{k})\eta_{\alpha\beta}(\boldsymbol{k};t). \tag{95}$$

Similarly, performing the integral (53), and in the expression (50) for $I(\boldsymbol{R},\boldsymbol{R}';t)$ replacing the general $\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t)$ by $H_{\alpha\lambda}(\boldsymbol{R}-\boldsymbol{R}'';t)$ [see (78) and preceding], we obtain

$$\begin{aligned} \boldsymbol{J}_F(t) = \frac{e}{\hbar}\sum_{\alpha,\lambda}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\left[\frac{\partial}{\partial\boldsymbol{k}}\big(H^0_{\alpha\lambda}(\boldsymbol{k}) - e\boldsymbol{\xi}_{\alpha\lambda}(\boldsymbol{k})\cdot\boldsymbol{E}(t)\big)\right] \\ \times\,\eta_{\lambda\alpha}(\boldsymbol{k};t), \end{aligned} \tag{96}$$

where we have used (87) for the transform (82) $H_{\alpha\lambda}(\boldsymbol{k};t)$. In matrix form we write (95) and (96) as

$$\begin{aligned} \boldsymbol{P}(t) &= e\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[\boldsymbol{\xi}\eta(t)], \\ \boldsymbol{J}_F(t) &= \frac{e}{\hbar}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}\big[\big(\boldsymbol{\partial}(H^0 - e\boldsymbol{\xi}\cdot\boldsymbol{E}(t))\big)\eta(t)\big], \end{aligned} \tag{97}$$

where $\partial^n = \partial/\partial k^n$. Using these results in the expression (94) for $\boldsymbol{J}(t)$, together with the dynamical Eq. (89), we obtain

$$\boldsymbol{J}(t) = e\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[\boldsymbol{v}\eta(t)], \tag{98}$$

where the matrix

$$\boldsymbol{v} = \boldsymbol{\partial}H^0 - \frac{i}{\hbar}\left[\boldsymbol{\xi}, H^0\right]. \tag{99}$$

We note that these results can be derived via an entirely different strategy that begins directly in the long-wavelength limit and calculates the response of the system to an applied electric field $\boldsymbol{E}(t)$ [33]; such an approach is of course more direct and much easier if only the long-wavelength limit is desired.

1. Basis transformations

Rather than work with the basis functions $\{\phi_{\alpha\boldsymbol{k}}(\boldsymbol{x})\}$ of (86) at each $\boldsymbol{k}$ it is often convenient to work with a new set of basis functions $\{\bar{\phi}_{n\boldsymbol{k}}(\boldsymbol{x})\}$, related to the original set at each $\boldsymbol{k}$ by a unitary transformation,

$$\bar{\phi}_{n\boldsymbol{k}}(\boldsymbol{x}) = \sum_\alpha\phi_{\alpha\boldsymbol{k}}(\boldsymbol{x})U_{\alpha n}(\boldsymbol{k}), \tag{100}$$

where at each $\boldsymbol{k}$ the $U_{\alpha n}(\boldsymbol{k})$ are elements of a unitary matrix U. With

$$\begin{aligned} \bar{H}^0 &\equiv U^\dagger H^0 U, \\ \bar{\eta} &\equiv U^\dagger\eta U, \end{aligned}$$

in our short-hand notation, we find that the equation for $\bar{\eta}(t)$ that follows from (89) is

$$i\hbar\frac{\partial\bar{\eta}(t)}{\partial t}=\big[\bar{H}^0-e\bar{\boldsymbol{\xi}}\cdot\boldsymbol{E}(t),\bar{\eta}(t)\big]-ie\boldsymbol{E}(t)\cdot\boldsymbol{\partial}\bar{\eta}(t), \tag{101}$$

where

$$\bar{\boldsymbol{\xi}}\equiv U^{\dagger}\boldsymbol{\xi}U+iU^{\dagger}\boldsymbol{\partial}U, \tag{102}$$

and the matrix $\bar{\boldsymbol{\xi}}$ at $\boldsymbol{k}$ has components

$$\bar{\boldsymbol{\xi}}_{nm}(\boldsymbol{k})=\frac{i}{\Omega_{uc}}\int_{uc}\bar{u}^*_{n\boldsymbol{k}}(\boldsymbol{x})\frac{\partial\bar{u}_{m\boldsymbol{k}}(\boldsymbol{x})}{\partial\boldsymbol{k}}d\boldsymbol{x}, \tag{103}$$

[cf. (88)] where $\bar{u}_{n\boldsymbol{k}}(\boldsymbol{x})$ is the periodic component, $\bar{u}_{n\boldsymbol{k}}(\boldsymbol{x}+\boldsymbol{R})=\bar{u}_{n\boldsymbol{k}}(\boldsymbol{x})$ for any lattice vector $\boldsymbol{R}$, associated with the basis function $\bar{\phi}_{n\boldsymbol{k}}(\boldsymbol{x})$,

$$\bar{\phi}_{n\boldsymbol{k}}(\boldsymbol{x})=\frac{1}{\sqrt{(2\pi)^3}}e^{i\boldsymbol{k}\cdot\boldsymbol{x}}\bar{u}_{n\boldsymbol{k}}(\boldsymbol{x}). \tag{104}$$

Such a transformation is often done as a prelude to a perturbation calculation, and chosen so that $\bar{H}^0$ is diagonal, but more general transformations can be considered. In terms of the new matrices, we find that the current density (98) can be written as

$$\boldsymbol{J}(t)=e\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[\bar{\boldsymbol{v}}\bar{\eta}(t)], \tag{105}$$

where the matrix

$$\bar{\boldsymbol{v}}=\boldsymbol{\partial}\bar{H}^0-\frac{i}{\hbar}\big[\bar{\boldsymbol{\xi}},\bar{H}^0\big] \tag{106}$$

[cf. (99)]. Thus the form of both the dynamical equations (101) and the expression (105) for the current density are invariant under such a set of unitary transformations $\{U(\boldsymbol{k})\}$. The same does *not* hold for the polarization and free current density that lead to the current density via (94). In place of (97), we obtain

$$\begin{aligned}\boldsymbol{P}(t)&=e\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[\bar{\boldsymbol{\xi}}\bar{\eta}(t)]\\&\quad-ie\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[(U^{\dagger}\boldsymbol{\partial}U)\bar{\eta}(t)],\end{aligned} \tag{107}$$

$$\begin{aligned}\boldsymbol{J}_F(t)=&\frac{e}{\hbar}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}\big[\big(\boldsymbol{\partial}(\bar{H}^0-e\bar{\boldsymbol{\xi}}\cdot\boldsymbol{E}(t))\big)\bar{\eta}(t)\big]\\&+\frac{e}{\hbar}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}\big[[(U^{\dagger}\boldsymbol{\partial}U),(\bar{H}^0-e\bar{\boldsymbol{\xi}}\cdot\boldsymbol{E}(t))]\bar{\eta}(t)\big]\\&+i\frac{e^2}{\hbar}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\mathrm{Tr}[(\boldsymbol{E}(t)\cdot\boldsymbol{\partial})(U^{\dagger}\boldsymbol{\partial}U)\bar{\eta}(t)].\end{aligned} \tag{108}$$

It is only in the Bloch basis (86) associated with the maximally localized Wannier functions that our expressions for $\boldsymbol{P}(t)$ and $\boldsymbol{J}_F(t)$ take a simple form (97), since it is those Wannier functions that were used for the introduction of our quantities $\boldsymbol{p}(\boldsymbol{x},t)$, $\boldsymbol{m}(\boldsymbol{x},t)$, $\rho_F(\boldsymbol{x},t)$, and $\boldsymbol{j}_F(\boldsymbol{x},t)$. This kind of dependence on the details of a unitary transformation changing the Bloch basis is not unique to our approach; indeed, the appearance of the second term on the right-hand side of (107) arises as well in the "modern theory of polarization," even in the limit of a topologically trivial insulator, and is associated with the "quantum of ambiguity" [15]. In the following section, we show that for a topologically trivial insulator we find the same expressions for the ground-state polarization and magnetization as in the "modern theory of polarization and magnetization," and it is easy to show that our expression for $\boldsymbol{J}_F$ in such a ground state vanishes even more generally, as expected. We will turn to our expressions for the ground-state polarization and magnetization for metals and topologically nontrivial insulators in a future publication.

2. *Perturbative calculation*

We close this section by using our approach to calculate the linear response of an insulator in the long-wavelength limit. We choose the transformation (100) to be that which diagonalizes the Hamiltonian, so

$$\bar{H}^0_{nm}(\boldsymbol{k})=\delta_{nm}\hbar\omega_n(\boldsymbol{k}),$$

and introducing a perturbation expansion of $\eta_{nm}(\boldsymbol{k};t)$ in orders of the electric field,

$$\eta_{nm}(\boldsymbol{k};t)=\eta^{(0)}_{nm}(\boldsymbol{k})+\eta^{(1)}_{nm}(\boldsymbol{k};t)+\dots$$

we take

$$\eta^{(0)}_{nm}(\boldsymbol{k})=f_n\delta_{nm},$$

where $f_n=0$ or 1 for each n, independent of $\boldsymbol{k}$. For an electric field

$$\boldsymbol{E}(t)=\boldsymbol{E}(\omega)e^{-i\omega t}+\boldsymbol{E}(-\omega)e^{i\omega t},$$

with $\boldsymbol{E}(-\omega)=\boldsymbol{E}^*(\omega)$, the usual perturbation treatment of (101) gives

$$\eta^{(1)}_{nm}(\boldsymbol{k};t)=\eta^{(1)}_{nm}(\boldsymbol{k};\omega)e^{-i\omega t}+\eta^{(1)}_{nm}(\boldsymbol{k};-\omega)e^{i\omega t},$$

where

$$\eta^{(1)}_{nm}(\boldsymbol{k};\omega)=\frac{e\bar{\boldsymbol{\xi}}_{nm}(\boldsymbol{k})\cdot\boldsymbol{E}(\omega)f_{mn}}{\hbar(\omega_{nm}(\boldsymbol{k})-\omega)},$$

where $f_{mn}=f_m-f_n$ and likewise for $\omega_{nm}(\boldsymbol{k})$. Then either calculating the first-order result for

$$\boldsymbol{J}^{(1)}(t)=\boldsymbol{J}(\omega)e^{-i\omega t}+\boldsymbol{J}(-\omega)e^{i\omega t}$$

directly from (105), or calculating the first order contributions to $\boldsymbol{P}(t)$ and $\boldsymbol{J}_F(t)$ from (105) and (106) and using the expressions in (94), we obtain

$$J^a(\omega)=\sigma^{ab}(\omega)E^b(\omega),$$

where the conductivity $\sigma^{ab}(\omega)$ is given by

$$\begin{aligned}\sigma^{ab}(\omega)=&-i\omega\frac{e^2}{\hbar}\sum_{n,m}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_m\omega_{nm}\big(\bar{\xi}^a_{mn}\bar{\xi}^b_{nm}+\bar{\xi}^b_{mn}\bar{\xi}^a_{nm}\big)}{\big(\omega^2_{nm}-\omega^2\big)}\\&-i\frac{e^2}{\hbar}\sum_{n,m}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_m\omega^2_{nm}\big(\bar{\xi}^a_{mn}\bar{\xi}^b_{nm}-\bar{\xi}^b_{mn}\bar{\xi}^a_{nm}\big)}{\big(\omega^2_{nm}-\omega^2\big)}.\end{aligned}$$

Note that in general the conductivity can be finite as $\omega\rightarrow 0$,

$$\lim_{\omega\rightarrow 0}\sigma^{ab}(\omega)=\frac{e^2}{\hbar}\sum_m\int\frac{d\boldsymbol{k}}{(2\pi)^3}f_m\left(\frac{\partial\bar{\xi}^a_{mm}}{\partial k^b}-\frac{\partial\bar{\xi}^b_{mm}}{\partial k^a}\right), \tag{109}$$

which is the well-known expression for the zero frequency transverse conductivity of a topologically nontrivial insulator arising from the Kubo formula[8] [28,29,34]; here we have used

$$\boldsymbol{\partial}\times\boldsymbol{\xi}-i\boldsymbol{\xi}\times\boldsymbol{\xi}=0,$$

and the fact that the corresponding equation holds for $\bar{\boldsymbol{\xi}}$ as well. Note that the zero frequency transverse conductivity is identified in our treatment with the "free" current density $\boldsymbol{J}_F$, as follows immediately from the expression (94) for $\boldsymbol{J}(t)$. This zero frequency response vanishes for a topologically trivial insulator; in fact, we show in Section II E that in linear response the full microscopic free current density $\boldsymbol{j}_F(\boldsymbol{x},t)$ of a topologically trivial insulator vanishes. In the long-wavelength limit being considered here, the only linear response of a topologically trivial insulator is from the polarization, $\boldsymbol{J}^{(1)}(t)=d\boldsymbol{P}^{(1)}(t)/dt$, where

$$\boldsymbol{P}^{(1)}(t)=\boldsymbol{P}(\omega)e^{-i\omega t}+\boldsymbol{P}(-\omega)e^{i\omega t},$$

and

$$\boldsymbol{P}(\omega)=\frac{e^2}{\hbar}\sum_{n,m}\int\frac{d\boldsymbol{k}}{(2\pi)^3}\frac{f_{mn}\bar{\boldsymbol{\xi}}_{mn}(\boldsymbol{k})(\bar{\boldsymbol{\xi}}_{nm}(\boldsymbol{k})\cdot\boldsymbol{E}(\omega))}{(\omega_{nm}(\boldsymbol{k})-\omega)},$$

the usual result from perturbation theory [10].

C. Ground-state moments in a topologically trivial insulator

Next we consider both the electric and magnetic dipole moments associated with the lattice sites in the ground state, restricting ourselves to a topologically trivial insulator at zero temperature. In such a system, all bands are either completely empty or completely filled, and each set of energy overlapping bands is topologically trivial as a whole. Then a maximally localized set of Wannier functions are associated with the filled bands, and another set with the empty bands [24–27], so we have

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}=f_\alpha\delta_{\alpha\beta}\delta_{\boldsymbol{R}\boldsymbol{R}'},$$

where $f_\alpha=0$ or 1, in the expressions (73) and (74) for $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$. Those quantities are then of course independent of time, and the expressions simplify considerably. Taking straight-line paths for $C(\boldsymbol{x},\boldsymbol{y})$ (see Appendix A), we have

$$\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})d\boldsymbol{w}=(x^i-y^i),$$

$$\int\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})d\boldsymbol{w}=\tfrac{1}{2}\epsilon^{jmk}(x^m-y^m),$$

and we find that the electric dipole moment $\boldsymbol{\mu}$ and magnetic dipole moment $\boldsymbol{\nu}$ of the charge-current distribution associated with each site are just

$$\boldsymbol{\mu}=\int\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x},$$

$$\boldsymbol{\nu}=\int\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x},$$

and are independent of $\boldsymbol{R}$, as expected. We obtain

$$\boldsymbol{\mu}=\sum_\alpha f_\alpha\boldsymbol{\mu}_\alpha,$$

$$\boldsymbol{\nu}=\sum_\alpha f_\alpha\boldsymbol{\nu}_\alpha,$$

where

$$\boldsymbol{\mu}_\alpha=e\int W_\alpha^*(\boldsymbol{x})\boldsymbol{x}W_\alpha(\boldsymbol{x})d\boldsymbol{x},$$

with $W_\alpha(\boldsymbol{x})\equiv W_{\alpha\boldsymbol{0}}(\boldsymbol{x})$, while $\boldsymbol{\nu}_\alpha$ is the sum of two contributions,

$$\boldsymbol{\nu}_\alpha=\bar{\boldsymbol{\nu}}_\alpha+\tilde{\boldsymbol{\nu}}_\alpha,$$

arising from the two contributions to $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x})$ in (74). The first term is an atomic-like contribution,

$$\bar{\nu}_\alpha^j=\frac{1}{4c}\epsilon^{jmk}\int x^mW_\alpha^*(\boldsymbol{x})\big(J^k(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_\alpha(\boldsymbol{x})\big)d\boldsymbol{x}+\frac{1}{4c}\epsilon^{jmk}\int x^m\big(J^k(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_\alpha(\boldsymbol{x})\big)^*W_\alpha(\boldsymbol{x})d\boldsymbol{x},\tag{110}$$

which, for a Hamiltonian of Schrödinger form (10), gives

$$\bar{\boldsymbol{\nu}}_\alpha=\frac{e}{2mc}\int W_\alpha^*(\boldsymbol{x})\left[\boldsymbol{x}\times\left(\frac{\hbar}{i}\frac{\partial W_\alpha(\boldsymbol{x})}{\partial\boldsymbol{x}}-\frac{e}{c}\boldsymbol{A}_{\text{static}}(\boldsymbol{x})W_\alpha(\boldsymbol{x})\right)\right]d\boldsymbol{x},$$

which is familiar from the corresponding expression in atomic physics, and

$$\tilde{\nu}_\alpha^j=\frac{1}{2\hbar c}\epsilon^{jmk}\sum_{\lambda,\boldsymbol{R}_1}R_1^m\text{Im}\big[H^{(0)}_{\alpha\boldsymbol{0};\lambda\boldsymbol{R}_1}\mu^k_{\lambda\boldsymbol{R}_1;\alpha\boldsymbol{0}}(\boldsymbol{R}_1)\big],\tag{111}$$

where $H^{(0)}_{\alpha\boldsymbol{0};\lambda\boldsymbol{R}_1}$ is given by (77) in the limit of no applied field, or more generally by

$$H^{(0)}_{\alpha\boldsymbol{R}_1;\lambda\boldsymbol{R}_2}\equiv\int W^*_{\alpha\boldsymbol{R}_1}(\boldsymbol{x})H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x}))W_{\lambda\boldsymbol{R}_2}(\boldsymbol{x})d\boldsymbol{x},\tag{112}$$

where we have defined

$$\boldsymbol{\mu}_{\beta\boldsymbol{R}_1;\alpha\boldsymbol{R}_2}(\boldsymbol{R})=e\int W^*_{\beta\boldsymbol{R}_1}(\boldsymbol{x})(\boldsymbol{x}-\boldsymbol{R})W_{\alpha\boldsymbol{R}_2}(\boldsymbol{x})d\boldsymbol{x}.$$

Here, $\tilde{\boldsymbol{\nu}}_\alpha$ is the itinerant contribution to magnetic moment defined earlier [14]; taking the macroscopic polarization and magnetization to be given by

$$\boldsymbol{P}=\frac{\boldsymbol{\mu}}{\Omega_{uc}},\quad\boldsymbol{M}=\frac{\boldsymbol{\nu}}{\Omega_{uc}}.$$

We are in agreement with earlier results from the modern theory of polarization and magnetization [34]. In a calculation such as this, using the basis of Wannier functions, the quantum of ambiguity arises because one must choose the lattice site with which a representative Wannier function of given type α is associated; the lattice sites with which the rest of the Wannier functions of that type are associated follow from translational symmetry.

D. Perturbative result for a topologically trivial insulator

We finally consider the general nature of the linear response of a topologically trivial insulator to an applied electromagnetic field. We look at the expression (50) for the link

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[8] In 2D, this is proportional to the net Chern number of the filled bands.

current and expand it into terms involving powers of the electromagnetic field,

$$I(\boldsymbol{R},\boldsymbol{R}';t)=I^{(0)}(\boldsymbol{R},\boldsymbol{R}')+I^{(1)}(\boldsymbol{R},\boldsymbol{R}';t)+\cdots,$$

which is achieved by expanding the terms of which it is composed in a similar way,

$$\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)=\bar{H}^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}+\bar{H}^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)+\cdots,$$
$$\eta_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)=\eta^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}+\eta^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)+\cdots$$

For a topologically trivial insulator, we have

$$\eta^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}=f_\alpha\delta_{\alpha\lambda}\delta_{\boldsymbol{R}\boldsymbol{R}'}\tag{113}$$

as above, and we identify $\bar{H}^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}=H^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}$, with $H^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}$ given by (112). Then it immediately follows that $I^{(0)}(\boldsymbol{R},\boldsymbol{R}')=0$, while

$$\begin{aligned}I^{(1)}&(\boldsymbol{R},\boldsymbol{R}';t)\\&=\frac{e}{i\hbar}\sum_{\alpha,\lambda}\big(\bar{H}^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)\eta^{(0)}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)-\eta^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)\bar{H}^{(1)}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\big)\\&\quad+\frac{e}{i\hbar}\sum_{\alpha,\lambda}\big(H^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}\eta^{(1)}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)-\eta^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)H^{(0)}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}\big).\end{aligned}\tag{114}$$

The first of these two terms gives

$$\frac{e}{i\hbar}\sum_{\alpha,\lambda}\big(\bar{H}^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)-\bar{H}^{(1)}_{\lambda\boldsymbol{R}';\alpha\boldsymbol{R}}(t)\big)f_\alpha\delta_{\alpha\lambda}\delta_{\boldsymbol{R}\boldsymbol{R}'},$$

which vanishes regardless of the form of $\bar{H}^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)$. To investigate the second term in (114), we look at the equation for $\eta^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}'}(t)$ that follows from a perturbative analysis of the general dynamical Eq. (37). Expanding terms in the usual way, we obtain

$$\begin{aligned}i\hbar\frac{\partial\eta^{(1)}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)}{\partial t}&=\sum_{\lambda,\boldsymbol{R}''}\big(H^{(0)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}\eta^{(1)}_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}(t)-\eta^{(1)}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}H^{(0)}_{\lambda\boldsymbol{R}'';\beta\boldsymbol{R}'}\big)\\&\quad+f_{\beta\alpha}H^{(1)}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'},\end{aligned}$$

and the inhomogeneous term in this equation is proportional to $f_{\beta\alpha}$. As can be confirmed by the formal solution of this equation in reciprocal space, $\eta^{(1)}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ will vanish unless one and only one of α and β is associated with a set of filled bands, so indeed $\eta^{(1)}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t)$ will be proportional to $f_{\alpha\beta}$. However, using this result in the second term of (114), we see that the second term will involve terms such as $f_{\alpha\beta}H^{(0)}_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}$, which vanish. So we have $I^{(1)}(\boldsymbol{R},\boldsymbol{R}';t)=0$.

Thus, to first order in the optical response of a topologically trivial insulator at zero temperature, the link current will vanish, and so we see from (49), (52), and (53) that $\boldsymbol{j}_F(\boldsymbol{x},t)=0$ and $\rho_F(\boldsymbol{x},t)$ is independent of time. The entire optical response to first order is described by $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$, even if the wavelength of light is comparable to or smaller than the lattice spacing.

IV. CONCLUSION

We have presented a general strategy for constructing microscopic polarization and magnetization fields, $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$, which together with microscopic "free" charge and current densities, $\rho_F(\boldsymbol{x},t)$ and $\boldsymbol{j}_F(\boldsymbol{x},t)$, can be used (2) to represent the expectation value of the microscopic electronic charge and current density operators, $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$. Our goal has been to write $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$ as sums over contributions from different sites (3), and we have done this by associating a set of localized orbitals with each site. In a periodic crystal, which has been the focus of our work, we have taken these to be maximally localized Wannier functions, each set of such functions associated with a set of bands that is topologically trivial; schemes exist to construct such Wannier functions, which are primarily *ab initio* based [35,36], and can readily be used to implement the formalism presented. The description that results is one of a lattice gauge theory, where the free charge and current densities are described by site charges and link currents, respectively; the polarization and magnetization fields associated with a given lattice site are then written in terms of the single-particle density operators associated with that site and its neighbors, and can be expected to be nonvanishing only in the neighborhood of the site.

While our site quantities $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t)$, together with our free charge and current densities, have been defined to include only valence and conduction electron contributions, the contributions from ion cores can be identified as well. In the simple case where the ions are considered fixed and approximated as point particles, there is a time-independent ionic charge density

$$\rho^{\text{ion}}(\boldsymbol{x})=\sum_{\boldsymbol{R}}\rho^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x}),$$

where

$$\rho^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x})=\sum_N q_N\delta(\boldsymbol{x}-\boldsymbol{R}-\boldsymbol{d}_N),$$

and where we assume that in each unit cell there are ions with charges q_N located at $\boldsymbol{R}+\boldsymbol{d}_N$. Following the strategy used for electrons, we can write $\rho^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x})$ as a contribution that would arise if all the ions were at the lattice site, and a time independent polarization,

$$\rho^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x})=\delta(\boldsymbol{x}-\boldsymbol{R})\sum_N q_N-\nabla\cdot\boldsymbol{p}^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x}),$$

where

$$\boldsymbol{p}^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x})=\int\boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\rho^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{y})d\boldsymbol{y}$$

[compare (55)]. The contributions of the ions can then be taken into account in our summary equations in Sec. II E by replacing the old $\rho_F(\boldsymbol{x},t)$ by

$$\rho^{\text{new}}_F(\boldsymbol{x},t)=\rho_F(\boldsymbol{x},t)+\sum_{N,\boldsymbol{R}}q_N\delta(\boldsymbol{x}-\boldsymbol{R})$$

in (69), and by replacing the old $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)$ by

$$\boldsymbol{p}^{\text{new}}_{\boldsymbol{R}}(\boldsymbol{x},t)=\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)+\boldsymbol{p}^{\text{ion}}_{\boldsymbol{R}}(\boldsymbol{x})$$

in (72). Note that in equilibrium, $\rho^{\text{new}}_F(\boldsymbol{x},t)$ vanishes and all the microscopic charge is associated with the polarization fields of valence electrons and ions. If the motion of the ions is also considered then additional terms, including contributions to the magnetization, appear as they did for valence and conduction electrons.

To move to macroscopic electrodynamics, spatial averages of the microscopic polarization and magnetization can now be identified as the macroscopic polarization and magnetization fields, and expansions of the polarization and magnetization fields associated with a site can be used to identify its electric and magnetic multipole moments; we plan to address these matters in a following publication. Benefits of this approach include the fact that the description of the interaction of the charges with the electromagnetic field involves the electric and magnetic fields themselves, rather than the scalar and vector potentials that describe them, and the fact that the structure of the expressions for $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$ in terms of their "constituent sources" $\boldsymbol{p}(\boldsymbol{x},t)$, $\boldsymbol{m}(\boldsymbol{x},t)$, $\rho_F(\boldsymbol{x},t)$, and $\boldsymbol{j}_F(\boldsymbol{x},t)$ are such that continuity is guaranteed by construction, even if approximations are made in describing those constituent sources.

We have restricted ourselves to electrons described in the independent particle approximation, with no interactions besides those that can be included in a mean-field treatment of the electromagnetic field. However, while including interactions between the electrons will make the description of the dynamics more difficult, much of the kinematics associated with defining the constituent sources will remain unchanged. Other simplifications we have employed, such as the omission of the contribution of the electron spin to the magnetization, would be easier to remedy. Also, although we have formulated our theory in a three-dimensional space, it is readily applicable to a two dimensional lattice, using some care in formulating the Fourier transforms and Wannier functions.

For a given $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$, there is not a unique way to construct the constituent sources, even if the set of localized orbitals being employed is fixed. For example, we have restricted ourselves to line integral forms (12) of the quantities that are responsible for relating $\boldsymbol{p}(\boldsymbol{x},t)$ and $\boldsymbol{m}(\boldsymbol{x},t)$ to $\langle\hat{\rho}(\boldsymbol{x},t)\rangle$ and $\langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle$. Yet the essential feature of those relators is only that they satisfy (13), not that they are of line integral form. And even within the line integral form we have used straight line paths in our examples, although the equations we derive are more general. The straight line path does seem the most natural, since it can be shown that it leads most naturally to the usual multipole expansion, but other paths would be worth exploring. The issue then is *not* what are the "correct" constituent sources $\boldsymbol{p}(\boldsymbol{x},t)$, $\boldsymbol{m}(\boldsymbol{x},t)$, $\rho_F(\boldsymbol{x},t)$, and $\boldsymbol{j}_F(\boldsymbol{x},t)$, since they cannot be uniquely defined, but whether or not a particular choice is convenient.

We have shown that our choice exhibits a number of interesting features: First, in a limit where the lattice sites are moved further away from each other, with the set of orbitals employed remaining fixed, our description flows naturally into that of a set of "isolated atoms" on a lattice, regardless of the wavelength of light. Second, in the long-wavelength limit of a uniform applied electric field, we found that the spatially averaged current density is the sum of a free current part and a contribution from the time derivative of the polarization. The first of these is responsible, for instance, for the transverse dc conductivity in a topologically nontrivial insulator; the second is the sole contribution to the linear response of a topologically trivial insulator. More generally, in a topologically trivial insulator, the linear optical response is due *solely* to induced microscopic polarization and magnetization fields, again regardless of the wavelength of light. This is as expected, since it is only to higher order that one would physically expect that injected quasiparticles could be driven by the electromagnetic field and lead to induced free charges and currents. Finally, we showed that in the ground state of a topologically trivial insulator the expressions for the polarization and the magnetization agree with results from the "modern theory of polarization and magnetization."

We believe these features suggest that our choice of constituent sources is worth developing further as a description of the ground state of systems of interest, and of the linear and nonlinear response of matter to radiation very generally. And we believe that the overall framework we have established here for introducing microscopic polarization and magnetization fields in extended systems will prove to be valuable for studying electronic dynamics at atomic scales.

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APPENDIX A: RELATOR EQUATIONS

We give a derivation of the relations (13) between relators that follows the spirit of Healy's [21], although our notation is different. We characterize the path from $\boldsymbol{y}$ to $\boldsymbol{x}$ by a function $\boldsymbol{z}(u)$ such that $\boldsymbol{z}(u_1)=\boldsymbol{y}$ and $\boldsymbol{z}(u_2)=\boldsymbol{x}$. Then from the first of (12), we have

$$s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\int_{u_1}^{u_2}du\frac{dz^i(u)}{du}\delta(\boldsymbol{w}-\boldsymbol{z}), \tag{A1}$$

and so

$$\begin{aligned}-\frac{\partial s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^i}&=-\int_{u_1}^{u_2}du\frac{dz^i(u)}{du}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^i}\\&=\int_{u_1}^{u_2}du\frac{dz^i(u)}{du}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial z^i}\\&=\int_{u_1}^{u_2}du\frac{d}{du}\delta(\boldsymbol{w}-\boldsymbol{z})\\&=\delta(\boldsymbol{w}-\boldsymbol{y})-\delta(\boldsymbol{w}-\boldsymbol{x}),\end{aligned}$$

which is the first of (13).

Moving to the second of (12), we have

$$\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\epsilon^{jmn}\int_{u_1}^{u_2}du\frac{dz^m}{du}\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z}), \tag{A2}$$

so

$$\epsilon^{ipj}\frac{\partial\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}=\epsilon^{ipj}\epsilon^{jmn}\int_{u_1}^{u_2}du\frac{dz^m}{du}\frac{\partial z^n}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^p}.$$

Now

$$\epsilon^{ipj}\epsilon^{jmn}=\epsilon^{jip}\epsilon^{jmn}=\delta^{im}\delta^{pn}-\delta^{in}\delta^{pm},$$

so

$$
\begin{aligned}
&\epsilon^{ipj}\frac{\partial\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}\\
&\quad= (\delta^{im}\delta^{pn}-\delta^{in}\delta^{pm})\int_{u_1}^{u_2} du\frac{dz^m}{du}\frac{\partial z^n}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^p}\\
&\quad= \int_{u_1}^{u_2} du\frac{dz^i}{du}\frac{\partial z^p}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^p}-\int_{u_1}^{u_2} du\frac{dz^p}{du}\frac{\partial z^i}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^p}\\
&\quad= -\int_{u_1}^{u_2} du\frac{dz^i}{du}\frac{\partial z^p}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial z^p}+\int_{u_1}^{u_2} du\frac{dz^p}{du}\frac{\partial z^i}{\partial x^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial z^p}\\
&\quad= -\int_{u_1}^{u_2} du\frac{dz^i}{du}\frac{\partial}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})+\int_{u_1}^{u_2} du\frac{\partial z^i}{\partial x^k}\frac{d}{du}\delta(\boldsymbol{w}-\boldsymbol{z}).
\end{aligned}
$$

Partially integrating the second term gives

$$
\begin{aligned}
&\epsilon^{ipj}\frac{\partial\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}\\
&\quad= -\int_{u_1}^{u_2} du\frac{dz^i}{du}\frac{\partial}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&\qquad-\int_{u_1}^{u_2} du\left(\frac{\partial}{\partial x^k}\frac{dz^i}{du}\right)\delta(\boldsymbol{w}-\boldsymbol{z})+\left[\frac{\partial z^i}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\right]_{u_1}^{u_2}\\
&\quad= -\frac{\partial}{\partial x^i}\left(\int_{u_1}^{u_2} du\frac{dz^i}{du}\delta(\boldsymbol{w}-\boldsymbol{z})\right)+\left[\frac{\partial z^i}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\right]_{u_1}^{u_2}.
\end{aligned}
$$

In the second term note the $z^i(u_2)=x^i$ and $z^i(u_1)=y^i$, so there will only be a contribution at u_2. Then recognizing the first term from (A1), we have

$$
\epsilon^{ipj}\frac{\partial\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}=-\frac{\partial}{\partial x^k}s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})+\delta^{ik}\delta(\boldsymbol{w}-\boldsymbol{x}),
$$

which is the second of (13).

The final expression is derived in much the same way. We have

$$
\beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\epsilon^{jmn}\int_{u_1}^{u_2} du\frac{dz^m}{du}\frac{\partial z^n}{\partial y^k}\delta(\boldsymbol{w}-\boldsymbol{z}), \tag{A3}
$$

so

$$
\begin{aligned}
&\epsilon^{ipj}\frac{\partial\beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}\\
&\quad= \epsilon^{ipj}\epsilon^{jmn}\int_{u_1}^{u_2} du\frac{dz^m}{du}\frac{\partial z^n}{\partial y^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&\quad= (\delta^{im}\delta^{pn}-\delta^{in}\delta^{pm})\int_{u_1}^{u_2} du\frac{dz^m}{du}\frac{\partial z^n}{\partial y^k}\frac{\partial\delta(\boldsymbol{w}-\boldsymbol{z})}{\partial w^p},
\end{aligned}
$$

and following exactly the strategy above we have

$$
\begin{aligned}
&\epsilon^{ipj}\frac{\partial\beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})}{\partial w^p}\\
&\quad= -\frac{\partial}{\partial y^i}\left(\int_{u_1}^{u_2} du\frac{dz^i}{du}\delta(\boldsymbol{w}-\boldsymbol{z})\right)+\left[\frac{\partial z^i}{\partial y^k}\delta(\boldsymbol{w}-\boldsymbol{z})\right]_{u_1}^{u_2}\\
&\quad= -\frac{\partial}{\partial y^i}s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})-\delta^{ik}\delta(\boldsymbol{w}-\boldsymbol{y}),
\end{aligned}
$$

because in the last term only the contribution from u_1 will survive. This is the third of (13).

Next we confirm that the relations (14) hold for symmetric paths $C(\boldsymbol{x},\boldsymbol{y})$, where for each and every $\boldsymbol{x}$ and $\boldsymbol{y}$ the path $C(\boldsymbol{x},\boldsymbol{y})$ is the "reverse" of the path $C(\boldsymbol{y},\boldsymbol{x})$. More precisely, if $C(\boldsymbol{x},\boldsymbol{y})$ is specified by giving $\boldsymbol{z}(u)$ as u varies from u_1 to u_2, with $\boldsymbol{z}(u_1)=\boldsymbol{y}$ and $\boldsymbol{z}(u_2)=\boldsymbol{x}$, then $C(\boldsymbol{y},\boldsymbol{x})$ is specified by the same $\boldsymbol{z}(u)$ as u varies from u_2 to u_1. We have

$$
s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\int_{C(\boldsymbol{x},\boldsymbol{y})}dz^i\delta(\boldsymbol{w}-\boldsymbol{z})=\int_{u_1}^{u_2} du\frac{dz^i(u)}{du}\delta(\boldsymbol{w}-\boldsymbol{z})
$$

and

$$
\begin{aligned}
s^i(\boldsymbol{w};\boldsymbol{y},\boldsymbol{x})&=\int_{C(\boldsymbol{y},\boldsymbol{x})}dz^i\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=\int_{u_2}^{u_1} du\frac{dz^i(u)}{du}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=-\int_{u_1}^{u_2} du\frac{dz^i(u)}{du}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=-s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}),
\end{aligned}
$$

while

$$
\begin{aligned}
\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})&=\epsilon^{jmn}\int_{C(\boldsymbol{x},\boldsymbol{y})}dz^m\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=\epsilon^{jmn}\int_{u_1}^{u_2}\frac{dz^m(u)}{du}\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})
\end{aligned}
$$

and

$$
\begin{aligned}
\beta^{jk}(\boldsymbol{w};\boldsymbol{y},\boldsymbol{x})&=\epsilon^{jmn}\int_{C(\boldsymbol{y},\boldsymbol{x})}dz^m\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=\epsilon^{jmn}\int_{u_2}^{u_1} du\frac{dz^m(u)}{du}\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=-\epsilon^{jmn}\int_{u_1}^{u_2} du\frac{dz^m(u)}{du}\frac{\partial z^n}{\partial x^k}\delta(\boldsymbol{w}-\boldsymbol{z})\\
&=-\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}).
\end{aligned}
$$

Finally, we consider the special case of a straight-line path $C(\boldsymbol{x},\boldsymbol{y})$ for each $\boldsymbol{x}$ and $\boldsymbol{y}$. That is, taking $u_1=0$ and $u_2=1$, for the path $C(\boldsymbol{x},\boldsymbol{y})$, we have

$$
\boldsymbol{z}=\boldsymbol{y}+u(\boldsymbol{x}-\boldsymbol{y}).
$$

Then we have

$$
\begin{aligned}
\frac{dz^i(u)}{du}&=x^i-y^i,\\
\frac{\partial z^n}{\partial x^k}&=u\delta^{nk},\\
\frac{\partial z^n}{\partial y^k}&=(1-u)\delta^{nk},
\end{aligned}
$$

and so from (A1) we have

$$
s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\int_0^1(x^i-y^i)\delta(\boldsymbol{w}-\boldsymbol{y}-u(\boldsymbol{x}-\boldsymbol{y}))du, \tag{A4}
$$

while from (A2) we have

$$
\alpha^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y})=\epsilon^{jmk}\int_0^1(x^m-y^m)\delta(\boldsymbol{w}-\boldsymbol{y}-u(\boldsymbol{x}-\boldsymbol{y}))udu, \tag{A5}
$$

and from (A3) we have

$$\beta^{jk}(\boldsymbol{w};\boldsymbol{x},\boldsymbol{y}) = \epsilon^{jmk}\int_0^1 (x^m - y^m)\delta(\boldsymbol{w}-\boldsymbol{y}-u(\boldsymbol{x}-\boldsymbol{y}))(1-u)du. \tag{A6}$$

Since a straight-line path is symmetric, in the terminology used above, we expect (A4)–(A6) to satisfy (14), and it is easy to confirm that they do.

APPENDIX B: ORTHOGONALIZATION OF STATES

In this Appendix, we use a short-hand notation, taking

$$(\alpha\boldsymbol{R}) \to n, \quad \boldsymbol{R} \to \boldsymbol{R}_n.$$

Our set of nonorthogonal states (24) are then labeled $\{W_n'(\boldsymbol{x},t)\}$; the elements of the overlap matrix $\mathbb{S}(t)$ characterizing them are

$$S_{nm}(t) \equiv \int W_n'^{*}(\boldsymbol{x},t)W_m'(\boldsymbol{x},t)d\boldsymbol{x} = e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)}\int W_n^*(\boldsymbol{x})e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{x},\boldsymbol{R}_m;t)}W_m(\boldsymbol{x})d\boldsymbol{x}.$$

Clearly $\mathbb{S}(t)$ is Hermitian, and although its matrix elements are in general not gauge-invariant, they can be written as

$$S_{nm}(t) = e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)}\hat{S}_{nm}(t),$$

where the $\hat{S}_{nm}(t)$ are gauge-invariant,

$$\hat{S}_{nm}(t) = \int W_n^*(\boldsymbol{x})e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{x},\boldsymbol{R}_m;t)}W_m(\boldsymbol{x})d\boldsymbol{x}$$

and since

$$\hat{S}^*_{nm}(t) = e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)}S^*_{nm}(t) = e^{-i\Phi(\boldsymbol{R}_m,\boldsymbol{R}_n;t)}S_{mn}(t) = \hat{S}_{mn}(t)$$

the matrix $\hat{\mathbb{S}}(t)$ is also Hermitian.

We seek a set $\{\bar{W}_n(\boldsymbol{x},t)\}$ spanned by the original set that are orthogonal,

$$\bar{W}_n(\boldsymbol{x},t) = \sum_p W_p'(\boldsymbol{x},t)C_{pn}(t), \tag{B1}$$

where

$$\int \bar{W}_n^*(\boldsymbol{x},t)\bar{W}_m(\boldsymbol{x},t)d\boldsymbol{x} = \sum_{p,l} C^*_{np}(t)S_{nl}(t)C_{lm}(t) = \delta_{nm},$$

or in matrix notation

$$\mathbb{C}^\dagger(t)\mathbb{S}(t)\mathbb{C}(t) = \mathbb{I}.$$

There are of course many matrices $\mathbb{C}(t)$ that can be found that satisfy this condition. However, the desired matrix $\mathbb{C}(t)$ yielding the minimization of (26) is the Hermitian matrix satisfying

$$\mathbb{C}(t) = \mathbb{S}^{-1/2}(t),$$

[23]. That is, it is the "Hermitian square root" of the inverse of the overlap matrix $\mathbb{S}(t)$.

To see the structure of $\mathbb{C}(t)$, and the nature of the resulting $\{\bar{W}_n(\boldsymbol{x},t)\}$, first introduce $\mathbb{T}(t)$ as the inverse of $\mathbb{S}(t)$,

$$\sum_l S_{nl}(t)T_{lm}(t) = \delta_{nm}$$

or

$$\sum_l \hat{S}_{nl}(t)e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_l;t)}T_{lm}(t) = \delta_{nm}.$$

Introducing $\hat{\mathbb{T}}(t)$ according to

$$\hat{T}_{lm}(t) = T_{lm}(t)e^{-i\Phi(\boldsymbol{R}_l,\boldsymbol{R}_m;t)},$$

we have

$$e^{-i\Phi(\boldsymbol{R}_m,\boldsymbol{R}_n;t)}\sum_l \hat{S}_{nl}(t)\hat{T}_{lm}(t)e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{R}_l,\boldsymbol{R}_m;t)} = \delta_{nm}$$

or

$$\sum_l \hat{S}_{nl}(t)\hat{T}_{lm}(t)e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{R}_l,\boldsymbol{R}_m;t)} = \delta_{nm},$$

and we see that the elements of $\hat{\mathbb{T}}(t)$ must be gauge-invariant, since everything else in the equation is. Since $\mathbb{C}(t)$ is the square root of $\mathbb{T}(t)$, we have

$$\sum_l C_{nl}(t)C_{lm}(t) = T_{nm}(t) = \hat{T}_{nm}(t)e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)}. \tag{B2}$$

Now the $\mathbb{C}(t)$ we seek is Hermitian, requiring

$$C^*_{nl}(t) = C_{ln}(t). \tag{B3}$$

Introducing $\hat{\mathbb{C}}(t)$ according to

$$\hat{C}_{nl}(t) = C_{nl}(t)e^{-i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_l;t)}, \tag{B4}$$

then using (B3) we see that

$$\hat{C}^*_{nl}(t) = C^*_{nl}(t)e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_l;t)} = C_{ln}(t)e^{-i\Phi(\boldsymbol{R}_l,\boldsymbol{R}_n;t)} = \hat{C}_{ln}(t),$$

and so the $\hat{\mathbb{C}}(t)$ we seek is Hermitian and, from (B2), satisfies

$$\sum_l \hat{C}_{nl}(t)\hat{C}_{lm}(t)e^{i[\Phi(\boldsymbol{R}_n,\boldsymbol{R}_l;t)+\Phi(\boldsymbol{R}_l,\boldsymbol{R}_m;t)]} = \hat{T}_{nm}(t)e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)} \tag{B5}$$

or

$$\sum_l \hat{C}_{nl}(t)\hat{C}_{lm}(t)e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{R}_l,\boldsymbol{R}_m;t)} = \hat{T}_{nm}(t),$$

a gauge-invariant equation, and so the matrix $\hat{\mathbb{C}}(t)$ we seek is gauge invariant.

In terms of our new quantities we can write (B1) as

$$\begin{aligned}\bar{W}_n(\boldsymbol{x},t) &= \sum_p W_p(\boldsymbol{x})e^{i\Phi(\boldsymbol{x},\boldsymbol{R}_p;t)}\hat{C}_{pn}(t)e^{i\Phi(\boldsymbol{R}_p,\boldsymbol{R}_n;t)}\\ &= e^{i\Phi(\boldsymbol{x},\boldsymbol{R}_n;t)}\sum_p W_p(\boldsymbol{x})\hat{C}_{pn}(t)e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{x};t)}e^{i\Phi(\boldsymbol{x},\boldsymbol{R}_p;t)}e^{i\Phi(\boldsymbol{R}_p,\boldsymbol{R}_n;t)}\\ &= e^{i\Phi(\boldsymbol{x},\boldsymbol{R}_n;t)}\chi_n(\boldsymbol{x},t),\end{aligned}$$

where

$$\chi_n(\boldsymbol{x},t) \equiv \sum_p W_p(\boldsymbol{x})\hat{C}_{pn}(t)e^{i\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_n;t)} \tag{B6}$$

is clearly gauge-invariant, thus establishing (27).

We can easily work out an expansion for the $\bar{W}_n(\boldsymbol{x},t)$ where the overlap between nonidentical $W_n'(\boldsymbol{x},t)$ is small. Defining a matrix $\mathfrak{s}$,

$$\mathfrak{s}(t) \equiv \mathbb{S}(t) - \mathbb{I},$$

where $\mathbb{I}$ is the identity matrix, we have

$$\begin{aligned}\mathbb{S}^{-1/2}(t) &= (\mathbb{I}+\mathfrak{s}(t))^{-1/2}\\ &= \mathbb{I} - \tfrac{1}{2}\mathfrak{s}(t) + \tfrac{3}{8}\mathfrak{s}^2(t) + \dots\end{aligned} \tag{B7}$$

where since $\mathbb{S}$ is Hermitian $\mathfrak{s}$ will be as well. In terms of components, we have

$$\begin{aligned}\mathfrak{s}_{nm}(t) &= S_{nm}(t) - \delta_{nm}\\ &= e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m;t)}\left[\left(\int W_n^*(\boldsymbol{x})e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{x},\boldsymbol{R}_m;t)}W_m(\boldsymbol{x})d\boldsymbol{x}\right) - \delta_{nm}\right]\\ &= e^{i\Phi(\boldsymbol{R}_n,\boldsymbol{R}_m)}\hat{\mathfrak{s}}_{nm}(t),\end{aligned}$$

where

$$\hat{\mathfrak{s}}_{nm}(t) = \left(\int W_n^*(\boldsymbol{x})e^{i\Delta(\boldsymbol{R}_n,\boldsymbol{x},\boldsymbol{R}_m;t)}W_m(\boldsymbol{x})d\boldsymbol{x}\right) - \delta_{nm}$$

is gauge invariant. Clearly the power series expansion (B7) yields the Hermitian square root of $\mathbb{S}$, and so we have

$$\begin{aligned}C_{pn}(t) &= \delta_{pn} - \frac{1}{2}\mathfrak{s}_{pn}(t) + \frac{3}{8}\sum_u \mathfrak{s}_{pu}(t)\mathfrak{s}_{un}(t) + \dots\\ &= \delta_{pn} - \frac{1}{2}\hat{\mathfrak{s}}_{pn}(t)e^{i\Phi(\boldsymbol{R}_p,\boldsymbol{R}_n;t)}\\ &\quad + \frac{3}{8}\sum_u \hat{\mathfrak{s}}_{pu}(t)\hat{\mathfrak{s}}_{un}(t)e^{i[\Phi(\boldsymbol{R}_p,\boldsymbol{R}_u;t)+\Phi(\boldsymbol{R}_u,\boldsymbol{R}_n;t)]} + \dots\end{aligned}$$

Then from (B4) we have

$$\begin{aligned}\hat{C}_{pn}(t) &= \delta_{pn} - \frac{1}{2}\hat{\mathfrak{s}}_{pn}(t)\\ &\quad + \frac{3}{8}\sum_u \hat{\mathfrak{s}}_{pu}(t)\hat{\mathfrak{s}}_{un}(t)e^{i\Delta(\boldsymbol{R}_p,\boldsymbol{R}_u,\boldsymbol{R}_n;t)} + \dots\end{aligned}$$

which is indeed gauge-invariant. Using this in (B6) yields

$$\begin{aligned}\chi_n(\boldsymbol{x},t) &= W_n(\boldsymbol{x}) - \frac{1}{2}\sum_p W_p(\boldsymbol{x})\hat{\mathfrak{s}}_{pn}(t)e^{i\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_n;t)}\\ &\quad + \frac{3}{8}\sum_{p,u} W_p(\boldsymbol{x})\hat{\mathfrak{s}}_{pu}(t)\hat{\mathfrak{s}}_{un}(t)e^{i\Delta(\boldsymbol{R}_p,\boldsymbol{R}_u,\boldsymbol{R}_n;t)}\\ &\quad \times e^{i\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_n;t)} + \dots\end{aligned}$$

Now defining as usual

$$\begin{aligned}&\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_u,\boldsymbol{R}_n;t)\\ &\quad \equiv \Phi(\boldsymbol{R}_n,\boldsymbol{x};t) + \Phi(\boldsymbol{x},\boldsymbol{R}_p;t) + \Phi(\boldsymbol{R}_p,\boldsymbol{R}_u;t) + \Phi(\boldsymbol{R}_u,\boldsymbol{R}_n;t)\\ &\quad = \Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_n;t) + \Delta(\boldsymbol{R}_p,\boldsymbol{R}_u,\boldsymbol{R}_n;t),\end{aligned}$$

we have

$$\begin{aligned}\chi_n(\boldsymbol{x},t) &= W_n(\boldsymbol{x}) - \frac{1}{2}\sum_p W_p(\boldsymbol{x})\hat{\mathfrak{s}}_{pn}(t)e^{i\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_n;t)}\\ &\quad + \frac{3}{8}\sum_{p,u} W_p(\boldsymbol{x})\hat{\mathfrak{s}}_{pu}(t)\hat{\mathfrak{s}}_{un}(t)e^{i\Delta(\boldsymbol{x},\boldsymbol{R}_p,\boldsymbol{R}_u,\boldsymbol{R}_n;t)} + \dots,\end{aligned}$$

and reverting to the original notation of the text this gives (28) as the lowest correction in the magnetic field.

APPENDIX C: THE TREATMENT OF AN ISOLATED ATOM

Here we review the treatment of the response of an isolated atom to an electromagnetic field, neglecting interactions between the electrons, following the spirit of the earlier work by Healy [21]. For simplicity, we treat the nucleus as fixed. Introducing an electron field operator $\psi(\boldsymbol{x},t)$ and beginning with minimal coupling, the electronic charge and current density operators are given by

$$\hat{\rho}(\boldsymbol{x},t) = e\psi^\dagger(\boldsymbol{x},t)\psi(\boldsymbol{x},t), \tag{C1}$$

$$\begin{aligned}\hat{\boldsymbol{j}}(\boldsymbol{x},t) &= \tfrac{1}{2}\psi^\dagger(\boldsymbol{x},t)\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))\psi(\boldsymbol{x},t)\big]\\ &\quad + \tfrac{1}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))\psi(\boldsymbol{x},t)\big]^\dagger\psi(\boldsymbol{x},t),\end{aligned} \tag{C2}$$

where, as in the main text, the function $\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))$ follows from $H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))$ in the usual fashion, $\mathfrak{p}_{mc}(\boldsymbol{x},t)$ is given by (8), and the electron field operator $\psi(\boldsymbol{x},t)$ satisfies the dynamical equation

$$\begin{aligned}i\hbar\frac{\partial\psi(\boldsymbol{x},t)}{\partial t} &= \big[\psi(\boldsymbol{x},t),\mathsf{H}_{mc}(t)\big]\\ &= \big[H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + e\phi(\boldsymbol{x},t)\big]\psi(\boldsymbol{x},t),\end{aligned}$$

where

$$\mathsf{H}_{mc}(t) = \int \psi^\dagger(\boldsymbol{x},t)\big(H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + e\phi(\boldsymbol{x},t)\big)\psi(\boldsymbol{x},t)d\boldsymbol{x}.$$

Assuming the electrons involved remain in a region of space about the nucleus, which we take to be at $\boldsymbol{R}$, we introduce a new field operator

$$\psi_{sp}(\boldsymbol{x},t) = e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi(\boldsymbol{x},t),$$

where $\Phi(\boldsymbol{x},\boldsymbol{R};t)$ is as given (15) in the main text. Now

$$\begin{aligned}i\hbar\frac{\partial\psi_{sp}(\boldsymbol{x},t)}{\partial t} &= \hbar\frac{\partial\Phi(\boldsymbol{x},\boldsymbol{R};t)}{\partial t}\psi_{sp}(\boldsymbol{x},t)\\ &\quad + e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\left(i\hbar\frac{\partial\psi(\boldsymbol{x},t)}{\partial t}\right).\end{aligned}$$

Since

$$\begin{aligned}&\hbar\frac{\partial\Phi(\boldsymbol{x},\boldsymbol{R};t)}{\partial t}\\ &\quad = \frac{e}{c}\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})\frac{\partial A^i(\boldsymbol{w},t)}{\partial t}d\boldsymbol{w}\\ &\quad = -e\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})E^i(\boldsymbol{w},t)d\boldsymbol{w}\\ &\qquad - e\int s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})\frac{\partial\phi(\boldsymbol{w},t)}{\partial w^i}d\boldsymbol{w}\end{aligned}$$

$$= -e\Omega^0_{\boldsymbol{R}}(\boldsymbol{x},t) + e\int \frac{\partial s^i(\boldsymbol{w};\boldsymbol{x},\boldsymbol{R})}{\partial w^i}\phi(\boldsymbol{w},t)d\boldsymbol{w}$$

$$= -e\Omega^0_{\boldsymbol{R}}(\boldsymbol{x},t) - e\phi(\boldsymbol{x},t) + e\phi(\boldsymbol{R},t),$$

where in the second to the last line we have used the definition (20) in the last line we have used the first of (13), we can write

$$e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\left(i\hbar\frac{\partial\psi(\boldsymbol{x},t)}{\partial t}\right)$$

$$= e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\big[H_0(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t)) + e\phi(\boldsymbol{x},t)\big]\psi(\boldsymbol{x},t)$$

$$= \big[H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t)) + e\phi(\boldsymbol{x},t)\big]\psi_{sp}(\boldsymbol{x},t),$$

[recall (19)] and we have

$$i\hbar\frac{\partial\psi_{sp}(\boldsymbol{x},t)}{\partial t} = \big(H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t)) - e\Omega^0_{\boldsymbol{R}}(\boldsymbol{x},t) + e\phi(\boldsymbol{R},t)\big)\psi_{sp}(\boldsymbol{x},t),$$

where note the term $e\phi(\boldsymbol{R},t)$ depends only on time and therefore will contribute only a global phase to $\psi_{sp}(\boldsymbol{x},t)$; it will not contribute to any operator values and thus can be dropped; we take

$$i\hbar\frac{\partial\psi_{sp}(\boldsymbol{x},t)}{\partial t} = \big(H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t)) - e\Omega^0_{\boldsymbol{R}}(\boldsymbol{x},t)\big)\psi_{sp}(\boldsymbol{x},t),$$

and then have

$$i\hbar\frac{\partial\psi_{sp}(\boldsymbol{x},t)}{\partial t} = \big[\psi_{sp}(\boldsymbol{x},t),\mathsf{H}_{sp}(t)\big], \tag{C3}$$

where

$$\mathsf{H}_{sp}(t) = \int \psi^\dagger_{sp}(\boldsymbol{x},t)\big(H_0(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t)) - e\Omega^0_{\boldsymbol{R}}(\boldsymbol{x},t)\big)\psi_{sp}(\boldsymbol{x},t)d\boldsymbol{x}. \tag{C4}$$

Looking at the electronic charge and current densities, we can write the first (C1) as

$$\hat{\rho}(\boldsymbol{x},t) = e\psi^\dagger_{sp}(\boldsymbol{x},t)\psi_{sp}(\boldsymbol{x},t), \tag{C5}$$

while the second becomes

$$\hat{\boldsymbol{j}}(\boldsymbol{x},t) = \frac{1}{2}\psi^\dagger_{sp}(\boldsymbol{x},t)e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi_{sp}(\boldsymbol{x},t)\big] + \frac{1}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}_{mc}(\boldsymbol{x},t))e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi_{sp}(\boldsymbol{x},t)\big]^\dagger e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\psi_{sp}(\boldsymbol{x},t)$$

or

$$\hat{\boldsymbol{j}}(\boldsymbol{x},t) = \frac{1}{2}\psi^\dagger_{sp}(\boldsymbol{x},t)\big[\boldsymbol{J}\big(\boldsymbol{x},e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\mathfrak{p}_{mc}(\boldsymbol{x},t)e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\big)\psi_{sp}(\boldsymbol{x},t)\big] + \frac{1}{2}\big[\boldsymbol{J}\big(\boldsymbol{x},e^{-i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\mathfrak{p}_{mc}(\boldsymbol{x},t)e^{i\Phi(\boldsymbol{x},\boldsymbol{R};t)}\big)\psi_{sp}(\boldsymbol{x},t)\big]^\dagger\psi_{sp}(\boldsymbol{x},t),$$

which can be written as

$$\hat{\boldsymbol{j}}(\boldsymbol{x},t) = \tfrac{1}{2}\psi^\dagger_{sp}(\boldsymbol{x},t)\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))\psi_{sp}(\boldsymbol{x},t)\big] + \tfrac{1}{2}\big[\boldsymbol{J}(\boldsymbol{x},\mathfrak{p}(\boldsymbol{x},\boldsymbol{R};t))\psi_{sp}(\boldsymbol{x},t)\big]^\dagger\psi_{sp}(\boldsymbol{x},t). \tag{C6}$$

For an isolated atom, we can define polarization and magnetization *operators* as

$$\begin{aligned}\hat{\boldsymbol{p}}(\boldsymbol{x},t) &\equiv \int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{w},\boldsymbol{R})\hat{\rho}(\boldsymbol{w},t)d\boldsymbol{w},\\ \hat{m}^j(\boldsymbol{x},t) &\equiv \frac{1}{c}\int \alpha^{jk}(\boldsymbol{x};\boldsymbol{w},\boldsymbol{R})\hat{j}^k(\boldsymbol{w},t)d\boldsymbol{w}.\end{aligned} \tag{C7}$$

Then from the properties of the relators, we find immediately that

$$\begin{aligned}\hat{\rho}(\boldsymbol{x},t) &= -\nabla\cdot\hat{\boldsymbol{p}}(\boldsymbol{x},t) + Q\delta(\boldsymbol{x}-\boldsymbol{R}),\\ \hat{\boldsymbol{j}}(\boldsymbol{x},t) &= \frac{\partial\hat{\boldsymbol{p}}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\hat{\boldsymbol{m}}(\boldsymbol{x},t) - \int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\hat{K}(\boldsymbol{y},t)d\boldsymbol{y},\end{aligned} \tag{C8}$$

where

$$Q = \int \hat{\rho}(\boldsymbol{x},t)d\boldsymbol{x},$$

$$\hat{K}(\boldsymbol{x},t) = \frac{\partial\hat{\rho}(\boldsymbol{x},t)}{\partial t} + \nabla\cdot\hat{\boldsymbol{j}}(\boldsymbol{x},t).$$

In arriving at (C8), we have only used (13) and the fact that the charge-current operators are only nonzero in a confined region of space near $\boldsymbol{R}$. We write the quantity Q without an operator hat because it is a conserved quantity and can be taken as a number; it is the total electron charge. Note however that local charge conservation (or the ansatz that it holds if we make various approximations in our equations) leads to $\hat{K}(\boldsymbol{x},t) = 0$. Hence we can write (C8) as

$$\begin{aligned}\hat{\rho}(\boldsymbol{x},t) &= -\nabla\cdot\hat{\boldsymbol{p}}(\boldsymbol{x},t) + Q\delta(\boldsymbol{x}-\boldsymbol{R}),\\ \hat{\boldsymbol{j}}(\boldsymbol{x},t) &= \frac{\partial\hat{\boldsymbol{p}}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\hat{\boldsymbol{m}}(\boldsymbol{x},t),\end{aligned} \tag{C9}$$

the standard form. There is a great advantage of determining $\hat{\rho}(\boldsymbol{x},t)$ and $\hat{\boldsymbol{j}}(\boldsymbol{x},t)$ (or their expectation values) by first determining $\hat{\boldsymbol{p}}(\boldsymbol{x},t)$ and $\hat{\boldsymbol{m}}(\boldsymbol{x},t)$ (or their expectation values). For if the former are found from the latter, regardless of how many approximations are involved in determining the latter we will still automatically have charge conservation.

For comparison with the next section of the appendices, we here define

$$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t) \equiv \int \boldsymbol{s}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\langle\hat{\rho}(\boldsymbol{y},t)\rangle d\boldsymbol{y},$$

$$m^j_{\boldsymbol{R}}(\boldsymbol{x},t) \equiv \frac{1}{c}\int \alpha^{jk}(\boldsymbol{x};\boldsymbol{y},\boldsymbol{R})\langle\hat{j}^k(\boldsymbol{y},t)\rangle d\boldsymbol{y},$$

and then from (C9) we can write

$$\begin{aligned}\langle\hat{\rho}(\boldsymbol{x},t)\rangle &= -\nabla\cdot\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t) + Q\delta(\boldsymbol{x}-\boldsymbol{R}),\\ \langle\hat{\boldsymbol{j}}(\boldsymbol{x},t)\rangle &= \frac{\partial\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x},t)}{\partial t} + c\nabla\times\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x},t).\end{aligned} \tag{C10}$$

Introducing a set of basis function $\{W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\}$, where α varies but $\boldsymbol{R}$ is fixed,

$$\psi_{sp}(\boldsymbol{x},t) = \sum_\alpha c_\alpha(t)W_{\alpha\boldsymbol{R}}(\boldsymbol{x}),$$

where

$$\{c_\alpha(t), c_\beta(t)\} = 0,$$
$$\{c_\alpha(t), c_\beta^\dagger(t)\} = \delta_{\alpha\beta},$$

we can write

$$\rho_{\boldsymbol{R}}(\boldsymbol{x}, t) = \sum_{\alpha,\beta} \rho_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R})\eta_{\alpha\beta}(t),$$
$$\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x}, t) = \sum_{\alpha,\beta} \boldsymbol{j}_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}; t)\eta_{\alpha\beta}(t), \tag{C11}$$

with $\rho_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R})$ and $\boldsymbol{j}_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}; t)$ given by (D4), and

$$\eta_{\alpha\beta}(t) \equiv \langle c_\beta^\dagger(t)c_\alpha(t)\rangle.$$

From the evolution Eq. (C3), we find that the dynamical of $\eta_{\alpha\beta}(t)$ are given by

$$i\hbar\frac{\partial \eta_{\alpha\beta}(t)}{\partial t} = \sum_{\lambda}\left(\bar{H}_{\alpha\lambda}(\boldsymbol{R}; t)\eta_{\lambda\beta}(t) - \eta_{\alpha\lambda}(t)\bar{H}_{\lambda\beta}(\boldsymbol{R}; t)\right), \tag{C12}$$

where

$$\bar{H}_{\alpha\lambda}(\boldsymbol{R}; t) = \frac{1}{2}\int W_{\alpha\boldsymbol{R}}^*(\boldsymbol{x})H_0(\boldsymbol{x}, \mathfrak{p}(\boldsymbol{x}, \boldsymbol{R}; t))W_{\lambda\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x}$$
$$+ \frac{1}{2}\int \left(H_0(\boldsymbol{x}, \mathfrak{p}(\boldsymbol{x}, \boldsymbol{R}; t))W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\right)^* W_{\lambda\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x}$$
$$- e\int W_{\alpha\boldsymbol{R}}^*(\boldsymbol{x})\Omega_{\boldsymbol{R}}^0(\boldsymbol{x}, t)W_{\lambda\boldsymbol{R}}(\boldsymbol{x})d\boldsymbol{x}. \tag{C13}$$

APPENDIX D: THE “ISOLATED ATOM LIMIT” OF A CRYSTAL

The isolated atom limit of our equations in Sec. II is identified by the assumption that $W_{\alpha\boldsymbol{R}}(\boldsymbol{x})$ and $W_{\beta\boldsymbol{R}'}(\boldsymbol{x})$ have no common support if $\boldsymbol{R} \neq \boldsymbol{R}'$. In this limit it follows from the definition (24) that the set of functions $\{W'_{\alpha\boldsymbol{R}}(\boldsymbol{x}, t)\}$ are mutually orthogonal, so $\bar{W}_{\alpha\boldsymbol{R}}(\boldsymbol{x}, t) \to W'_{\alpha\boldsymbol{R}}(\boldsymbol{x}, t)$ and, from (24,27), we see $\chi_{\alpha\boldsymbol{R}}(\boldsymbol{x}, t) \to W_{\alpha\boldsymbol{R}}(\boldsymbol{x})$. Then from (35), we have

$$\bar{H}_{\alpha\boldsymbol{R};\lambda\boldsymbol{R}''}(t) \to \delta_{\boldsymbol{R}\boldsymbol{R}''}\bar{H}_{\alpha\lambda}(\boldsymbol{R}; t), \tag{D1}$$

with $\bar{H}_{\alpha\lambda}(\boldsymbol{R}; t)$ is given by (C13). Assuming no initial correlation between the electronic motion in the individual atoms, from (33) and (36), we have

$$\eta_{\alpha\boldsymbol{R};\beta\boldsymbol{R}'}(t) \to \delta_{\boldsymbol{R}\boldsymbol{R}'}\eta_{\alpha\beta}(\boldsymbol{R}; t) \tag{D2}$$

at least initially, and this condition will then be maintained as the dynamics evolve according to (37), which reduces to

$$i\hbar\frac{\partial \eta_{\alpha\beta}(\boldsymbol{R}; t)}{\partial t} = \sum_{\lambda}\left(\bar{H}_{\alpha\lambda}(\boldsymbol{R}; t)\eta_{\lambda\beta}(\boldsymbol{R}; t) - \eta_{\alpha\lambda}(\boldsymbol{R}; t)\bar{H}_{\lambda\beta}(\boldsymbol{R}; t)\right). \tag{D3}$$

for each $\boldsymbol{R}$. Comparing with (C12) we see that this is indeed the dynamics expected for a collection of isolated atoms. Finally, (39) becomes

$$G_{\boldsymbol{R}}(\boldsymbol{x}, \boldsymbol{y}; t) = i\sum_{\alpha,\beta} \eta_{\alpha\beta}(\boldsymbol{R}; t)W_{\beta\boldsymbol{R}}^*(\boldsymbol{y})W_{\alpha\boldsymbol{R}}(\boldsymbol{x}),$$

and for use in (44) we can take $\rho_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x}, \boldsymbol{R}; t) \to \rho_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R})$ and $\boldsymbol{j}_{\beta\boldsymbol{R}';\alpha\boldsymbol{R}''}(\boldsymbol{x}, \boldsymbol{R}; t) \to \boldsymbol{j}_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}; t)$, where from (45) and (46), we obtain

$$\rho_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}) = eW_{\beta\boldsymbol{R}}^*(\boldsymbol{x})W_{\alpha\boldsymbol{R}}(\boldsymbol{x}),$$
$$\boldsymbol{j}_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}; t) = \frac{1}{2}W_{\beta\boldsymbol{R}}^*(\boldsymbol{x})\left(\boldsymbol{J}(\boldsymbol{x}, \mathfrak{p}(\boldsymbol{x}, \boldsymbol{R}; t))W_{\alpha\boldsymbol{R}}(\boldsymbol{x})\right)$$
$$+ \frac{1}{2}\left(\boldsymbol{J}^*(\boldsymbol{x}, \mathfrak{p}(\boldsymbol{x}, \boldsymbol{R}; t))W_{\beta\boldsymbol{R}}^*(\boldsymbol{x})\right)W_{\alpha\boldsymbol{R}}(\boldsymbol{x}), \tag{D4}$$

so (44) become

$$\rho_{\boldsymbol{R}}(\boldsymbol{x}, t) \to \sum_{\alpha,\beta} \rho_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R})\eta_{\alpha\beta}(\boldsymbol{R}; t) = -ie\left[G_{\boldsymbol{R}}(\boldsymbol{x}, \boldsymbol{y}; t)\right]_{\boldsymbol{y}\to\boldsymbol{x}},$$
$$\boldsymbol{j}_{\boldsymbol{R}}(\boldsymbol{x}, t) \to \sum_{\alpha,\beta} \boldsymbol{j}_{\beta\alpha}(\boldsymbol{x}, \boldsymbol{R}; t)\eta_{\alpha\beta}(\boldsymbol{R}; t)$$
$$= -ie\left[\boldsymbol{\mathcal{J}}_{\boldsymbol{R}}(\boldsymbol{x}, \boldsymbol{y}; t)G_{\boldsymbol{R}}(\boldsymbol{x}, \boldsymbol{y}; t)\right]_{\boldsymbol{y}\to\boldsymbol{x}}. \tag{D5}$$

From (D1) and (D2), it follows from (50) that $I(\boldsymbol{R}, \boldsymbol{R}'; t) = 0$, and from (49) each site charge [see (48)] is independent of time,

$$Q_{\boldsymbol{R}} = \int \rho_{\boldsymbol{R}}(\boldsymbol{x}, t)d\boldsymbol{x} = e\sum_{\alpha} \eta_{\alpha\alpha}(\boldsymbol{R}; t),$$

and from (52) and (53), we have

$$\rho_F(\boldsymbol{x}) = \sum_{\boldsymbol{R}} Q_{\boldsymbol{R}}\delta(\boldsymbol{x} - \boldsymbol{R}), \quad \boldsymbol{j}_F(\boldsymbol{x}, t) = 0, \tag{D6}$$

where the first is independent of time. Now since the Wannier functions associated with different sites are assumed to have no common support, from charge conservation (47), we must have $K_{\boldsymbol{R}}(\boldsymbol{x}, t) = 0$ for all $\boldsymbol{R}$, since at any given $\boldsymbol{x}$ at most one $K_{\boldsymbol{R}}(\boldsymbol{x}, t)$ can contribute to the sum (47); together with the second of (D6), this guarantees that $\tilde{\boldsymbol{j}}(\boldsymbol{x}, t) = 0$ [see (59)]. With that, and the use of (D5), our general expressions (73) and (74) reduce to

$$\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x}, t) = \sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\left[\int \boldsymbol{s}(\boldsymbol{x}; \boldsymbol{y}, \boldsymbol{R})\rho_{\beta\alpha}(\boldsymbol{y}, \boldsymbol{R})d\boldsymbol{y}\right]\eta_{\alpha\beta}(\boldsymbol{R}; t) \tag{D7}$$

and

$$m_{\boldsymbol{R}}^j(\boldsymbol{x}, t) = \frac{1}{c}\sum_{\alpha,\beta,\boldsymbol{R}',\boldsymbol{R}''}\left[\int \alpha^{jk}(\boldsymbol{x}; \boldsymbol{y}, \boldsymbol{R})j_{\beta\alpha}^k(\boldsymbol{y}, \boldsymbol{R}; t)d\boldsymbol{y}\right]\eta_{\alpha\beta}(\boldsymbol{R}; t). \tag{D8}$$

With the aid of the first of (57) and (67),

$$\boldsymbol{p}(\boldsymbol{x}, t) = \sum_{\boldsymbol{R}} \boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x}, t),$$
$$\boldsymbol{m}(\boldsymbol{x}, t) = \sum_{\boldsymbol{R}} \boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x}, t),$$

so from the first of (58) and (68), together with (D6), we can write

$$\langle\hat{\rho}(\boldsymbol{x}, t)\rangle = \sum_{\boldsymbol{R}}\left(-\nabla\cdot\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x}, t) + Q_{\boldsymbol{R}}\delta(\boldsymbol{x} - \boldsymbol{R})\right),$$
$$\langle\hat{\boldsymbol{j}}(\boldsymbol{x}, t)\rangle = \sum_{\boldsymbol{R}}\left(\frac{\partial \boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x}, t)}{\partial t} + \nabla\times\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x}, t)\right), \tag{D9}$$

with of course the dynamics of $\boldsymbol{p}_{\boldsymbol{R}}(\boldsymbol{x}, t)$ and $\boldsymbol{m}_{\boldsymbol{R}}(\boldsymbol{x}, t)$ given by the use of the solution of (D3) in (D7) and (D8). Comparing (C10) with (D9) we see that the latter is indeed what we would expect for the charge-current density of a collection of isolated atoms.

Naturally, in a calculation with a finite set of Wannier functions one cannot expect $K_{\boldsymbol{R}}(\boldsymbol{x}, t) = 0$, or even the sum to be zero. But this situation arose as well for an isolated atom in Appendix C. There, and indeed as was done in the general derivation in Sec. II, the approach is to envision a calculation with an infinite set of basis functions, and construct equations for the charge and current densities in terms of the polarization and magnetization fields. By their very structure the equations guarantee charge conservation, even if the set of basis functions is truncated and the resulting expressions for the polarization and magnetization fields are only approximate.

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