Microscopic polarization and magnetization fields in extended systems

Perry T. Mahon,1,∗ Rodrigo A. Muniz,1,2, † and J. E. Sipe1, ‡

1Department of Physics, University of Toronto, Toronto, Ontario M5S 1A7, Canada
2Department of Electrical Engineering and Computer Science, University of Michigan, Ann Arbor, MI 48109, USA
(Dated: November 13, 2018)

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. 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.

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, \( P(x, t) \) and \( M(x, t) \), which appear in the description of the macroscopic charge and current densities,

\[
\begin{align*}
\varrho(x, t) &= -\nabla \cdot P(x, t) + \varrho_F(x, t), \\
J(x, t) &= \frac{\partial P(x, t)}{\partial t} + \rho \nabla \times M(x, t) + J_F(x, t),
\end{align*}
\]

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, \( \varrho_F(x, t) \) and \( J_F(x, t) \), 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 \( P(x, t) \) and \( M(x, t) \) to include contributions from higher multipole moments of constituent molecules [2, 3]. The work of Power, Zienau, Wooley [4], 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: 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. But 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].

A different strategy is that of the “modern theory of polarization”, [11, 12] which has focussed on gapped systems where physical intuition suggests that to lowest order none of the response to slowly varying fields should

∗ pmahon@physics.utoronto.ca
† rodrigo.a.muniz@gmail.com
‡ sipe@physics.utoronto.ca
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 \( \mathbf{J} \) via the equation \( d\mathbf{J}/dt \equiv \mathbf{J} \). Since \( \mathbf{J} \) can be calculated for an adiabatically applied uniform electric field, at least the change in \( \mathbf{P} \) can be identified. Magnetizations, \( \mathbf{M} \), associated with unperturbed systems can also be identified [13, 14], 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 magnetic field over the unit cell on the optical response, and the polarization and magnetization fields that are associated with the induced current density, \( \mathbf{J} \), can be identified locally [22]. Rather, we construct expressions for \( \mathbf{P}(x, t) \) and \( \mathbf{M}(x, t) \) in terms of electron Green functions and their expansions in terms of localized basis functions associated with each site. 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 the process of constructing \( \mathbf{P}(x, t) \) and \( \mathbf{M}(x, t) \) we can confirm that they are indeed related to the microscopic charge and current densities near \( \mathbf{R} \). 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 other than those that could be included in a mean field treatment of the electromagnetic field. Hence it is
sufficient to consider only the lesser, equal time 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 nuclei 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. 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].

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

II. MICROSCOPIC POLARIZATION AND MAGNETIZATION FIELDS

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

$$G_{mc}(x, y; t) = i \langle \psi^\dagger(y, t) \psi(x, t) \rangle,$$  \hspace{1cm} (4)

where the subscript $mc$ denotes the usual minimal coupling procedure to include the effect of an external, classical electromagnetic field specified by a scalar potential $\phi(x, t)$ and a vector potential $A(x, t)$; 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

$$i\hbar \frac{\partial G_{mc}(x, y; t)}{\partial t} = K_{mc}(x, y; t)G_{mc}(x, y; t),$$  \hspace{1cm} (5)

where

$$K_{mc}(x, y; t) = H_{mc}(x, t) - H^*_mc(y, t),$$  \hspace{1cm} (6)

with the “script” fonts (such as $K_{mc}$ and $H_{mc}$) denoting differential operators acting on the functions that follow, at and in the neighborhood of the spatial variable(s) indicated. We have

$$H_{mc}(x, t) = H_0(x, p_{mc}(x, t)) + e\phi(x, t),$$  \hspace{1cm} (7)

where

$$p_{mc}(x, t) = p(x) - \frac{e}{c}A(x, t),$$  \hspace{1cm} (8)

with

$$p(x) = \frac{\hbar}{i} \frac{\partial}{\partial x} - \frac{e}{c}A_{static}(x).$$  \hspace{1cm} (9)

In (9) we have included the possibility of the presence of a static, periodic magnetic field described by a vector potential $A_{static}(x)$, where $A_{static}(x) = A_{static}(x + R)$ for any lattice site position $R$; note that we distinguish such a static, periodic vector potential from the “external” electromagnetic field associated with $\phi(x, t)$ and $A(x, t)$. 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

$$H_0^{Sch}(x, p(x)) = \frac{\langle p(x) \rangle^2}{2m} + V(x),$$  \hspace{1cm} (10)

but more general forms can be considered. In a system that is periodic before the application of an external electromagnetic field, we have $H_0(x + R, p(x)) = H_0(x, p(x))$, 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 equation (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

\[ H_0(x, p(x)) \rightarrow H_0(x, p_{mc}(x, t)) + e\phi(x, t). \]

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

\[ \langle \hat{\rho}(x, t) \rangle = -ie \left[ G_{mc}(x, y; t) \right]_{y \rightarrow x}, \tag{11} \]
\[ \langle \hat{j}(x, t) \rangle = \left[ J_{mc}(x, y; t) G_{mc}(x, y; t) \right]_{y \rightarrow x}, \]

where

\[ J_{mc}(x, y; t) = -\frac{i}{2} \left( J(x, p_{mc}(x, t)) + J(y, p_{mc}^*(y, t)) \right), \]

and the function \( J(x, p_{mc}(x, t)) \) follows from \( H_0(x, p_{mc}(x, t)) \) in the usual fashion [29]. For the Schrödinger Hamiltonian, for example, we have

\[ J(x, p_{mc}(x, t)) = \frac{e}{m} p_{mc}(x, t). \]

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,

\[ E(x, t) = -\nabla \phi(x, t) - \frac{1}{c} \frac{\partial A(x, t)}{\partial t}, \]
\[ B(x, t) = \nabla \times A(x, t). \]

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 [30, 31] as

\[ s^i(w; x, y) = \int_{C(x, y)} dz^i \delta(w - z), \tag{12} \]
\[ \alpha^{jk}(w; x, y) = e^{jmn} \int_{C(x, y)} dz^m \frac{\partial z^n}{\partial x^j} \delta(w - z), \]
\[ \beta^{jk}(w; x, y) = e^{jmn} \int_{C(x, y)} dz^m \frac{\partial z^n}{\partial y^j} \delta(w - z), \]

where for each two points \( x \) and \( y \) we use \( C(x, y) \) to indicate some path from \( y \) to \( x \). Here and below superscripts indicate Cartesian components, \( e^{jmn} \) is the Levi-Civita symbol, and repeated Cartesian components are summed over [32]. Regardless of the path chosen, the quantities (12) satisfy

\[ \frac{\partial s^i(w; x, y)}{\partial w^j} = -\delta(w - x) + \delta(w - y), \tag{13} \]
\[ \frac{\partial s^i(w; x, y)}{\partial x^j} = \delta^{ik} \delta(w - x) - \epsilon^{ijp} \frac{\partial \alpha^{kp}(w; x, y)}{\partial w^p}, \]
\[ \frac{\partial s^i(w; x, y)}{\partial y^j} = -\delta^{ik} \delta(w - y) - \epsilon^{ijp} \frac{\partial \beta^{kp}(w; x, y)}{\partial w^p}, \]

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

\[ s^i(w; x, y) + s^i(w; y, x) = 0, \tag{14} \]
\[ \alpha^{jk}(w; x, y) + \beta^{jk}(w; y, x) = 0. \]

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

### A. The global Green function

We now use these relators to introduce a new Green function, \( G_{gl}(x, y; t) \) [33]. The subscript \( gl \) denotes “global”, in that no special point is introduced, and yet the dynamical equation for \( G_{gl}(x, y; t) \) and the expressions for the 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 \( C(x, y) \), or in choosing sets of relators satisfying (13) that are even more general than (12). While straight lines between \( y \) and \( x \) are almost always chosen for the paths \( C(x, y) \) 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.

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

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

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

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

As we always use symmetric relators we obtain

\[ G_{gl}^*(x, y; t) = -G_{gl}(y, x; t), \]

where \( G_{gl}(x, y; t) \) is the global Green function.
following from the fact that $G_{mc}(x, y; t)$ trivially satisfies this relation. While each factor on the right-hand-side of (16) is gauge dependent, their product is not. For the dynamical equation for $G_{gl}(x, y; t)$ is found to be
\[
 i\hbar \frac{\partial G_{gl}(x, y; t)}{\partial t} = \mathcal{K}_{gl}(x, y; t)G_{gl}(x, y; t),
\]
(17)
where
\[
 \mathcal{K}_{gl}(x, y; t) = H_0(x, p(x, y; t)) - H_0^*(y, p(y, x; t)) - e\Omega^0_y(x, t),
\]
(18)
with
\[
 p^k(x, y; t) \equiv p^k(x) - \frac{e}{c}\Omega^k_y(x, t),
\]
(19)
and
\[
 \Omega^0_y(x, t) \equiv \int s^l(w; x, y)E^l(w, t)dw,
\]
\[
 \Omega^k_y(x, t) \equiv \int \alpha^l(w; x, y)B^l(w, t)dw.
\]
(20)
Now using (11,16), we can write the charge and current densities as
\[
 \langle \hat{\rho}(t) \rangle = -i\epsilon \left[ G_{gl}(x, y; t) \right]_{y \rightarrow x},
\]
\[
 \langle \hat{\mathbf{j}}(t) \rangle = \left[ \mathcal{J}_{gl}(x, y; t)G_{gl}(x, y; t) \right]_{y \rightarrow x},
\]
(21)
where
\[
 \mathcal{J}_{gl}(x, y; t) = -\frac{i}{2} \left( \mathbf{J}(x, p(x, y; t)) + J^*(y, p(y, x; t)) \right).
\]
Each these quantities is clearly gauge invariant.

**B. Wannier functions and adjusted Wannier functions**

While the global gauge invariant Green function $G_{gl}(x, y; t)$ 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 $\{W_{\alpha R}(x)\}$, or “orbitals”, labeled by the lattice site $R$ and a “type index” $\alpha$. We take the functions in the set $\{W_{\alpha R}(x)\}$ to be orthogonal,
\[
 \int W^*_\beta R'(x) W_{\alpha R}(x) dx = \delta_{\beta \alpha} \delta_{R'R}.
\]
(22)

In the special case of a periodic system, which is our main focus here, we have
\[
 W_{\alpha R}(x) = W_{\alpha}(x - R),
\]
(23)
where we identify $W_{\alpha}(x)$ as a function localized near the origin, and the set $\{W_{\alpha R}(x)\}$ 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 [34] [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, $A(x, t) \rightarrow \tilde{A}(t)$, the set $\{\tilde{W}_{\alpha R}(x, t)\}$ of modified orbitals
\[
 W'_{\alpha R}(x, t) = e^{i\phi(x, R; t)} W_{\alpha R}(x)
\]
(24)
are orthonormal and often useful in calculations. Yet for nonuniform vector potentials the functions in the set $\{W'_{\alpha R}(x)\}$ 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 $\{\tilde{W}_{\alpha R}(x, t)\}$,
\[
 \int \tilde{W}^*_\beta R'(x) \tilde{W}_{\alpha R}(x, t) dx = \delta_{\beta \alpha} \delta_{R'R}.
\]
(25)
which are as close as possible to the functions $\{W'_{\alpha R}(x, t)\}$, in the sense that
\[
 \sum_{\alpha, R} \left| \int \tilde{W}_{\alpha R}(x, t) - W'_{\alpha R}(x, t) \right|^2 dx
\]
(26)
is a minimum at each time $t$. We refer to these new functions as “adjusted Wannier functions”. For a finite system the set $\{\tilde{W}_{\alpha R}(x, t)\}$ can be found numerically if the sum over $\alpha$ is truncated. In any case, it always follows that the $\{\tilde{W}_{\alpha R}(x, t)\}$ are of the form
\[
 \tilde{W}_{\alpha R}(x, t) = e^{i\phi(x, R; t)} \chi_{\alpha R}(x, t),
\]
(27)
where the functions in the set $\{\chi_{\alpha R}(x, t)\}$ 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 $\chi_{\alpha R}(x, t)$; the first two terms are
\[
 \chi_{\alpha R}(x, t) = W_{\alpha R}(x) - \frac{1}{2} i \sum_{\beta, R'} W^*_{\beta R'}(x) \left[ \int W^*_{\beta R'}(y) \Delta(R', y, R; t) W_{\alpha R}(y) dy \right] + ...
\]
(28)
(see Appendix B), where very generally
\[
 \Delta(x, y, z; t) \equiv \Phi(z, x; t) + \Phi(x, y; t) + \Phi(y, z; t)
\]
(29)
is a gauge invariant quantity, since it involves only the flux of the magnetic field through the surface identified by $x, y, z$ and the paths connecting them.
Neglecting the spin degree of freedom, which could be included in a straight-forward way, we expand the field operator,

\[ \psi(x, t) = \sum_{\alpha} a^\alpha_R(t) W^\alpha_{R}(x, t), \tag{30} \]

where

\[ \left\{ a^\alpha_R(t), a^{\beta\prime}_R(t) \right\} = 0, \]

\[ \left\{ a^\alpha_R(t), a^{\beta\prime}_R(t) \right\} = \delta_{\alpha\beta}\delta_{RR'}, \tag{31} \]

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 \( G_{mc}(x, y; t) \) takes the form

\[ G_{mc}(x, y; t) = i \sum_{\alpha,\beta, R, R'} \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t) W^\beta'_{R'}(y, t) W^\alpha_{R}(x, t), \tag{32} \]

where

\[ \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t) = \left\langle a^\dagger_{\beta'; R'}(t) a^\alpha_R(t) \right\rangle. \tag{33} \]

From the dynamical equation (5) for \( G_{mc}(x, y; t) \) we can immediately find the equations of motion for the \( \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t) \),

\[ i\hbar \frac{\partial \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t)}{\partial t} = \sum_{\lambda, R''} \left( e^{\phi(R, R''; t)} \tilde{H}^\lambda_{\alpha; R, \beta; R} \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t) \right. \]

\[ - \bar{\eta}^\lambda_{\alpha; R, \beta; R} e^{\phi(R', R''; t)} \left. \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t) \right) \]

\[ - e^{\Omega^\phi_{R''}(R, t)} \bar{\eta}^\alpha_{\alpha; R, \beta; R'}(t), \tag{34} \]

where

\[ \Omega^\phi_{y}(x, t) \equiv - \int s^i(z; x, y) \frac{\partial \phi(z, t)}{\partial z^i} dz, \]

and the matrix \( \tilde{H} \) is Hermitian, \( \tilde{H}^\star_{\alpha; R, \lambda; R''; t} = \tilde{H}_{\lambda; R, \alpha; R''; t} \), with elements that are gauge invariant.

\[ \tilde{H}^\lambda_{\alpha; R, \beta; R'}(t) = \sum_{\lambda, R''} \left( e^{\phi(R, R''; t)} \right. \]

\[ \sum_{\alpha, \beta, R, R'} \left[ \bar{\eta}^\lambda_{\alpha; R, \beta; R}(t) \frac{\delta}{\delta \chi^\lambda_{\alpha; R, \beta; R'}(x, t)} \right. \]

The equations (34,35) can be understood as the microscopic underpinnings of the equations of a lattice gauge theory. These expressions display five main features: (a) the functions \( \Phi(R, R''; t) \) and \( \Omega^\phi_{R''}(R, t) \) allow for an arbitrary variation of the scalar and vector potential as one moves between neighboring sites; (b) the “hopping” matrix elements \( \tilde{H}^\lambda_{\alpha; R, \beta; R'}(t) \) involve a set of states (labeled by Greek letters) at each site, and thus are matrices even for fixed \( R \) and \( R'' \); (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 charge and current densities can be constructed once the \( \bar{\eta}^\lambda_{\alpha; R, \beta; R'}(t) \) are found and \( G_{mc}(x, y; t) \) 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 \( \tilde{H}^\lambda_{\alpha; R, \beta; R'}(t) \) we will construct the microscopic polarization and magnetization fields associated with each 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

$$\eta_{\alpha R;\beta R'}(t) \equiv \tilde{\eta}_{\alpha R;\beta R'}(t)e^{i\Omega(R';R,t)}, \quad (36)$$

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

$$i\hbar \frac{\partial \eta_{\alpha R;\beta R'}(t)}{\partial t} = \sum_{\lambda, R''} e^{i\Delta(R,R'',R';t)} \left( \tilde{H}_{\alpha R;\lambda R''}(t)\eta_{\lambda R'',\beta R'}(t) \right. - \eta_{\alpha R;\lambda R''}(t)\tilde{H}_{\lambda R'',\beta R'}(t) \left. - e^{i\Omega(R';R,t)}\eta_{\alpha R;\beta R'}(t) \right). \quad (37)$$

Although the form of (37) is a bit unusual because of the factor $\exp(\Omega(R,R',R';t))$, note that all terms appearing in this equation for $\eta_{\alpha R;\beta R'}(t)$ are gauge invariant. Also, since any initial $\eta_{\alpha R;\beta R'}(t)$ before any fields are applied is trivially gauge invariant, then $\eta_{\alpha R;\beta R'}(t)$ itself will be gauge-invariant even as the fields are introduced.

We can write $G_{\beta}(x,y;t)$ in terms of the $\eta_{\alpha R;\beta R'}(t)$ via (16,32,36), and from the orthonormality relations (25) we can write the $\eta_{\alpha R;\beta R'}(t)$ in terms of $G_{\beta}(x,y;t)$; writing these results in terms of the $\chi_{\alpha R}(x,t)$ of (27) we have

$$G_{\beta}(x,y;t) = \int \frac{d^d\Phi}{(2\pi)^d} \frac{1}{2\pi} e^{i\Phi(x,y,t)} \frac{\partial}{\partial \Phi(x,y,t)} \chi_{\alpha R}(x,t) \sum_{\alpha, R; R'} \eta_{\alpha R;\beta R'}(t) \chi_{\alpha R'}(x,t) \quad (38)$$

where in general

$$\Delta(x,u,v,y;t) = \Phi(y,x,t) + \Phi(x,u,t) + \Phi(u,v,t) + \Phi(v,y,t)$$

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

$$G_{\beta}^*(x,y;t) = -G_{\beta}(x,y;t),$$

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

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

and then have

$$G_{\beta}(x,y;t) = \sum_R e^{-i\Delta(x,y,R;R',t)} G_R(x,y;t). \quad (40)$$

The quantities (39) are chosen so that the gauge invariant phase factors $\exp(-i\Delta(x,y,R,R';t))$ appear in the decomposition (40) for reasons that will become clear below [35].

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

$$\begin{align*}
\langle \tilde{\rho}(x,t) \rangle & = \sum_{\alpha} \rho_{\beta}(x,t), \\
\langle \tilde{j}(x,t) \rangle & = \sum_{\alpha} j_{\beta}(x,t),
\end{align*}$$

where

$$\begin{align*}
\rho_{\beta}(x,t) & = -ie \left[ G_R(x,y;t) \right]_{y \to x} \\
j_{\beta}(x,t) & = \left[ J_R(x,y;t) G_R(x,y;t) \right]_{y \to x},
\end{align*}$$

with

$$\begin{align*}
J_R(x,y;t) & = -\frac{i}{2} \left( J(x,p(x,R,t)) + J^*(y,p(y,R,t)) \right).
\end{align*}$$

In particular, we obtain

$$\begin{align*}
\rho_{\beta}(x,t) & = \sum_{\alpha, \beta, R; R'} \rho_{\beta R;\alpha R'}(x,R,t) \eta_{\alpha R',\beta R'}(t), \\
j_{\beta}(x,t) & = \sum_{\alpha, \beta, R; R'} j_{\beta R';\alpha R'}(x,R,t) \eta_{\alpha R',\beta R'}(t),
\end{align*}$$

where

$$\begin{align*}
\rho_{\beta R';\alpha R'}(x,R,t) & = e^{\frac{\Phi(x)}{2}} \left( \delta_{R'R} + \delta_{R'R'} \right) e^{i\Delta(R',x,R';t)} \\
& \times \chi_{\beta R'}^*(x,t) \chi_{\alpha R'}(x,t)
\end{align*}$$

and

$$\begin{align*}
J_{\beta R';\alpha R'}(x,R,t) & = e^{\frac{\Phi(x)}{2}} \left( \delta_{R'R} + \delta_{R'R'} \right) e^{i\Delta(R',x,R';t)} \\
& \times \chi_{\alpha R'}^*(x,t) \chi_{\beta R'}(x,t).
\end{align*}$$
\[ \hat{J}_{\beta R':\alpha R''}(x, R; t) = \frac{1}{4} \delta_{RR''} e^{i\Delta(x, R', R'': t)} \chi^*_\beta R''(x, t) \left( J(x, p(x, R; t)) \chi^*_\alpha R''(x, t) \right) \]
\[ + \frac{1}{4} \delta_{RR'} \left( J^*(x, p(x, R; t)) \chi^*_\beta R''(x, t) \right) e^{i\Delta(x, R', R'': t)} \chi^*_\alpha R''(x, t) \]
\[ + \frac{1}{4} \delta_{RR''} \left( J^*(x, p(x, R; t)) e^{i\Delta(x, R', R'': t)} \chi^*_\beta R'(x, t) \right) \chi^*_\alpha R''(x, t) \]
\[ + \frac{1}{4} \delta_{RR'} \chi^*_\beta R''(x, t) \left( J(x, p(x, R; t)) e^{i\Delta(x, R', R'': t)} \chi^*_\alpha R''(x, t) \right). \]

Note that in the expression (39) for \( G_R(x, y; t) \) the lattice site \( R \) always appears as one of the indices of the single-particle density matrix. Then, since \( \chi_{oR}(x, t) \) can be reasonably expected to be nonzero only for \( x \) close to \( R \) (see the expansion (28)), we can expect the \( \rho_R(x, t) \) and \( J_R(x, t) \) of (42) to be nonzero only for \( x \) close to \( R \) as well.

D. Site polarizations and magnetizations

The equations (42, 43) are precisely the ones that would be written down in a model for isolated atoms, where there \( G_R(x, y; t) \) would be the Green function for the \( R^{th} \) atom. The “special point” \( R \) has been identified for the charge-current distribution about lattice site \( R \), 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 \( \exp(-i\Delta(x, y, R; t)) \) were introduced in (40). A difference between our problem and that of isolated atoms, of course, is that in general
\[ K_R(x, t) = \nabla \cdot J_R(x, t) + \frac{\partial \rho_R(x, t)}{\partial t} \neq 0, \]

since electrons can move from the region nearest one lattice site to regions nearest others. Thus generally the site charges
\[ Q_R(t) = \int \rho_R(x, t) dx = e \sum_{\alpha} \eta_{o\alpha R}(t) \]
are time dependent. From the dynamical equation (37) we find that we can write
\[ \frac{dQ_R(t)}{dt} = \sum_{R'} I(R, R'; t), \]
where
\[ I(R, R'; t) = \frac{e}{\hbar} \sum_{\alpha, \lambda} \left( \tilde{H}_{\alpha R\lambda R'}(t) \eta_{\lambda R''; oR}(t) \right) \]
\[ - \eta_{oR\lambda R'}(t) \tilde{H}_{\lambda R''; oR}(t). \]
(50)
Since
\[ I(R', R; t) = -I(R, R'; t), \]
(51)
\( I(R, R'; t) \) can be interpreted as the net current flowing from site \( R' \) to site \( R \) and thus as a link current.

With the site charges and link currents defined, we can now define microscopic “free” charge and current densities associated with them,
\[ \rho_{f}(x, t) = \sum_{R} Q_R(t) \delta(x - R), \]
\[ j_{f}(x, t) = \frac{1}{2} \sum_{R, R'} s(x; R, R') I(R, R'; t). \]
(53)
The first simply takes the free charge density to be the sum of the changes associated with each lattice site placed at that lattice site. The second introduces a microscopic current density by distributing the net current from site \( R' \) to \( R \) along the path from \( R' \) to \( R \) defined by \( C(R, R') \). From (49,51) and the second of the properties (13) we immediately find
\[ \nabla \cdot j_f(x, t) + \frac{\partial \rho_f(x, t)}{\partial t} = 0. \]
(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 \( p_R(t) \) as
\[ p_R(t) = \int s(x; y, R) \rho_f(y, t) dy, \]
(55)
and define preliminary site magnetization fields as
\[ \tilde{m}_R^i(x, t) = \frac{1}{c} \int \alpha^{jk}(x; y, R') j_R^k(y, t) dy. \]
(56)
Since \( \rho_R(x, t) \) and \( j_R(x, t) \) are nonzero only for \( x \) close to \( R \), we can expect \( p_R(t) \) and \( \tilde{m}_R(x, t) \) to share that property as well. Introducing associated total microscopic polarization and magnetization fields,
\[ p(x, t) = \sum_R p_R(x, t), \]
\[ \tilde{m}(x, t) = \sum_R \tilde{m}_R(x, t), \]
(57)
with the use of (13) we obtain
\[ \langle \dot{\rho}(x, t) \rangle = -\nabla \cdot p(x, t) + \rho_F(x, t), \] (58)
\[ \langle \dot{j}(x, t) \rangle = \frac{\partial p(x, t)}{\partial t} + c \nabla \times \tilde{m}(x, t) + \tilde{j}(x, t) + j_F(x, t), \]
where
\[ \tilde{j}(x, t) = -\sum_R \int s(x; y, R) K_R(y, t) dy - j_F(x, t). \] (59)

In contrast with the problem of isolated atoms, here in general we have a time dependent \( \rho_F(x, t) \), a nonzero \( \dot{j}_F(x, t) \), and a nonzero \( \tilde{j}(x, t) \), all arising because the functions in \(\{K_R(x, t)\}\) are generally nonzero as charge moves from site to site. Nonetheless, since total charge is conserved we have
\[ \sum_R K_R(x, t) = 0, \]
and it is easy to confirm that
\[ \nabla \cdot \tilde{j}(x, t) = 0. \]

To complete our treatment of the site polarizations and magnetizations we express the divergenceless \( \tilde{j}(x, t) \) in terms of the curls of magnetizations associated with the different sites. We begin by writing
\[ \tilde{j}(x, t) = \sum_{\alpha, \beta} \tilde{j}_{\beta R''; R'}(x, t) \eta_{\alpha R''; \beta R'}(t), \]
where the expressions (47) and (53) for \( K_R(y, t) \) and \( j_F(x, t) \) respectively are used to identify \( \tilde{j}_{\beta R''; R'}(x, t) \),
\[ \tilde{j}_{\beta R''; R'}(x, t) = -\sum_R \int s(x; y, R) \Gamma_{R, R''}^{R', \beta} \eta_{\alpha R''; \beta R'}(t) dt \]
\[ - \frac{1}{2} \sum_{R, R'} s(x; R, R') \alpha R'' \beta R''' \eta_{\alpha R''; \beta R'}(t), \] (60)
where
\[ \eta_{\alpha R''; \beta R'}(t) = \delta_{R'' R'} \delta_{R R'} \tilde{H}_{\beta R', \alpha R'}(t) \]
\[ - \delta_{R'' R'} \delta_{R R'} \tilde{H}_{\beta R', \alpha R'}(t), \] (61)
and
\[ \Gamma_{R}^{R', \beta R'}(x, t) = \]
\[ \nabla \cdot \tilde{j}_{\beta R''; R'}(x, R; t) + \frac{\partial \rho_{\beta R''; R'}(x, R; t)}{\partial t} \]
\[ + \frac{1}{\hbar} \sum_{\mu, \nu, R_1, R_2} \rho_{\mu R_2; \nu R_1}(x, R_1, R_2) \delta_{\mu R_2; \nu R_1, R_2}(t), \]
with
\[ \delta_{\mu R_2; \nu R_1, R_2}(t) = \delta_{\beta \nu} \delta_{\alpha R_2} e^{i\Delta(R_1, R_2, t) \tilde{H}_{\beta R', \alpha R'}(t)} - \delta_{\nu \alpha} \delta_{\beta R_2} e^{i\Delta(R_1, R_2, t) \tilde{H}_{\beta \nu R', \alpha R'}(t)} - \delta_{\nu \alpha} \delta_{\beta R_2} \delta_{\mu R_1} \delta_{\mu R_2} \tilde{H}_{\beta R', \alpha R'}(t). \] (62)

We associate portions of \( \tilde{j}(x, t) \) with each lattice site in an obvious way,
\[ \tilde{j}(x, t) = \sum_R \tilde{j}_R(x, t), \]
where
\[ \tilde{j}_R(x, t) = \sum_{\alpha, \beta, R''} \tilde{j}_{\beta R''; R'}(x, R; t) \eta_{\alpha R''; \beta R'}(t), \] (64)
with
\[ \tilde{j}_{\beta R''; R'}(x, R; t) = \frac{1}{2} (\delta_{R R'} + \delta_{R' R'}) \tilde{j}_{\beta R''; R'}(x, t). \] (65)
We then define
\[ \tilde{m}_R^j(x, t) = \frac{1}{c} \int \alpha^k(x, y; R) \tilde{j}^k_R(y, t) dy, \] (66)
which, following the arguments used above for \( p_R(x, t) \) and \( m_R(x, t) \), can be expected to be nonzero only for \( x \) close to \( R \). Using the relator properties (13), we obtain
\[ \tilde{j}_R(x, t) = c \nabla \times \tilde{m}_R(x, t), \]
as desired. We can now write
\[ m_R(x, t) \equiv \tilde{m}_R(x, t) + \tilde{m}_R(x, t) \]
and, with
\[ m(x, t) = \sum_R m_R(x, t), \] (67)
we can write the second of (58) as
\[ \langle \dot{j}(x, t) \rangle = \frac{\partial p(x, t)}{\partial t} + c \nabla \times m(x, t) + j_F(x, t). \] (68)

E. Summary

We can now separate the “free” and “bound” charge and current densities, writing the total expectation values of the microscopic charge and current density operators as
\[ \langle \dot{\rho}(x, t) \rangle = \rho_B(x, t) + \rho_F(x, t), \]
\[ \langle \dot{j}(x, t) \rangle = \dot{j}_B(x, t) + j_F(x, t). \]
The free charge density is given in terms of site charges \( Q_R(t) \) by (52) and the free current density in terms of link currents \( I(R, R'; t) \) by (53); the site charges evolve due to 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
\[ \rho_B(x, t) = -\nabla \cdot p(x, t), \] (69)
\[ j_B(x, t) = \frac{\partial p(x, t)}{\partial t} + c \nabla \times m(x, t), \]
which guarantee that the bound charge and current densities satisfy continuity well,
\[ \nabla \cdot j_B(x, t) + \frac{\partial \rho_B(x, t)}{\partial t} = 0. \quad (70) \]
The microscopic polarization and magnetization fields can be broken up into site contributions,
\[ p(x, t) = \sum_R p_R(x, t), \quad (71) \]
\[ m(x, t) = \sum_R m_R(x, t). \]

\[
m^j_R(x, t) = \frac{1}{c} \sum_{\alpha, \beta, R', R''} \left[ \int \alpha^j(x; y, R) \left( j^k_{\beta R' \alpha R''}(y, R; t) + j^k_{\beta R' \alpha R''}(y, R; t) \right) dy \right] \eta_{\alpha R' \beta R''}(t), \quad (73)\]

where \( j^k_{\beta R' \alpha R''}(y, R; t) \) and \( j^k_{\beta R' \alpha R''}(y, R; t) \) 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 \( \eta_{\alpha R' \beta R''}(t) \), 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 \( I(R, R'; t) \) 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 \( \{p_R(x, t)\} \) and \( \{m_R(x, t)\} \) the bound charge and current densities (69) will satisfy continuity (70). 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 \( W_\alpha(R) \) and \( W_\beta(R') \) are assumed to have no common support if \( R \neq R' \). 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
\[
\langle \dot{\rho}(x, t) \rangle = \sum_R \left( -\nabla \cdot p_R(x, t) + Q_R \delta(x - R) \right), \quad (74) \]
\[
\langle \dot{j}(x, t) \rangle = \sum_R \left( \frac{\partial p_R(x, t)}{\partial t} + \nabla \times m_R(x, t) \right), \]
where \( Q_R \) is the fixed electronic charge associated with lattice site \( R \), and \( p_R(x, t) \) and \( m_R(x, t) \) 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 (74). 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 \( E(x, t) \rightarrow E(t) \), 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, \( H_0(x, p(x)) \), using the expression (9). This is a standard model often used to calculate the optical response of crystals [10]. For the paths \( C(x, y) \) in (12) we take straight lines.

With \( B(x, t) = 0 \) and \( E(t) \) uniform we have \( \Delta(x, y, z; t) \rightarrow 0 \) from (29), \( \chi_{\alpha R}(x, t) \rightarrow W_\alpha(R) \) from (28) and the discussion preceding it, \( \Omega^0_\alpha(y; t) \rightarrow (y - x) \cdot E(t) \) from (20) and the choice of a straight-line path, and \( \eta^{k}(x, y; t) \rightarrow \eta^{k}(x) \) from (19). The dynamical equation (37) then simplifies to
\[
\frac{i}{\hbar} \frac{\partial \eta_{\alpha R \beta R'}}{\partial t} = \sum_{\lambda, R''} \left( H_{\alpha R \lambda R''}(t) \eta_{\lambda R'' \beta R'}(t) - \eta_{\alpha R \lambda R''}(t) H_{\lambda R'' \beta R'}(t) \right) \\
- \left( e(R - R') \cdot E(t) \right) \eta_{\alpha R \beta R'}(t), \quad (75)\]
where from (35) we have used
\[ H_{α,R;R'}(t) \rightarrow H_{α,R;R'}(t) \]
\[ = \int W_{α,R}^*(x)H_0(x,p(x))W_{α,R'}(x)dx \]
\[ - \frac{e}{2} E(t) \cdot \int \left( W_{α,R}^*(x)(x-R')W_{α,R'}(x) \right) \]
\[ + W_{α,R}^*(x)(x-R)W_{α,R'}(x)dx, \]

as well as the Hermiticity of \( H_0(x,p(x)) \). Using the orthogonality of the Wannier functions (22) and translational invariance (23) we can manipulate this into the form
\[ H_{α,R;R'}(t) = H_{α}(R - R'; t), \]
where
\[ H_{α}(R; t) = \int W_{α,R}^*(x-R)H_0(x,p(x))W_{α}(x)dx \]
\[ - eE(t) \cdot \int W_{α,R}^*(x-R)xW_{α}(x)dx. \]

We now look at the transform of \( η_{α,R;R'}(t) \) into crystal momentum space. Using the identities
\[ \frac{Ω_{uc}}{(2π)^3} \int _{BZ} e^{i(k-R')R}dk = δ_{RR'}, \]
\[ \frac{Ω_{uc}}{(2π)^3} \sum _{R,R'} e^{i(k-k')R} = δ(k-k'), \]

where we restrict \( k \) and \( k' \) to the first Brillouin zone, and \( R \) and \( R' \) are lattice sites, we introduce the Fourier transform
\[ η_{α;k;k'}(t) = \frac{Ω_{uc}}{(2π)^3} \sum _{R,R'} e^{i(k'-R'-kR)}η_{α,R;R'}(t), \]

and from the dynamical equation (75) for \( η_{α,R;R'}(t) \) we obtain the corresponding equations for \( η_{α;k;k'}(t) \),
\[ i\hbar \frac{∂η_{α;k;k'}(t)}{∂t} = \sum _{γ} \left( H_{αγ}(k; t)η_{γ;k;k'}(t) - η_{α;k;γ'}(t)H_{γβ}(k'; t) \right) \]
\[ - ieE(t) \cdot \left( \frac{∂}{∂k} + \frac{∂}{∂k'} \right) η_{α;k;k'}(t), \]

where
\[ H_{αγ}(k; t) = \sum _{R} e^{-ik'R}H_{αγ}(R; t). \]

From the inverse Fourier transform of (79),
\[ η_{α,R;R'}(t) = \frac{Ω_{uc}}{(2π)^3} \int e^{-i(k'-R'-kR)}η_{α;k;k'}(t)dkdk', \]

where we have used (78), we see that if we have a state that shares the translational symmetry of the lattice, for which \( η_{α,R;R'}(t) \) depends only on \( (R - R') \), we have \( η_{α;k;k'}(t) \) of the form
\[ η_{α;k;k'}(t) = η_{α,β}(k; t)δ(k - k'). \]

If this holds initially then the dynamical equation (80) guarantees that it will hold at all later times, with
\[ i\hbar \frac{∂η_{α,β}(k; t)}{∂t} = \sum _{γ} \left( H_{αγ}(k; t)η_{γ,β}(k; t) - η_{α,γ}(k; t)H_{γ,β}(k; t) \right) \]
\[ - ieE(t) \cdot \frac{∂η_{α,β}(k; t)}{∂k}. \]

We can write (81) in a more familiar form by introduce a Bloch function associated with each Wannier function,
\[ φ_{αk}(x) = \sqrt{\frac{Ω_{uc}}{(2π)^3}} \sum _{R} e^{ikx}u_{αk}(x), \]

They are orthonormal according to
\[ \int φ_{αk}^*(x)φ_{βk'}(x)dx = δ_{αβ}δ(k - k'), \]

and in the second line of (85) we have introduced the periodic function \( u_{αk}(x) \), where \( u_{αk}(x) = u_{αk}(x + R) \) for any lattice constant \( R \). Using the inverse relation of (85),
\[ W_{αR}(x) = \sqrt{\frac{Ω_{uc}}{(2π)^3}} \int e^{-ikx}φ_{αk}(x)dx \]
in (77), we find we can write (81) as
\[ H_{αγ}(k; t) = \frac{1}{Ω_{uc}} \int \left( W_{α}^*(x-R)H_0(x,p(x))W_{γ}(x)dx \right) \]
\[ = \frac{1}{Ω_{uc}} \int u_{αk}^*(x)H_0(x,p(x) + hk)u_{γk}(x)dx, \]

with the second integral ranging over any unit cell, and
\[ ξ_{αγ}(k) = \frac{1}{Ω_{uc}} \int W_{α}^*(x-R)xW_{γ}(x)dx \]

where \( ξ_{αγ}(k) \) 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 [11]. Dropping the matrix indices and writing, for example, \( \eta(k; t) \) for the matrix with elements \( \eta_{\alpha\beta}(k; t) \), we can then write (84) in the standard matrix form,

\[
\frac{i \hbar}{\partial t} \eta(k; t) = \left[ \hat{H}^0(k) - e \xi(k) \cdot \mathbf{E}(t), \eta(k; t) \right] - ie \mathbf{E}(t) \cdot \frac{\partial \eta(k; t)}{\partial k}.
\]

When considering optical response, with the electric field treated as uniform, one is usually interested in the average of the magnetization term will vanish, and we will only recover contributions from the polarization \( p(x, t) \) and the free current density \( j_F(x, t) \). Using the decomposition (71) of \( p(x, t) \) into contributions from different lattice sites, and noting that the \( p_R(x, t) \) for different \( R \) will be the same except for a translation associated with the difference in the lattice sites, we can introduce a spatially averaged polarization

\[
P(t) = \frac{1}{\Omega_{uc}} \int p_R(x, t) dx,
\]

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

\[
j_F(x, t) = \sum_R j_{F,R}(x, t),
\]

where

\[
j_{F,R}(x, t) = \frac{1}{2} \sum_{R'} s(x; R, R') I(R, R'; t),
\]

we can introduce a free current density,

\[
J_F(t) = \frac{1}{\Omega_{uc}} \int j_{F,R}(x, t) dx,
\]

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

\[
J(t) = \frac{dP(t)}{dt} + J_F(t).
\]

Using the expression (55,44) for \( p_R(x, t) \) and \( p_R(y, t) \), together with the form (12) for \( s(x; y, R) \), the use of the Fourier decomposition (82) yields

\[
P(t) = e \sum_{\alpha, \beta} \int \frac{dk}{(2\pi)^3} \xi_{\beta\alpha}(k) \eta_{\alpha\beta}(k; t).
\]

Similarly, performing the integral (53), and in the expression (50) for \( I(R, R'; t) \) replacing the general \( \hat{H}_{\alpha R, \lambda R'}(t) \) by \( H_{\alpha \lambda}(R - R'; t) \) (see (77) and preceding), we obtain

\[
J_F(t) = \frac{e}{\hbar} \sum_{\alpha, \lambda} \int \frac{dk}{(2\pi)^3} \left( \frac{\partial}{\partial k} \left( H_{\alpha \lambda}^0(k) - e \xi_{\alpha \lambda}(k) \cdot \mathbf{E}(t) \right) \right) \eta_{\lambda \alpha}(k; t),
\]

where we have used (86) for the transform (81) \( H_{\alpha \lambda}(k; t) \). In matrix form we write (94,95) as

\[
P(t) = e \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \xi(t) \right],
\]

\[
J_F(t) = \frac{e}{\hbar} \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \left( \partial(H^0 - e \xi \cdot \mathbf{E}(t)) \right) \eta(t) \right],
\]

where \( \partial^n = \partial / \partial k^n \). Using these results in the expression (93) for \( J(t) \), together with the dynamical equation (88) we obtain

\[
J(t) = e \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ v \eta(t) \right],
\]

where the matrix

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

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 \( \mathbf{E}(t) \) [36]; 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 \( \{ \phi_{\alpha k}(x) \} \) of (85) at each \( k \) it is often convenient to work with a new set of basis functions \( \{ \tilde{\phi}_{\alpha k}(x) \} \), related to the original set at each \( k \) by a unitary transformation,

\[
\tilde{\phi}_{\alpha k}(x) = \sum_{\alpha} \phi_{\alpha k}(x) U_{\alpha\alpha}(k),
\]

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

\[
\tilde{H}^0 \equiv U^\dagger H^0 U,
\]

\[
\tilde{\eta} \equiv U^\dagger \eta U,
\]

in our short-hand notation, we find that the equation for \( \tilde{\eta}(t) \) that follows from (88) is

\[
\frac{i\hbar}{\partial t} \frac{\partial \tilde{\eta}(t)}{\partial t} = \left[ \tilde{H}^0 - e \tilde{\xi} \cdot \mathbf{E}(t), \tilde{\eta}(t) \right] - ie \mathbf{E}(t) \cdot \partial \tilde{\eta}(t),
\]

(100)
where
\[ \xi \equiv U^\dagger \xi U + iU^\dagger \partial U, \]
and the matrix \( \tilde{\xi} \) at \( k \) has components
\[ \tilde{\xi}_{nm}(k) = \frac{i}{\Omega_{uc}} \int_{uc} \bar{u}_{nk}(x) \frac{\partial \bar{u}_{mk}(x)}{\partial k} \ dx, \]
(cf. (87)) where \( \bar{u}_{nk}(x) \) is the periodic component, \( \bar{u}_{nk}(x + R) = \bar{u}_{nk}(x) \) for any lattice vector \( R \), associated with the basis function \( \phi_{nk}(x) \),
\[ \tilde{\phi}_{nk}(x) = \frac{1}{\sqrt{(2\pi)^3}} e^{i\mathbf{k} \cdot \mathbf{x}} \bar{u}_{nk}(x). \]

Such a transformation is often done as a prelude to a perturbation calculation, and chosen so that \( H^0 \) is diagonal, but more general transformations can be considered. In terms of the new matrices we find that the current density (97) can be written as
\[ J(t) = e \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \bar{\eta} \tilde{\eta}(t) \right], \]
where the matrix
\[ \bar{\eta} = \partial H^0 - \frac{i}{\hbar} [\tilde{\xi}, H^0] \]
(cf. (98)). Thus the form of both the dynamical equations (100) and the expression (104) for the current density are invariant under such a set of unitary transformations \( \{ U(k) \} \). The same does not hold for the polarization and free current density that lead to the current density via (93). In place of (96) we obtain
\[ P(t) = e \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \tilde{\xi} \bar{\eta}(t) \right], \]
\[ J_P(t) = \frac{\hbar}{e} \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \left( \frac{\partial}{\partial k} U^\dagger U \right) \bar{\eta}(t) \right], \]
\[ J_P(t) = \frac{\hbar}{e} \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \left( \frac{\partial}{\partial k} U^\dagger U \right) \eta(t) \right] \]
\[ + i \frac{\hbar}{e} \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \left( \frac{\partial}{\partial k} U^\dagger U \right) \bar{\eta}(t) \right] \]
\[ + i \frac{\hbar}{e} \int \frac{dk}{(2\pi)^3} \text{Tr} \left[ \left( \frac{\partial}{\partial k} U^\dagger U \right) \tilde{\eta}(t) \right]. \]

It is only in the Bloch basis (85) associated with the maximally localized Wannier functions that our expressions for \( P(t) \) and \( J_P(t) \) take a simple form (96), since it is those Wannier functions that were used for the introduction of our quantities \( p(x, t), \ m(x, t), \ rho_P(x, t) \) and \( j_P(x, t) \). 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 (106) 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" [14]. 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 \( J_P \) 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. Perturbation 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 (99) to be that which diagonalizes the Hamiltonian, so
\[ \bar{H}_{nm}(k) = \delta_{nm} \hbar \omega_n(k), \]
and introducing a perturbation expansion of \( \eta_{nm}(k; t) \) in orders of the electric field,
\[ \eta_{nm}(k; t) = \eta_{nm}^{(0)}(k) + \eta_{nm}^{(1)}(k; t) + \ldots \]
we take
\[ \eta_{nm}^{(0)}(k) = f_n \delta_{nm}, \]
where \( f_n = 0 \) or 1 for each \( n \), independent of \( k \). For an electric field
\[ E(t) = E(\omega)e^{-i\omega t} + E(-\omega)e^{i\omega t}, \]
with \( E(-\omega) = E^*(\omega) \), the usual perturbation treatment of (100) gives
\[ \eta_{nm}^{(1)}(k; t) = \eta_{nm}^{(1)}(k; \omega)e^{-i\omega t} + \eta_{nm}^{(1)}(k; -\omega)e^{i\omega t}, \]
where
\[ \eta_{nm}^{(1)}(k; \omega) = \frac{\epsilon_{nm}(k) \cdot E(\omega)f_{nm}}{\hbar(\omega_n(k) - \omega)}, \]
where \( f_{nm} = f_m - f_n \) and likewise for \( \omega_{nm}(k) \). Then either calculating the first order result for
\[ J^{(1)}(t) = J(\omega)e^{-i\omega t} + J(-\omega)e^{i\omega t} \]
directly from (104), or calculating the first order contributions to \( P(t) \) and \( J_P(t) \) from (104,105) and using the expressions in (93), we obtain
\[ J^a(\omega) = \sigma^{ab}(\omega)E^b(\omega), \]
where the conductivity \( \sigma^{ab}(\omega) \) is given by
\[ \sigma^{ab}(\omega) = -i\omega \sum_{n,m} \int \frac{dk}{(2\pi)^3} \frac{f_{nm}(\omega_{nm}(k) - \epsilon_n + \epsilon_m)}{(\omega_{nm}^2 - \omega^2)}, \]
\[ - i\omega \sum_{n,m} \int \frac{dk}{(2\pi)^3} \frac{f_{nm}(\omega_{nm}(k) - \epsilon_n + \epsilon_m)}{(\omega_{nm}^2 - \omega^2)}. \]
Note that in general the conductivity can be finite as \( \omega \to 0, \)
\[
\lim_{\omega \to 0} \sigma^{ab}(\omega) = \frac{e^2}{h} \sum_{n,m} \frac{d}{2\pi} \int \frac{dk}{\omega_{nm}} \left( \frac{\partial \xi_{nm}}{\partial k^b} - \frac{\partial \xi_{nm}}{\partial k^a} \right),
\]
(108)
which is the expression for the zero frequency transverse conductivity of a topologically nontrivial insulator [37][28]; here we have used
\[
\partial \times \xi - i \xi \times \xi = 0,
\]
and the fact that the corresponding equation holds for \( \tilde{\xi} \) as well. Note that the zero frequency transverse conductivity is identified in our treatment with the “free” current density \( \mathbf{J}_F, \) as follows immediately from the expression (93) for \( \mathbf{J}(t) \). This zero frequency response vanishes for a topologically trivial insulator is from the polarization, \( \mathbf{J}^{(1)}(t) = d\mathbf{P}^{(1)}(t)/dt, \) where
\[
\mathbf{P}^{(1)}(t) = \mathbf{P}(\omega)e^{-i\omega t} + \mathbf{P}(-\omega)e^{i\omega t},
\]
and
\[
\mathbf{P}(\omega) = \frac{e^2}{h} \sum_{n,m} \int \frac{dk}{(2\pi)^3} \frac{f_{mn}(k) (\xi_{nm}(k) \cdot \mathbf{E}(\omega))}{(\omega_{nm}(k) - \omega)},
\]
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. 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
\[
\eta_{\alpha R;\beta R'} = f_{\alpha} \delta_{\alpha \beta} \delta_{RR'},
\]
where \( f_{\alpha} = 0 \) or 1, in the expressions (72,73) for \( \mathbf{p}_R(x,t) \) and \( \mathbf{m}_R(x,t) \). Those quantities are then of course independent of time, and the expressions simplify considerably. Taking straight-line paths for \( C(x,y) \) (see Appendix A) we have
\[
\int s^i(w;x,y)dw = (x^i - y^i),
\]
\[
\int \alpha^{jk}(w;x,y)dw = \frac{1}{2} \epsilon^{ijk}(x^m - y^m),
\]
and we find that the electric dipole moment \( \mu \) and magnetic dipole moment \( \nu \) of the charge-current distribution associated with each site are just
\[
\mu = \int \mathbf{p}_R(x)dx,
\]
\[
\nu = \int \mathbf{m}_R(x)dx,
\]
and are independent of \( \mathbf{R} \), as expected. We obtain
\[
\mu_{\alpha} = e \int W_{\alpha}(x)W_{\alpha}(x)dx,
\]
\[
\nu_{\alpha} = \mathbf{p}_{\alpha} + \mathbf{d}_{\alpha},
\]
with \( W_{\alpha}(x) \equiv W_{\alpha 0}(x) \), while \( \nu_{\alpha} \) is the sum of two contributions,
\[
\nu_{\alpha} = \nu_{\alpha} + \nu_{\alpha},
\]
arising from the two contributions to \( \mathbf{m}_R(x) \) in (73). The first term is an atomic-like contribution,
\[
\bar{\nu}_{\alpha} = \frac{1}{4\epsilon} \int x^m W_{\alpha}(x) \left( j^k(x,p(x))W_{\alpha}(x) \right) dx
\]
\[
+ \frac{1}{4\epsilon} \int x^m \left( j^k(x,p(x))W_{\alpha}(x) \right)^* W_{\alpha}(x) dx,
\]
(109)
which, for a Hamiltonian of Schrödinger form (10), gives
\[
\bar{\nu}_{\alpha} = \frac{e}{2\mu e} \int W_{\alpha}^*(x) \left( x \times \left( \frac{\hbar}{i} \frac{\partial W_{\alpha}(x)}{\partial x} - \frac{e}{c} \mathbf{A}_{\text{static}}(x)W_{\alpha}(x) \right) \right) dx,
\]
which is familiar from the corresponding expression in atomic physics, and
\[
\bar{\nu}_{\alpha} = \frac{1}{2\mu e} \int \sum_{\lambda_i R_i} R_i^m \text{Im} \left[ H_{\alpha 0;\lambda_i R_i}^R \mu_{\lambda_i R_i;\alpha 0}^{R_i}(R_1) \right],
\]
(110)
where \( H_{\alpha 0;\lambda_i R_i}^R \) is given by (76) in the limit of no applied field, or more generally by
\[
H_{\alpha R_1;\lambda R_2}^{(0)} = \int W_{\alpha R_1}^*(x)H_0(x,p(x))W_{\lambda R_2}(x)dx,
\]
(111)
where we have defined
\[
\mu_{\beta R_1;\alpha R_2}^{R_2}(R) = e \int W_{\beta R_1}(x)(x - R)W_{\alpha R_2}(x)dx.
\]
Here $\nu_0$ is the itinerant contribution to magnetic moment defined earlier [13]; taking the macroscopic polarization and magnetization to be given by

\[ P = \frac{\mu}{\Omega_{\text{uc}}}, \]
\[ M = \frac{\nu}{\Omega_{\text{uc}}}. \]

We are in agreement with earlier results from the modern theory of polarization and magnetization [38]. 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 $\alpha$ 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 current, and expand it into terms involving powers of the electromagnetic field,

\[ I(R, R'; t) = I^{(0)}(R, R') + I^{(1)}(R, R'; t) + \ldots, \]

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

\[ \tilde{H}_{\alpha R \lambda R'}(t) = \tilde{H}^{(0)}_{\alpha R \lambda R'} + \tilde{H}^{(1)}_{\alpha R \lambda R'}(t) + \ldots, \]
\[ \eta^{(0)}_{\alpha R \lambda R'}(t) = \eta^{(0)}_{\alpha R \lambda R'} + \eta^{(1)}_{\alpha R \lambda R'}(t) + \ldots. \]

For a topologically trivial insulator we have

\[ \eta^{(0)}_{\alpha R \lambda R'} = f_\alpha \delta_{\alpha \lambda} \delta_{RR'}, \tag{112} \]

as above, and we identify $\tilde{H}^{(0)}_{\alpha R \lambda R'} = H^{(0)}_{\alpha R \lambda R'}$, with $H^{(0)}_{\alpha R \lambda R'}$ given by (111). Then it immediately follows that $I^{(0)}(R, R') = 0$, while

\[ I^{(1)}(R, R'; t) = \frac{e}{i\hbar} \sum_{\alpha, \lambda} \left( \langle H^{(1)}_{\alpha R \lambda R'}(t) \eta^{(0)}_{\lambda R' ; \alpha R}(t) - \eta^{(0)}_{\alpha R \lambda R'}(t) H^{(1)}_{\lambda R' ; \alpha R}(t) \rangle \right) \]
\[ + \frac{e}{i\hbar} \sum_{\alpha, \lambda} \left( \langle H^{(0)}_{\alpha R \lambda R'}(t) \eta^{(1)}_{\lambda R' ; \alpha R}(t) - \eta^{(1)}_{\alpha R \lambda R'}(t) H^{(0)}_{\lambda R' ; \alpha R}(t) \rangle \right). \tag{113} \]

The first of these two terms gives

\[ \frac{e}{i\hbar} \sum_{\alpha, \lambda} \left( \tilde{H}^{(1)}_{\alpha R \lambda R'}(t) - H^{(1)}_{\lambda R' ; \alpha R}(t) \right) f_\alpha \delta_{\alpha \lambda} \delta_{RR'}, \]

which vanishes regardless of the form of $\tilde{H}^{(1)}_{\alpha R \lambda R'}(t)$. To investigate the second term in (113), we look at the equation for $\eta^{(1)}_{\alpha R \lambda R'}(t)$ that follows from a perturbative analysis of the general dynamical equation (37). Expanding terms in the usual way we obtain

\[ i\hbar \frac{\partial \eta^{(1)}_{\alpha R \lambda R'}(t)}{\partial t} = \sum_{\lambda', R''} \left( \tilde{H}^{(0)}_{\alpha R \lambda R'}(t) \eta^{(1)}_{\lambda R' ; \alpha R''}(t) - \eta^{(1)}_{\alpha R \lambda R'}(t) H^{(0)}_{\lambda R' ; \alpha R''} \right) \]
\[ + f_{\beta \lambda} \tilde{H}^{(1)}_{\alpha R \beta R'}, \]

and the inhomogeneous term in this equation is proportional to $f_{\beta \lambda}$. As can be confirmed by the formal solution of this equation in reciprocal space, $\eta^{(1)}_{\alpha R \lambda R'}(t)$ will vanish unless one and only one of $\alpha$ and $\beta$ is associated with a set of filled bands, so indeed $\eta^{(1)}_{\alpha R \lambda R'}(t)$ will be proportional to $f_{\beta \lambda}$. But using this result in the second term of (113), we see that the second term will involve terms such as $f_{\beta \lambda} \tilde{H}^{(0)}_{\alpha R \beta R'}$, which vanish. So we have

\[ I^{(1)}(R, R'; t) = 0. \]

Thus to first order in the optical response of a topologically trivial insulator the link current will vanish, and so we see from (49,52,53) that $j_{\rho}(x, t) = 0$ and $j_{\mu}(x, t)$ is independent of time. The entire optical response to first order is described by $p(x, t)$ and $m(x, t)$, 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, $p(x, t)$ and $m(x, t)$, which together with microscopic "free" charge and current densities, $\rho_{\mu}(x, t)$ and $j_{\rho}(x, t)$, can be used (2) to represent the expectation value of the microscopic charge and current density operators, $\langle \hat{\rho}(x, t) \rangle$ and $\langle \hat{j}(x, t) \rangle$. Our goal has been to write $p(x, t)$ and $m(x, t)$ 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. 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 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 non-vanishing only in the neighborhood of the site. Spatial averages of the microscopic polarization and magnetization can 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 \( \langle \hat{\rho}(x, t) \rangle \) and \( \langle \hat{j}(x, t) \rangle \) in terms of their “constituent sources” \( p(x, t), m(x, t), \rho_F(x, t), \) and \( j_F(x, t) \) 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. But 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 neglect of the motion of nuclei and 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 possible to restrict the formalism to a two dimensional lattice, using some care in formulating the Fourier transforms and Wannier functions.

For a given \( \langle \hat{\rho}(x, t) \rangle \) and \( \langle \hat{j}(x, t) \rangle \) 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 \( p(x, t) \) and \( m(x, t) \) to \( \langle \hat{\rho}(x, t) \rangle \) and \( \langle \hat{j}(x, t) \rangle \). 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 \( p(x, t), m(x, t), \rho_F(x, t), \) and \( j_F(x, t) \), 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 quasi-particles 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.

V. ACKNOWLEDGEMENTS

This work was supported by the Natural Sciences and Engineering Research Council of Canada (NSERC). P.T.M. acknowledges a PGS-D scholarship from NSERC. We thank Sylvia Swiecicki for many helpful discussions in the early stages of this work.

VI. APPENDICES

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 \( y \) to \( x \) by a function \( z(u) \) such that \( z(u_1) = y \) and \( z(u_2) = x \). Then from the first of (12) we have

\[
s_i(w; x, y) = \int_{u_1}^{u_2} du \frac{dz^i(u)}{du} \delta(w - z),
\]

and so

\[
- \frac{\partial s^i(w; x, y)}{\partial w^i} = - \int_{u_1}^{u_2} du \frac{dz^i(u)}{du} \frac{\partial \delta(w - z)}{\partial w^i} = \int_{u_1}^{u_2} du \frac{dz^i(u)}{du} \frac{\partial \delta(w - z)}{\partial z^i} = \int_{u_1}^{u_2} du \frac{d}{du} \delta(w - z) = \delta(w - y) - \delta(w - x),
\]

which is the first of (13).
Moving to the second of (12) we have
\[
\alpha^{jk}(w; x, y) = e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \delta(w - z), \quad (A2)
\]
so
\[
e^{ipj} \alpha^{jk}(w; x, y) = e^{ipj} e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \delta(w - z).
\]
Now
\[
e^{ipj} e^{jmn} = e^{ipj} e^{jmn} = \delta^{im} \delta^{pn} - \delta^{in} \delta^{pm},
\]
so
\[
e^{ipj} \frac{\partial \alpha^{jk}(w; x, y)}{\partial u^p}
\]
\[
= \left( \delta^{im} \delta^{pn} - \delta^{in} \delta^{pm} \right) \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \frac{\partial \delta(w - z)}{\partial w^p}
\]
\[
= \int_{u_1}^{u_2} du \frac{dz^i}{du} \frac{\partial \delta(w - z)}{\partial u^p}
\]
\[
- \int_{u_1}^{u_2} du \left( \frac{\partial}{\partial x^k} \frac{dz^i}{du} \right) \delta(w - z) + \left[ \frac{\partial z^i}{\partial x^k} \delta(w - z) \right]_{u_1}^{u_2}
\]
\[
= - \frac{\partial}{\partial u^p} \left( \int_{u_1}^{u_2} du \frac{dz^i}{du} \delta(w - z) \right) + \left[ \frac{\partial z^i}{\partial x^k} \delta(w - z) \right]_{u_1}^{u_2}.
\]
Partially integrating the second term gives
\[
e^{ipj} \frac{\partial \alpha^{jk}(w; x, y)}{\partial u^p}
\]
\[
= - \int_{u_1}^{u_2} du \frac{dz^i}{du} \frac{\partial \delta(w - z)}{\partial u^p}
\]
\[
- \int_{u_1}^{u_2} du \left( \frac{\partial}{\partial x^k} \frac{dz^i}{du} \right) \delta(w - z) + \left[ \frac{\partial z^i}{\partial x^k} \delta(w - z) \right]_{u_1}^{u_2}
\]
\[
= - \frac{\partial}{\partial u^p} \left( \int_{u_1}^{u_2} du \frac{dz^i}{du} \delta(w - z) \right) + \left[ \frac{\partial z^i}{\partial x^k} \delta(w - z) \right]_{u_1}^{u_2}.
\]
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
\[
e^{ipj} \frac{\partial \alpha^{jk}(w; x, y)}{\partial u^p} = - \frac{\partial}{\partial x^k} s^i(w; x, y) + \delta^{ik} \delta(w - x),
\]
which is the second of (13).

The final expression is derived in much the same way. We have
\[
\beta^{jk}(w; x, y) = e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \delta(w - z), \quad (A3)
\]
so
\[
e^{ipj} \frac{\partial \beta^{jk}(w; x, y)}{\partial u^p}
\]
\[
= e^{ipj} e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \delta(w - z)
\]
\[
= \left( \delta^{im} \delta^{pn} - \delta^{in} \delta^{pm} \right) \int_{u_1}^{u_2} du \frac{dz^m}{du} \frac{dz^n}{du} \frac{\partial \delta(w - z)}{\partial w^p}.
\]
and following exactly the strategy above we have
\[
e^{ipj} \frac{\partial \beta^{jk}(w; x, y)}{\partial u^p}
\]
\[
= - \frac{\partial}{\partial y^k} \left( \int_{u_1}^{u_2} du \frac{dz^j}{du} \delta(w - z) + \left[ \frac{\partial z^i}{\partial y^k} \delta(w - z) \right]_{u_1}^{u_2} \right)
\]
\[
= - \frac{\partial}{\partial y^k} s^i(w; x, y) - \delta^{ik} \delta(w - y),
\]
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 \( C(x, y) \), where for each and every \( x \) and \( y \) the path \( C(x, y) \) is the “reverse” of the path \( C(y, x) \). More precisely, if \( C(x, y) \) is specified by giving \( z(u) \) as \( u \) varies from \( u_1 \) to \( u_2 \), with \( z(u_1) = y \) and \( z(u_2) = x \), then \( C(y, x) \) is specified by the same \( z(u) \) as \( u \) varies from \( u_2 \) to \( u_1 \). We have
\[
s^i(w; x, y) = \int_{C(x,y)} dz^i \delta(w - z)
\]
\[
= \int_{u_1}^{u_2} du \frac{dz^i(u)}{du} \delta(w - z),
\]
and
\[
s^i(w; y, x) = \int_{C(y,x)} dz^i \delta(w - z)
\]
\[
= \int_{u_2}^{u_1} du \frac{dz^i(u)}{du} \delta(w - z)
\]
\[
= - \int_{u_1}^{u_2} du \frac{dz^i(u)}{du} \delta(w - z)
\]
\[
= - s^i(w; x, y),
\]
while
\[
\alpha^{jk}(w; x, y) = e^{jmn} \int_{C(x,y)} dz^m \frac{dz^n}{dz^k} \delta(w - z)
\]
\[
= e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m(u)}{du} \frac{dz^n}{dz^k} \delta(w - z),
\]
and
\[
\beta^{jk}(w; y, x) = e^{jmn} \int_{C(y,x)} dz^m \frac{dz^n}{dz^k} \delta(w - z)
\]
\[
= e^{jmn} \int_{u_2}^{u_1} du \frac{dz^m(u)}{du} \frac{dz^n}{dz^k} \delta(w - z)
\]
\[
= - e^{jmn} \int_{u_1}^{u_2} du \frac{dz^m(u)}{du} \frac{dz^n}{dz^k} \delta(w - z)
\]
\[
= - \alpha^{jk}(w; x, y).
\]
Finally, we consider the special case of a straight-line path \(C(x, y)\) for each \(x\) and \(y\). That is, taking \(u_1 = 0\) and \(u_2 = 1\), for the path \(C(x, y)\) we have
\[
z = y + u(x - y)
\]
Then we have
\[
\frac{dz^i(u)}{du} = x^i - y^i,
\]
\[
\frac{\partial z^n}{\partial x^k} = u\delta^{nk}
\]
and so from (A1) we have
\[
s^i(w; x, y) = 
\int_0^1 (x^i - y^i)\delta(w - y - u(x - y))du,
\]
while from (A2) we have
\[
\alpha^{jk}(w; x, y) = 
\int_0^1 (x^m - y^m)\delta(w - y - u(x - y))udu,
\]
and from (A3) we have
\[
\beta^{jk}(w; x, y) = 
\int_0^1 (x^m - y^m)\delta(w - y - u(x - y))(1 - u)du.
\]
Since a straight-line path is symmetric, in the terminology used above, we expect (A4,A5,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
\[
(\alpha R) \rightarrow n
\]
\[
R \rightarrow R_n
\]
Our set of non-orthogonal states (24) are then labelled \(\{W_n'(x, t)\}\); the elements of the overlap matrix \(S(t)\) characterizing them are
\[
S_{nm}(t) = \int W_n(x, t)W_m(x, t)dx
\]
\[
e^{i\Phi(R_n, R_m; t)} \int W_n^*(x)e^{i\Delta(R_n, x, R_m; t)}W_m(x)dx
\]
Clearly \(S(t)\) is Hermitian, and although its matrix elements are in general not gauge-invariant, they can be written as
\[
S_{nm}(t) = e^{i\Phi(R_n, R_m; t)} \hat{S}_{nm}(t),
\]
where the \(\hat{S}_{nm}(t)\) are gauge-invariant,
\[
\hat{S}_{nm}(t) = \int W_n^*(x)e^{i\Delta(R_n, x, R_m; t)}W_m(x)dx
\]
and since
\[
\hat{S}_{nm}(t) = e^{i\Phi(R_n, R_m; t)}S_{nm}(t)
\]
the matrix \(\hat{S}(t)\) is also Hermitian.

We seek a set \(\{W_n(x, t)\}\) spanned by the original set that are orthogonal,
\[
\hat{W}_n(x, t) = \sum_p W_p'(x, t)C_{pn}(t),
\]
where
\[
\int \hat{W}_n^*(x, t)\hat{W}_m(x, t)dx = \sum_{p, l} C_{np}'(t)C_{lm}(t) = \delta_{nm},
\]
or in matrix notation
\[
\hat{C}^\dagger(t)\hat{S}(t)\hat{C}(t) = I.
\]
There are of course many matrices \(\hat{C}(t)\) that can be found that satisfy this condition. However, the desired matrix \(\hat{C}(t)\) yielding the minimization of (26) is the Hermitian matrix satisfying
\[
\hat{C}(t) = S^{-1/2}(t),
\]
[23]. That is, it is the “Hermitian square root” of the inverse of the overlap matrix \(S(t)\).

To see the structure of \(\hat{C}(t)\), and the nature of the resulting \(\{\hat{W}_n(x, t)\}\), first introduce \(T(t)\) as the inverse of \(S(t)\),
\[
\sum_l S_{nl}(t)T_{lm}(t) = \delta_{nm},
\]
or
\[
\sum_l \hat{S}_{nl}(t)e^{i\Phi(R_n, R_l; t)}T_{lm}(t) = \delta_{nm}.
\]
Introducing \(\hat{T}(t)\) according to
\[
\hat{T}_{lm}(t) = T_{lm}(t)e^{-i\Phi(R_l, R_m; t)}
\]
we have
\[
e^{-i\Phi(R_m, R_l; t)}\sum_l \hat{S}_{nl}(t)\hat{T}_{lm}(t)e^{i\Delta(R_n, R_l, R_m; t)} = \delta_{nm},
\]
or
\[
\sum_l \hat{S}_{nl}(t)\hat{T}_{lm}(t)e^{i\Delta(R_n, R_l, R_m; t)} = \delta_{nm},
\]
and we see that the elements of $\hat{T}(t)$ must be gauge-invariant, since everything else in the equation is. Since $C(t)$ is the square root of $T(t)$ we have
\[ \sum_l C_{nl}(t)C_{lm}(t) = T_{nm}(t) = \hat{T}_{nm}(t)e^{i\Phi(R_n, R_m; t)}. \] (B2)

Now the $C(t)$ we seek is Hermitian, requiring
\[ C^*_{nl}(t) = C_{ln}(t). \] (B3)

Introducing $\hat{C}(t)$ according to
\[ \hat{C}_{nl}(t) = C_{nl}(t)e^{-i\Phi(R_n, R_l; t)}, \] (B4)
then using (B3) we see that
\[ \hat{C}^*_{nl}(t) = C^*_{nl}(t)e^{i\Phi(R_n, R_l; t)} = C_{ln}(t)e^{-i\Phi(R_l, R_n; t)} = \hat{C}_{ln}(t), \]
and so the $\hat{C}(t)$ we seek is Hermitian and, from (B2), satisfies
\[ \sum_l \hat{C}_{nl}(t)\hat{C}_{lm}(t)e^{i\Phi(R_n, R_l; t)+\Phi(R_l, R_m; t)} = \hat{T}_{nm}(t)e^{i\Phi(R_n, R_m; t)}, \] (B5)
or
\[ \sum_l \hat{C}_{nl}(t)\hat{C}_{lm}(t)e^{i\Delta(R_n, R_l, R_m; t)} = \hat{T}_{nm}(t), \]
a gauge-invariant equation, and so the matrix $\hat{C}(t)$ we seek is gauge invariant.

In terms of our new quantities we can write (B1) as
\[ \bar{W}_n(x, t) = \sum_p W_p(x)e^{i\Phi(x, R_p; t)}\hat{C}_{pn}(t)e^{i\Phi(R_p, x; t)} = e^{i\Phi(x, R_p; t)}\sum_p W_p(x)\hat{C}_{pn}(t)e^{i\Phi(R_p, x; t)} \times e^{i\Phi(x, R_p; t)}e^{i\Phi(R_p, x; t)} = e^{i\Phi(x, R_p; t)}\chi_n(x, t), \]
where
\[ \chi_n(x, t) \equiv \sum_p W_p(x)\hat{C}_{pn}(t)e^{i\Delta(x, R_p, R_n; t)} \] (B6)
is clearly gauge-invariant, thus establishing (27).

We can easily work out an expansion for the $\bar{W}_n(x, t)$ where the overlap between nonidentical $W_n(x, t)$ is small. Defining a matrix $s$,
\[ s(t) = S(t) - I, \]
where $I$ is the identity matrix, we have
\[ S^{-1/2}(t) = (I + s(t))^{-1/2} = I - \frac{1}{2}s(t) + \frac{3}{8}s^2(t) + \ldots \] (B7)
where since $S$ is Hermitian $s$ will be as well. In terms of components we have
\[ s_{nm}(t) = S_{nm}(t) - \delta_{nm} = e^{i\Phi(R_n, R_m; t)} \times \left[ \left( \int W_n^*(x)e^{i\Delta(R_n, x, R_m; t)}W_m(x)dx \right) - \delta_{nm} \right] = e^{i\Phi(R_n, R_m)}\hat{s}_{nm}(t), \]
where
\[ \hat{s}_{nm}(t) = \left( \int W^*_n(x)e^{i\Delta(R_n, x, R_m; t)}W_m(x)dx \right) - \delta_{nm} \]
is gauge invariant. Clearly the power series expansion (B7) yields the Hermitian square root of $S$, and so we have
\[ C_{pn}(t) = \delta_{pn} - \frac{1}{2}\hat{s}_{pn}(t) + \frac{3}{8}\sum_u \hat{s}_{pu}(t)\hat{s}_{un}(t) + \ldots \]
\[ = \delta_{pn} - \frac{1}{2}\hat{s}_{pn}(t)e^{i\Phi(R_p, R_n; t)} + \frac{3}{8}\sum_u \hat{s}_{pu}(t)\hat{s}_{un}(t)e^{i\Phi(R_p, R_n; t)} + \ldots \]
Then from (B4) we have
\[ \hat{C}_{pn}(t) = \delta_{pn} - \frac{1}{2}\hat{s}_{pn}(t) + \frac{3}{8}\sum_u \hat{s}_{pu}(t)\hat{s}_{un}(t)e^{i\Delta(R_p, R_n, R_m; t)} + \ldots \]
which is indeed gauge-invariant. Using this in (B6) yields
\[ \chi_n(x, t) = W_n(x) - \frac{1}{2}\sum_p W_p(x)\hat{s}_{pn}(t)e^{i\Delta(x, R_p, R_n; t)} + \frac{3}{8}\sum_{p, u} W_p(x)\hat{s}_{pu}(t)\hat{s}_{un}(t)e^{i\Delta(x, R_p, R_n; t)} \times e^{i\Delta(x, R_p, R_n; t)} + \ldots \]
Now defining as usual
\[ \Delta(x, R_p, R_u, R_n; t) \equiv \Phi(R_n, x; t) + \Phi(x, R_p; t) + \Phi(R_p, R_u; t) + \Phi(R_u, R_n; t) = \Delta(x, R_p, R_u; t) + \Delta(R_p, R_u, R_n; t), \]
we have
\[ \chi_n(x, t) = W_n(x) - \frac{1}{2}\sum_p W_p(x)\hat{s}_{pn}(t)e^{i\Delta(x, R_p, R_n; t)} + \frac{3}{8}\sum_{p, u} W_p(x)\hat{s}_{pu}(t)\hat{s}_{un}(t)e^{i\Delta(x, R_p, R_n; t)} + \ldots, \]
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 Healey [21]. For simplicity we treat the nucleus as fixed. Introducing an electron field operator \( \psi(x, t) \) and beginning with minimal coupling the charge and current density operators are given by

\[
\dot{\psi}(x, t) = e\frac{\partial}{\partial t} \psi(x, t),
\]

\[
\dot{J}(x, t) = \frac{1}{2} \psi^\dagger(x, t) \left[ J(x, p_{mc}(x, t)) \psi(x, t) \right] + \frac{1}{2} \left[ J(x, p_{mc}(x, t)) \psi(x, t) \right]^\dagger \psi(x, t),
\]

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

\[
i\hbar \frac{\partial \psi(x, t)}{\partial t} = [\psi(x, t), H_{mc}(t)]
\]

\[
= \left( H_0(x, p_{mc}(x, t)) + e\phi(x, t) \right) \psi(x, t),
\]

where

\[
H_{mc}(t) = \int \psi^\dagger(x, t) \left( H_0(x, p_{mc}(x, t)) + e\phi(x, t) \right) \psi(x, t) dx.
\]

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

\[
\psi_{sp}(x, t) = e^{-i\Phi(x, \mathbf{R}, t)} \psi(x, t),
\]

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

\[
i\hbar \frac{\partial \psi_{sp}(x, t)}{\partial t} = \hbar \frac{\partial \Phi(x, \mathbf{R}, t)}{\partial t} \psi_{sp}(x, t)
\]

\[
+ e^{-i\Phi(x, \mathbf{R}, t)} \left( i\hbar \frac{\partial \psi(x, t)}{\partial t} \right).
\]

Since

\[
\hbar \frac{\partial \Phi(x, \mathbf{R}, t)}{\partial t} = \frac{e}{c} \int s^1(w; x, \mathbf{R}) \frac{\partial A^1(w, t)}{\partial t} dw
\]

\[
- e \int s^1(w; x, \mathbf{R}) E^1(w, t) dw
\]

\[
- e \int s^1(w; x, \mathbf{R}) \frac{\partial \phi(w, t)}{\partial w^1} dw
\]

\[
= -e\Omega_{sp}^0(x, t) + e \int \frac{\partial s^1(w; x, \mathbf{R})}{\partial w^1} \phi(w, t) dw
\]

\[
= -e\Omega_{sp}^0(x, t) + e \phi(x, t) + e\phi(\mathbf{R}, t),
\]

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

\[
e^{-i\Phi(x, \mathbf{R}, t)} \left( i\hbar \frac{\partial \psi(x, t)}{\partial t} \right)
\]

\[
= e^{-i\Phi(x, \mathbf{R}, t)} \left( H_0(x, p_{mc}(x, t)) + e\phi(x, t) \right) \psi(x, t)
\]

\[
= \left( H_0(x, p(\mathbf{R}, t)) + e\phi(\mathbf{R}, t) \right) \psi_{sp}(x, t),
\]

(recall (19)) and we have

\[
i\hbar \frac{\partial \psi_{sp}(x, t)}{\partial t} = \left( H_0(x, p(\mathbf{R}, t)) - e\Omega_{sp}^0(x, t) \right) \psi_{sp}(x, t),
\]

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

\[
i\hbar \frac{\partial \psi_{sp}(x, t)}{\partial t} = \left( H_0(x, p(\mathbf{R}, t)) - e\Omega_{sp}^0(x, t) \right) \psi_{sp}(x, t),
\]

and then have

\[
i\hbar \frac{\partial \psi_{sp}(x, t)}{\partial t} = [\psi_{sp}(x, t), H_{sp}(t)],
\]

where

\[
H_{sp}(t) = \int \psi_{sp}^\dagger(x, t) \left( H_0(x, p(\mathbf{R}, t)) - e\Omega_{sp}^0(x, t) \right) \psi_{sp}(x, t) dx.
\]

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

\[
\dot{\psi}(x, t) = e\psi_{sp}^\dagger(x, t) \psi_{sp}(x, t),
\]

while the second becomes

\[
\dot{J}(x, t) = \frac{1}{2} \psi_{sp}^\dagger(x, t) \left[ J(x, p_{mc}(x, t)) \psi_{sp}(x, t) \right] e^{i\Phi(x, \mathbf{R}, t)}
\]

\[
+ \frac{1}{2} \left[ J(x, p_{mc}(x, t)) \psi_{sp}(x, t) \right]^\dagger e^{i\Phi(x, \mathbf{R}, t)}
\]

\[
\times \psi_{sp}(x, t),
\]

or

\[
\dot{J}(x, t) = \frac{1}{2} \psi_{sp}^\dagger(x, t) \left[ J(x, e^{-i\Phi(x, \mathbf{R}, t)} p_{mc}(x, t)) \psi_{sp}(x, t) \right] e^{i\Phi(x, \mathbf{R}, t)}
\]

\[
+ \frac{1}{2} \left[ J(x, e^{-i\Phi(x, \mathbf{R}, t)} p_{mc}(x, t)) \psi_{sp}(x, t) \right]^\dagger e^{i\Phi(x, \mathbf{R}, t)}
\]

\[
\times \psi_{sp}(x, t),
\]
which can be written as
\[
\dot{j}(x, t) = \frac{1}{2} \psi_{sp}^\dagger(x, t) \left[ J(x, p(x, R; t)) \psi_{sp}(x, t) \right] + \frac{1}{2} \left[ J(x, p(x, R; t)) \psi_{sp}(x, t) \right]^\dagger \psi_{sp}(x, t).
\] (C6)

For an isolated atom we can define polarization and magnetization operators as
\[
\hat{p}(x, t) \equiv \int s(x; w, R)\hat{p}(w, t)dw, \quad \text{and} \quad \hat{\rho}(x, t) \equiv \int \alpha(x; w, R)\hat{\rho}(w, t)dw.
\] (C7)

Then from the properties of the relators we find immediately that
\[
\begin{align*}
\dot{\rho}(x, t) &= -\nabla \cdot \hat{p}(x, t) + Q\delta(x - R), \quad \text{(C8)} \\
\dot{j}(x, t) &= \frac{\partial \hat{p}(x, t)}{\partial t} + c\nabla \times \hat{\rho}(x, t) \\
&\quad - \int s(x; y, R)\hat{K}(y, t)dy,
\end{align*}
\] where
\[
Q = \int \dot{\rho}(x, t)dx,
\]
\[
\hat{K}(x, t) = \frac{\partial \hat{\rho}(x, t)}{\partial t} + \nabla \cdot \hat{j}(x, t).
\]

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 \(R\). 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 \(\hat{K}(x, t) = 0\). Hence we can write (C8) as
\[
\begin{align*}
\dot{\rho}(x, t) &= -\nabla \cdot \hat{p}(x, t) + Q\delta(x - R), \quad \text{(C9)} \\
\dot{j}(x, t) &= \frac{\partial \hat{p}(x, t)}{\partial t} + c\nabla \times \hat{\rho}(x, t),
\end{align*}
\] the standard form. There is a great advantage of determining \(\dot{\rho}(x, t)\) and \(\dot{j}(x, t)\) (or their expectation values) by first determining \(\hat{p}(x, t)\) and \(\hat{\rho}(x, t)\) (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
\[
\begin{align*}
p_R(x, t) &\equiv \int s(x; w, R)\langle \hat{p}(w, t) \rangle dw, \\
m_R^j(x, t) &\equiv \frac{1}{e} \int \alpha^j(x; w, R)\langle \hat{j}^k(w, t) \rangle dw,
\end{align*}
\] and then from (C9) we can write
\[
\langle \dot{\rho}(x, t) \rangle = -\nabla \cdot p_R(x, t) + Q\delta(x - R),
\] (C10)
\[
\langle \dot{j}(x, t) \rangle = \frac{\partial p_R(x, t)}{\partial t} + c\nabla \times m_R(x, t).
\]

Introducing a set of basis function \(\{W_{\alpha R}(x)\}\), where \(\alpha\) varies but \(R\) is fixed,
\[
\psi_{sp}(x, t) = \sum_\alpha c_\alpha(t)W_{\alpha R}(x),
\] where
\[
\{c_\alpha(t), c_\beta(t)\} = 0, \\
\{c_\alpha(t), \bar{c}_\beta(t)\} = \delta_{\alpha\beta},
\] we can write
\[
\begin{align*}
\rho_R(x, t) &= \sum_{\alpha, \beta} \rho_{\alpha\beta}(x, R)\eta_{\alpha\beta}(t), \\
j_R(x, t) &= \sum_{\alpha, \beta} j_{\alpha\beta}(x, R; t)\eta_{\alpha\beta}(t),
\end{align*}
\] with \(\rho_{\alpha\beta}(x, R)\) and \(j_{\alpha\beta}(x, R; t)\) given by (D4), and
\[
\eta_{\alpha\beta}(t) \equiv \langle c_\beta^\dagger(t)c_\alpha(t) \rangle.
\]

From the evolution equation (C3) we find that the dynamical of \(\eta_{\alpha\beta}(t)\) are given by
\[
\begin{align*}
\frac{i\hbar}{\partial t} \eta_{\alpha\beta}(t) &= \sum_\lambda \left( \tilde{H}_{\alpha\lambda}(R; t)\eta_{\lambda\beta}(t) - \eta_{\alpha\lambda}(t)\tilde{H}_{\lambda\beta}(R; t) \right),
\end{align*}
\] (C12)

where
\[
\tilde{H}_{\alpha\lambda}(R; t) = \frac{1}{2} \int W_{\alpha R}^\ast(x)H_0(x, p(x, R; t))W_{\lambda R}(x)dx \\
&+ \frac{1}{2} \int \left( H_0(x, p(x, R; t))W_{\alpha R}(x) \right)^\ast W_{\lambda R}(x)dx \\
&- e \int W_{\alpha R}(x)\Omega_{\lambda R}(x, t)W_{\lambda R}(x)dx
\] (C13)

Appendix D: The “isolated atom limit” of a crystal

The isolated atom limit of our equations in Section II is identified by the assumption that \(W_{\alpha R}(x)\) and \(W_{\beta R'}(x)\) have no common support if \(R \neq R'\). In this limit it follows from the definition (24) that the set of functions \(\{W_{\alpha R}(x, t)\}\) are mutually orthogonal, so \(W_{\alpha R}(x, t) \rightarrow W'_{\alpha R}(x, t)\) and, from (24,27), we see \(\chi_{\alpha R}(x, t) \rightarrow W_{\alpha R}(x)\). Then from (35) we have
\[
\tilde{H}_{\alpha R;\beta R'}(t) \rightarrow \delta_{RR'}\tilde{H}_{\alpha\lambda}(R; t),
\] (D1)
with $\tilde{H}_{\alpha\lambda}(R; t)$ is given by (C13). Assuming no initial correlation between the electronic motion in the individual atoms, from (33,36) we have

$$\eta_{\alpha R; R'}(t) \rightarrow \delta_{RR'}\eta_{\alpha\beta}(R; t)$$  \hspace{1cm} (D2)

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

$$i\hbar \frac{\partial \eta_{\alpha\beta}(R; t)}{\partial t} = \sum_{\lambda} \left( \tilde{H}_{\alpha\lambda}(R; t)\eta_{\lambda\beta}(R; t) - \eta_{\alpha\lambda}(R; t)\tilde{H}_{\lambda\beta}(R; t) \right).$$  \hspace{1cm} (D3)

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

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

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

$$\rho_{\beta\alpha}(x, R) = e W_{\beta R}^*(x) W_{\alpha R}(x),$$  \hspace{1cm} (D4)

$$j_{\beta\alpha}(x, R; t) = \frac{1}{2} W_{\beta R}^*(x) \left( J(x,p(x, R; t)) W_{\alpha R}(x) \right) + \frac{1}{2} \left( J'(x,p(x, R; t)) W_{\beta R}^*(x) \right) W_{\alpha R}(x),$$

so (44) become

$$\rho_R(x, t) \rightarrow \sum_{\alpha,\beta} \rho_{\beta\alpha}(x, R) \eta_{\alpha\beta}(R; t)$$ \hspace{1cm} (D5)

$$= -ie \left[ G_R(x, y; t) \right]_{y \rightarrow x},$$

$$j_R(x, t) \rightarrow \sum_{\alpha,\beta} j_{\beta\alpha}(x, R; t) \eta_{\alpha\beta}(R; t)$$

$$= -ie \left[ J_R(x, y; t) G_R(x, y; t) \right]_{y \rightarrow x}.$$

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

$$Q_R = \int \rho_R(x, t) dx = e \sum_{\alpha} \eta_{\alpha\alpha}(R; t),$$

and from (52,53) we have

$$\rho_R(x) = \sum_R Q_R \delta(x - R),$$  \hspace{1cm} (D6)

$$j_R(x, t) = 0,$$

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 $K_R(x, t) = 0$ for all $R$, since at any given $x$ at most one $K_R(x, t)$ can contribute to the sum (47); together with the second of (D6), this guarantees that $\tilde{j}(x, t) = 0$ (see (59)). With that, and the use of (D5), our general expressions (72,73) reduce to

$$p_R(x, t) = \sum_{\alpha,\beta, R', R''} \left[ \int s(x; y, R) \rho_{\beta\alpha}(y, R; t) dy \right] \eta_{\alpha\beta}(R; t)$$  \hspace{1cm} (D7)

and

$$m_R^j(x, t) = \frac{1}{c} \sum_{\alpha,\beta, R', R''} \left[ \int \alpha^k(x; y, R) j_{\beta\alpha}^k(y, R; t) dy \right] \eta_{\alpha\beta}(R; t).$$  \hspace{1cm} (D8)

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

$$p(x, t) = \sum_R p_R(x, t),$$

$$m(x, t) = \sum_R m_R(x, t),$$

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

$$\langle \dot{p}(x, t) \rangle = \sum_R \left( -\nabla \cdot p_R(x, t) + Q_R \delta(x - R) \right),$$  \hspace{1cm} (D9)

$$\langle \dot{j}(x, t) \rangle = \sum_R \left( \frac{\partial p_R(x, t)}{\partial t} + \nabla \times m_R(x, t) \right),$$

with of course the dynamics of $p_R(x, t)$ and $m_R(x, t)$ given by the use of the solution of (D3) in (D7,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 $K_R(x, t) = 0$, 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 Section 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.
As a trivial example, consider another.

For a review and references to original work see C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Photons and Atoms. Introduction to Quantum Electrodynamics*, John Wiley and Sons, Inc., 1989.

ForareviewandreferencestooriginalworkseeC.Cohen-
H.A.Lorentz.*The Theory of Electrons*. Columbia University Press, 1909.

L. Rosenfeld. *Theory of Electrons*. North-Holland Publishing Company, 1951.

A.D. Fokker. *Phil. Mag.*, 404, 1920.

R. Zeyher. H. Bliz. M. Cardona. *Solid State Communications*, 19:57–60, 1976.

J. E. Sipe. Ed Ghahramani. *Phys. Rev. B*, 93, 2016.

J. E. Sipe. *Phil. Mag.*, 80:197202, 2007.

E.I. Blount. *Phil. Mag.*, 322:835, 2013.

Sang Wook Kim, Kangjun Seo, and Bruno Uchoa. *Phys. Rev. B*, 97, 2018.

See, e.g., Michael E. Peskin and Daniel V. Schroeder, *An Introduction to quantum field theory*, Addison-Wesley, Reading, USA, 1995.

Sylvia D. Swiecicki. J.E. Sipe. *Annals of Physics*, 338:260–277, 2013.

Sylvia D. Swiecicki. J.E. Sipe. *Phys. Rev. B*, 90:125115, 2014.

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

This extends earlier work [30, 31, 39].

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

This is a further development of a strategy introduced earlier [31].

Rodrigo A. Muniz, J.L. Cheng, and J.E. Sipe, in preparation.

In 2D this is proportional to the net Chern number of the filled bands.

Di Xiao. Ming-Che Chang. Qian Niu. *Reviews of Modern Physics*, 82:1959, 2010.

M. Levanda. V. Fleurov. *J. Phys.: Condens. Matter*, 6:7889, 1994.

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

This extends earlier work [30, 31, 39].

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

This is a further development of a strategy introduced earlier [31].

Rodrigo A. Muniz, J.L. Cheng, and J.E. Sipe, in preparation.

In 2D this is proportional to the net Chern number of the filled bands.

Di Xiao. Ming-Che Chang. Qian Niu. *Reviews of Modern Physics*, 82:1959, 2010.

M. Levanda. V. Fleurov. *J. Phys.: Condens. Matter*, 6:7889, 1994.

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

This extends earlier work [30, 31, 39].

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

This is a further development of a strategy introduced earlier [31].

Rodrigo A. Muniz, J.L. Cheng, and J.E. Sipe, in preparation.

In 2D this is proportional to the net Chern number of the filled bands.

Di Xiao. Ming-Che Chang. Qian Niu. *Reviews of Modern Physics*, 82:1959, 2010.

M. Levanda. V. Fleurov. *J. Phys.: Condens. Matter*, 6:7889, 1994.