Polarization and localization in insulators: Generating function approach

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We develop the theory and practical expressions for the full quantum-mechanical distribution of the intrinsic macroscopic polarization of an insulator in terms of the ground state wave function. The central quantity is a cumulant generating function, which yields, upon successive differentiation, all the cumulants and moments of the probability distribution of the center of mass $\mathbf{X}/N$ of the electrons, defined appropriately to remain valid for extended systems obeying twisted boundary conditions. The first moment is the average polarization, where we recover the well-known Berry phase expression. The second cumulant gives the mean-square fluctuation of the polarization, which defines an electronic localization length ξ_i along each direction i: $\xi_i^2 = (\langle X_i^2 \rangle - \langle X_i \rangle^2)/N$. It follows from the fluctuation-dissipation theorem that in the thermodynamic limit ξ_i diverges for metals and is a finite, measurable quantity for insulators. In noninteracting systems ξ_i^2 is related to the spread of the Wannier functions. It is possible to define for correlated insulators maximally localized ''many-body Wannier functions,'' which for large N become localized in disconnected regions of the high-dimensional configuration space, establishing a direct connection with Kohn's theory of the insulating state. Interestingly, the expression for ξ_i^2, which involves the second derivative of the wave function with respect to the boundary conditions, is directly analogous to Kohn's formula for the ''Drude weight'' as the second derivative of the energy.

I. INTRODUCTION

An insulator is distinguished from a conductor at zero temperature by its vanishing dc conductivity and its ability to sustain a macroscopic polarization $\mathbf{P}_{mac}$, both with and without an applied electric field.[1,2] In the classical theory of electromagnetism in materials this distinction is often cast in terms of the difference between ''free charges'' that carry the dc current in a conductor and polarizable ''bound charges'' in an insulator.[2,3] Such a description conflicts with the fact that, even in highly ionic solids, the electrons are not well localized near the ions, and there is appreciable interpenetration between the ionic charge densities.[4–7] The inadequacy of such a textbook picture is particularly striking in the case of covalent insulators, whose charge density is delocalized, as in metals.[3] Therefore the qualitative difference between metals and insulators is not apparent from inspection of the charge distribution, and the correct notion of electronic localization in insulators versus delocalization in conductors must be sought elsewhere.

As shown by Kohn,[8,9] localization is a property of the many-electron wave function: insulating behavior arises whenever the ground state wave function of an extended system breaks up into a sum of functions $\Psi_{\mathbf{M}}$, which are localized in essentially disconnected regions $\mathcal{R}_{\mathbf{M}}$ of the high-dimensional configuration space. When using periodic boundary conditions on a supercell containing N electrons, an insulating wave function can be written as

$$\Psi(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \sum_{\mathbf{M}=-\infty}^{+\infty} \Psi_{\mathbf{M}}(\mathbf{x}_1, \ldots, \mathbf{x}_N), \qquad (1)$$

where for a large supercell $\Psi_{\mathbf{M}}$ and $\Psi_{\mathbf{M}'}$ have an exponentially small overlap for $\mathbf{M}' \neq \mathbf{M}$. Hence, electronic localization in insulators does not occur in real space (charge density) but in configuration space (wave function). Kohn argued that such disconnectedness is in fact the signature of an insulating wave function.

The other aspect of insulators, macroscopic polarization, is a subject about which there has been much debate. In a confined system with open boundary conditions, it is simply the dipole moment of the charge distribution divided by the volume.[1,2] In the case of an extended system, such as a periodic crystalline solid, the situation is far less clear: although there exist well-known expressions for dielectric response functions,[10,11] the very definition of macroscopic polarization as a bulk property independent of surface termination remained controversial for a long time. Only in recent years has a theory of polarization emerged—the so-called Berry phase formulation[12–17]—for the average $\mathbf{P}_{mac}$ in terms of the bulk ground state wave function of an insulating crystal. This theory shows that in general the information about the macroscopic polarization of an extended insulating system is not in the charge density, but in the wave function. This important finding is consistent with the well-known fact that the dipole moment of a periodic, continuous charge distribution is ill defined,[18,19] since the expressions for the first moment of the charge distribution are valid for confined systems but do not have a well-defined thermodynamic limit independent of the surface. The only instance where a meaningful dipole moment per unit volume can be assigned to an extended crystal is when the charge distribution in the unit cell can be resolved into contributions that are localized in nonoverlapping regions and can be ascribed[3] to identifiable atoms (ions, molecules) (the so-called ''Clausius-Mossotti limit''). However, such a limit is rather unrealistic for most insulators, with the possible exception of some organic and molecular crystals.

In this paper we present a comprehensive theory of electronic polarization and localization in bulk insulators that generalizes the Berry phase theory and merges with Kohn's

theory of localization in the insulating state. By introducing the generating function formalism, we show the rigorous connections between polarization and localization, establish relations to experimentally measureable quantities, and provide formulas for practical calculations.

The paper is organized as follows: in Sec. II we summarize some of the recent developments in the field, pointing to some of the open issues that will be addressed in this work, and collect relevant equations for later reference. In Sec. III is introduced the central concept upon which the present formulation is based, the generating function applied to the quantum probability distribution of the macroscopic polarization. The simple case of a confined system is discussed first, and then we show how to modify the expressions to deal with extended systems. In Sec. IV we establish the fluctuation-dissipation relation between the quadratic quantum fluctuations of the polarization and the absorptive part of the conductivity for an extended system. In Sec. V we discuss the qualitative differences between the quadratic fluctuations in insulators and in conductors in terms of a properly defined localization length; for insulators we derive an inequality involving the localization length and the optical gap. In Sec. VI we introduce ''many-body Wannier functions'' and establish the connection between the present formalism and Kohn's theory of localization in the insulating state. Discretized formulas for the polarization and localization length that can be used in numerical many-body calculations are derived in Sec. VII. A discussion of our results is presented in Sec. VIII.

II. OVERVIEW OF RECENT DEVELOPMENTS

A meaningful definition of average macroscopic polarization that is generally applicable—to both confined and extended insulating systems—and relates closely to the way polarization is experimentally measured can be obtained by taking as the more basic concept the *change* in polarization induced by a slow change in some parameter λ in the Hamiltonian.[20,21] The resulting expressions are in terms of the derivative $\partial \mathbf{P}_{\text{mac}}^{(\lambda)}/\partial\lambda$, and Resta[21] proposed to calculate the finite change in bulk polarization as

$$\Delta\mathbf{P}_{\text{mac}} = \int_0^1 d\lambda \frac{\partial\mathbf{P}_{\text{mac}}^{(\lambda)}}{\partial\lambda}. \tag{2}$$

Specializing to the case of extended systems and considering the variation in λ as an adiabatic time evolution, $\partial\mathbf{P}_{\text{mac}}^{(\lambda)}/\partial\lambda$ is the spatially averaged adiabatic current flowing through the bulk; thus this equation gives $\Delta\mathbf{P}_{\text{mac}}$ as an integrated bulk current. According to classical electrodynamics,[1,2] this adiabatic polarization current is

$$\frac{\partial\mathbf{P}_{\text{mac}}^{(\lambda)}}{\partial\lambda} = \frac{1}{V}\int_V d\mathbf{r}\, \mathbf{j}_{\text{bulk}}^{(\lambda)} = \frac{1}{V}\mathbf{J}^{(\lambda)}, \tag{3}$$

where V is the volume of the system and $\mathbf{j}_{\text{bulk}}^{(\lambda)}$ is the current density in the bulk, where λ plays the role of time in the usual expression for the current (for a derivation see, for example, Ref. 14). $\mathbf{J}^{(\lambda)}$ can be expressed as the adiabatic limit of a Kubo formula for the current.[21,22] In its usual form the Kubo formula involves a summation over all eigenstates

(ground state and excited states), which not only makes it impractical for actual calculations, but also fails to reflect the fact that electric polarization is a ground state property.

In the Berry phase theory of polarization,[12–16] the Kubo formula is recast in a form that only depends on the ground state wave function; in order to arrive at such an expression, it is convenient to impose *twisted boundary conditions* over the volume V on the many-body wave function, which becomes labeled by $\mathbf{k}$:

$$\Psi_{\mathbf{k}}^{(\lambda)}(\mathbf{x}_1, \ldots, \mathbf{x}_i + \mathbf{L}, \ldots, \mathbf{x}_N)$$
$$= e^{\imath\mathbf{k}\cdot\mathbf{L}}\Psi_{\mathbf{k}}^{(\lambda)}(\mathbf{x}_1, \ldots, \mathbf{x}_i, \ldots, \mathbf{x}_N), \tag{4}$$

where N is the number of electrons in the system, $\mathbf{L} = (L_1, 0, 0)$, for example, and $-\pi/L_i \leq k_i \leq \pi/L_i$ (in the case of an orthorhombic supercell). It is useful to introduce the following wave function:

$$|\Phi_{\mathbf{k}}^{(\lambda)}\rangle = e^{-\imath\mathbf{k}\cdot\hat{\mathbf{X}}}|\Psi_{\mathbf{k}}^{(\lambda)}\rangle, \tag{5}$$

where $\hat{\mathbf{X}} = \Sigma_{i=1}^N \hat{\mathbf{x}}_i$, so that $\hat{\mathbf{X}}/N$ is the position operator for the center of mass of the N electrons in the volume V. $\Phi_{\mathbf{k}}^{(\lambda)}$ can be regarded as the many-body analog of the cell-periodic part of the Bloch function in the single-electron case. It obeys periodic boundary conditions, and the $\mathbf{k}$ dependence is transferred from the boundary conditions to the Hamiltonian: if $\Psi_{\mathbf{k}}^{(\lambda)}$ is the ground state of the Hamiltonian $\hat{H}^{(\lambda)}$, then $\Phi_{\mathbf{k}}^{(\lambda)}$ is the ground state of the Hamiltonian

$$\hat{H}^{(\lambda)}(\mathbf{k}) = e^{-\imath\mathbf{k}\cdot\hat{\mathbf{X}}}\hat{H}^{(\lambda)}e^{\imath\mathbf{k}\cdot\hat{\mathbf{X}}}, \tag{6}$$

which, for a nonrelativistic Hamiltonian without spin-orbit coupling, can be obtained from $\hat{H}^{(\lambda)}$ by performing the gauge transformation $\hat{\mathbf{p}}_i \rightarrow \hat{\mathbf{p}}_i + \hbar\mathbf{k}$ on the momentum operator of each particle (for the single-particle analog, see Ref. 23.) Using the function $\Phi_{\mathbf{k}}^{(\lambda)}$, the Kubo formula for the adiabatic electronic current $\mathbf{J}_{\mathbf{k}}^{(\lambda)}$ for a particular choice of twisted boundary conditions can be expressed as[16,24,25]

$$\mathbf{J}_{\mathbf{k}}^{(\lambda)} = 2q_e \,\text{Im}\langle\partial_{\mathbf{k}}\Phi_{\mathbf{k}}^{(\lambda)}|\partial_\lambda\Phi_{\mathbf{k}}^{(\lambda)}\rangle, \tag{7}$$

which indeed only depends on the bulk ground state wave function. Substituting this expression into Eq. (3) and using Eq. (2), it can be shown that the net change in electronic polarization along the path parametrized by λ is

$$(\Delta P_{\text{el}})_i = \frac{\imath q_e}{(2\pi)^3}\int d\mathbf{k}\int_0^1 d\lambda[\langle\partial_\lambda\Phi_{\mathbf{k}}^{(\lambda)}|\partial_{k_i}\Phi_{\mathbf{k}}^{(\lambda)}\rangle$$
$$- \langle\partial_{k_i}\Phi_{\mathbf{k}}^{(\lambda)}|\partial_\lambda\Phi_{\mathbf{k}}^{(\lambda)}\rangle], \tag{8}$$

where the integral in $\mathbf{k}$ is over all twisted boundary conditions.

As expected for a measurable quantity, the above expression, as well as Eq. (7), are invariant under gauge transformations of the form

$$\Psi_{\mathbf{k}}^{(\lambda)} \rightarrow e^{\imath\varphi(\mathbf{k},\lambda)}\Psi_{\mathbf{k}}^{(\lambda)}, \tag{9}$$

where $\varphi(\mathbf{k},\lambda)$ is a smooth, real function. The ground state wave functions Ψ at $\mathbf{k}$ and $\mathbf{k}+\mathbf{G}$, where $\mathbf{G}$ is a basis vector of the reciprocal lattice of the cell of volume V, can differ at

most by an overall phase factor, since the boundary conditions are the same and the ground state is assumed to be nondegenerate:[25]

$$\Psi_{\mathbf{k}}^{(\lambda)} = e^{\iota\Theta(\mathbf{k},\lambda;\mathbf{G})}\Psi_{\mathbf{k}+\mathbf{G}}^{(\lambda)}. \tag{10}$$

If $\Theta(\mathbf{k},\lambda;\mathbf{G})$, which is at our disposal, is chosen to be independent of λ, then it can be shown that the net change in polarization becomes simply

$$\Delta\mathbf{P}_{\mathrm{el}} = \mathbf{P}_{\mathrm{el}}^{(1)} - \mathbf{P}_{\mathrm{el}}^{(0)}, \tag{11}$$

where

$$\mathbf{P}_{\mathrm{el}}^{(\lambda)} = \frac{\iota q_e}{(2\pi)^3}\int d\mathbf{k}\langle\Phi_{\mathbf{k}}^{(\lambda)}|\partial_{\mathbf{k}}\Phi_{\mathbf{k}}^{(\lambda)}\rangle, \tag{12}$$

and again the integral in $\mathbf{k}$ is over all twisted boundary conditions. Equations (8), (11), and (12) form the central result of Ref. 14, which gives the many-body generalization of the Berry phase theory of polarization, originally formulated by King-Smith and Vanderbilt for independent electrons.[12,13] In the derivation it is assumed that the ground state is isolated from the excited states by a finite energy gap, and that there are no long-range correlations.[25] In the independent-electron theory the Berry phase formula in terms of Bloch functions can be recast in terms of localized Wannier functions,[26] yielding the intuitive result that the electronic polarization is given by the sum of the centers of charge of the occupied Wannier functions.[12,13] Unfortunately, Wannier functions are only defined in a one-electron framework. In Sec. VI B we will introduce functions that are in some sense the many-body counterpart of Wannier functions (we will term them ''many-body Wannier functions''), and can be chosen to be localized in configuration space. In terms of those functions, a localized description of polarization can be presented for the correlated case. Moreover, they establish the link between the Berry phase theory of polarization and Kohn's theory of localization.[8,9]

The following comments should be made about the equations above: unlike Eq. (8), which involves an integral over λ, Eqs. (11) and (12) only depend on the endpoints, $\lambda=0$ and $\lambda=1$. The tradeoff is that whereas the former gives the exact change in polarization along the path, the latter give it only modulo a ''quantum''[12–14] (this ''quantum of polarization'' is discussed in Appendix C). A related aspect is the behavior of the equations under gauge transformations: unlike Eqs. (7) and (8), Eqs. (11) and (12) are not completely gauge invariant. As mentioned previously, they were obtained by assuming $\partial_\lambda\Theta(\mathbf{k},\lambda;\mathbf{G})=0$, and therefore the resulting $\Delta\mathbf{P}_{\mathrm{el}}$ is only invariant (modulo the quantum) under transformations that preserve the condition $\Theta(\mathbf{k},\lambda=1;\mathbf{G}) = \Theta(\mathbf{k},\lambda=0;\mathbf{G})$. Moreover, in order to be able to interpret Eq. (12) at a *single* λ as the electronic polarization,[13,27] one has to impose the stronger condition $\Theta(\mathbf{k},\lambda;\mathbf{G})\equiv0$, or $\Psi_{\mathbf{k}+\mathbf{G}}^{(\lambda)}=\Psi_{\mathbf{k}}^{(\lambda)}$. Gauges that obey this condition are known as ''periodic gauges.''[12]

It has been pointed out by Aligia[28] that this analysis needs to be modified in cases where there is a fractional number of electrons per primitive cell. The idea of a ''periodic gauge'' needs to be extended to relate wave functions separated by multiples of the smallest $\mathbf{G}$, and the integral in Eq. (12)

needs to be extended accordingly. For the sake of simplicity, in the main text we will assume integer filling, and in Appendix D we indicate how to modify the formulas in order to deal with fractional filling.

We note that in its present form the theory of bulk polarization only gives the *average* of the quantum distribution of $\mathbf{P}_{\mathrm{mac}}$. In Secs. III C and III D we will extended it to deal with the full distribution. In particular, its quadratic spread will turn out to be a very relevant quantity, since it is intimately related to electronic localization. To our knowledge this was first discussed by Kudinov,[29] who proposed to measure the degree of electronic localization in insulators precisely in terms of the mean-square quantum fluctuation of the ground state polarization. Kudinov proposed a criterion to differentiate between insulators and conductors based on the scaling with sample volume V of the mean-square quantum fluctuation of the net dipole moment, $\langle\Delta\hat{\mathbf{d}}^2\rangle = \langle\hat{d}^2\rangle - \langle\hat{\mathbf{d}}\rangle^2$; here $\hat{\mathbf{d}}$ is the dipole moment operator, $\langle\cdots\rangle$ means the expectation value over the ground state, and $\Delta\hat{\mathbf{d}} = \hat{\mathbf{d}} - \langle\hat{\mathbf{d}}\rangle$. Using the fluctuation-dissipation theorem,[30–32] Kudinov related this quantity to the optical conductivity:

$$\frac{\langle\Delta\hat{d}_i^2\rangle}{V} = \frac{\hbar}{\pi}\int_0^\infty d\omega\,\mathrm{Im}\chi_{ii}(\omega), \tag{13}$$

where $\chi(\omega)$ is the electric susceptibility tensor and $i=x,y,z$. Using Eq. (13), Kudinov showed that as $V\to\infty$, $\langle\Delta\hat{\mathbf{d}}^2\rangle/V$ remains finite in insulators, whereas it diverges in conductors. Only confined systems with open boundary conditions were considered, and thus the issue of how to deal with $\mathbf{P}_{\mathrm{mac}}$ in extended systems was not addressed. Our formulation leads to similar expressions, but with carefully defined bulk quantities that have a well-defined thermodynamic limit.

The ideas from the Berry phase theory of polarization have recently been extended in order to address the problem of localization. This effort was initiated in Ref. 33, where non-interacting electron systems with a band gap were considered. For such systems it is natural to attempt to quantify the degree of localization of the electrons in terms of the spread of the occupied Wannier functions. Marzari and Vanderbilt proposed to measure that spread via the quantity

$$\Omega = \sum_{n=1}^{M} [\langle r^2\rangle_n - \langle\mathbf{r}\rangle_n^2], \tag{14}$$

where $\langle\cdots\rangle_n$ means the expectation value over the nth occupied Wannier function in the unit cell (whose total number M equals the number of filled bands.) Since the electronic polarization is given by the sum of the centers of charge of the occupied Wannier functions,[12,13] this expression is very appealing in its interpretation as the spread of the charge distribution of the Wannier functions. It should be noted however, that unlike the sum of the centers of charge, the sum of the quadratic spreads is not invariant under gauge transformations of the Wannier functions,[33] and so Ω cannot be used directly as a measure of any physical quantity. Nevertheless, Marzari and Vanderbilt were able to decompose it into a sum of two positive terms: a gauge-invariant part, Ω_{I},

plus a gauge-dependent term, which they minimized to obtain maximally localized Wannier functions.

We will show that Ω_I gives the mean-square fluctuation of the bulk polarization, thus obeying a relation analogous to Eq. (13) (see Appendix A). Moreover, in the same way that Ω measures the spread of the Wannier functions, for independent electrons Ω_I measures the spread of Kohn's functions Ψ_M,[8,9] which can be interpreted as maximally localized many-body Wannier functions; this is discussed in Sec. VI B and Appendix B.

In Ref. 33 it was shown that Ω_I be rewritten as

$$\Omega_I = \frac{v}{(2\pi)^3} \int_{BZ} d\mathbf{k} \, \mathrm{Tr} \, g(\mathbf{k}), \qquad (15)$$

where Tr denotes the trace, v is the volume of the unit cell, the integral is over the Brillouin zone, and $g(\mathbf{k})$ is the tensor

$$g_{ij}(\mathbf{k}) = \mathrm{Re} \sum_{n=1}^{M} \langle \partial_{k_i} u_{n\mathbf{k}} | \partial_{k_j} u_{m\mathbf{k}} \rangle$$
$$- \sum_{n=1}^{M} \sum_{m=1}^{M} \langle \partial_{k_i} u_{n\mathbf{k}} | u_{m\mathbf{k}} \rangle \langle u_{m\mathbf{k}} | \partial_{k_j} u_{n\mathbf{k}} \rangle, \quad (16)$$

where $u_{n\mathbf{k}}$ is the cell-periodic part of the Bloch function. This tensor is a metric that can be used to determine the ''quantum distance'' along a given path in in $\mathbf{k}$ space.[33] In Sec. III D we generalize this tensor to the many-body case, and in Sec. IV we relate it to the measurable polarization fluctuations via the fluctuation-dissipation relation.

All of the above expressions for the polarization and localization involve integrals over $\mathbf{k}$. More recently, alternative expressions have been proposed which use only periodic boundary conditions ($\mathbf{k}=0$).[16,17,34] These are sometimes called ''single-point'' formulas. The basic quantity in this formulation is, in one dimension,

$$z_N = \langle \Psi_{\mathbf{k}=0} | e^{i(2\pi/L)\hat{X}} | \Psi_{\mathbf{k}=0} \rangle, \qquad (17)$$

where $\Psi_{\mathbf{k}=0}$ is the ground state many-body wave function obeying periodic boundary conditions over a cell of length L with N electrons, and as before $\hat{X} = \sum_{i=1}^{N} \hat{x}_i$. Resta[17] showed that in the thermodynamic limit the electronic polarization is given by

$$P_{el} = \lim_{N \to \infty} \frac{q_e}{2\pi} \mathrm{Im} \ln z_N. \qquad (18)$$

However, the nature of the approximations involved at finite L, and the precise relation between Eqs. (12) and (18) as a function of the size of the system, were not clarified; this is a matter of crucial importance for the usefulness of the expressions in practical calculations, and is discussed in Sec. VII C.

Resta and Sorella[34] proposed to measure the electronic localization length in one-dimensional (1D) insulators as[35]

$$\xi = \sqrt{\mathcal{D}}/(2\pi n_0), \qquad (19)$$

where $n_0 = N/L$ and

$$\mathcal{D} = -\lim_{N \to \infty} N \ln|z_N|^2. \qquad (20)$$

They showed that if the electrons are uncorrelated, then for insulators ξ is simply related to the 1D version of Ω_I defined in Eq. (15) ($\xi^2 = \Omega_I/M$), and therefore it is finite, whereas for metals it diverges (even before taking the limit $N \to \infty$). Then they proposed that a similar behavior should occur when the electrons are correlated (with the difference that in general ξ diverges for correlated conductors only after taking the thermodynamic limit).

In the present work we generalize Eq. (19) to many dimensions (see Sec. VII B), and give an explicit many-body derivation that, similarly to Ω_I in the uncorrelated case, ξ^2 measures the polarization fluctuations in correlated extended systems. Similarly to the continuum formulas involving an average over twisted boundary conditions,[28] these formulas require modification when there is a noninteger number of electrons per cell[36] (see Appendix D).

III. GENERATING FUNCTION FORMALISM

A. Definitions

Generating functions play a central role in the theory of statistics,[37] and have been applied to many problems in physics.[38,39] Loosely speaking, a generating function of a distribution is some function that yields, upon successive differentiation, the moments of the distribution, or some combination thereof. Two kinds of generating functions will be of interest to us: the *characteristic function* $C_{\mathbf{X}}(\boldsymbol{\alpha})$, and its logarithm, the *cumulant generating function*. If $\mathbf{X}$ is a vector of d variables $X_1, \ldots, X_d$ with a normalized joint probability distribution function $p(X_1, \ldots, X_d) = p(\mathbf{X})$, the characteristic function is defined as

$$C_{\mathbf{X}}(\boldsymbol{\alpha}) = \int_{-\infty}^{+\infty} e^{-i\boldsymbol{\alpha} \cdot \mathbf{X}} p(\mathbf{X}) d\mathbf{X} \equiv \langle e^{-i\boldsymbol{\alpha} \cdot \mathbf{X}} \rangle, \qquad (21)$$

where $\boldsymbol{\alpha} \cdot \mathbf{X} = \sum_{i=1}^{d} \alpha_i X_i$.[40] The d-dimensional moments can be extracted directly from $C_{\mathbf{X}}(\boldsymbol{\alpha})$:

$$\langle X_1^{n_1} \cdots X_d^{n_d} \rangle = i^n \partial_{\alpha_1^{n_1} \cdots \alpha_d^{n_d}}^n C_{\mathbf{X}}(\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0}, \qquad (22)$$

where $n = \sum_{i=1}^{d} n_i$. The cumulants are obtained in a similar way from $\ln C_{\mathbf{X}}(\boldsymbol{\alpha})$:

$$\langle X_1^{n_1} \cdots X_d^{n_d} \rangle_c = i^n \partial_{\alpha_1^{n_1} \cdots \alpha_d^{n_d}}^n \ln C_{\mathbf{X}}(\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0}, \qquad (23)$$

where, following the notation of Ref. 38, $\langle \cdots \rangle_c$ denotes the *cumulant average*, which in general is different from the simple average $\langle \cdots \rangle$ associated with the moments:

$$\langle X_i \rangle_c = \langle X_i \rangle,$$

$$\langle X_i^2 \rangle_c = \langle X_i^2 \rangle - \langle X_i \rangle^2 = \langle \Delta X_i^2 \rangle,$$

$$\langle X_i X_j \rangle_c = \langle X_i X_j \rangle - \langle X_i \rangle \langle X_j \rangle. \qquad (24)$$

An important property of cumulants is that they can be explicitly represented solely in terms of the lower moments, and vice versa. More precisely—and this is very relevant for what follows—for $n>1$ they can be expressed in terms of the *central moments* $\langle \Delta X_1^{m_1} \Delta X_2^{m_2} \Delta X_3^{m_3} \rangle$, where $\Delta X_i = X_i - \langle X_i \rangle$ and $m_1 + m_2 + m_3 \leq n$, and thus they are independent

of the mean $\langle X_i \rangle$. Moreover, provided that the characteristic function exists, the set of all the moments or cumulants completely determines the distribution.

B. Polarization distribution in confined systems

Let us consider a neutral 3D system of finite volume V containing N electrons and N_n nuclei, on which we impose open boundary conditions. The dipole moment operator is $\hat{\mathbf{d}} = q_e \hat{\mathbf{X}} + q_n \hat{\mathbf{X}}_n$, where $\hat{\mathbf{X}} = \Sigma_{i=1}^{N} \hat{\mathbf{x}}_i$ and $\hat{\mathbf{X}}_n = \Sigma_{i=1}^{N_n} \hat{\mathbf{x}}_i^n$. The average dipole moment of the system is $\mathbf{d} = \langle \Psi | \hat{\mathbf{d}} | \Psi \rangle = \int \mathbf{r} \rho(\mathbf{r}) d\mathbf{r} = V \mathbf{P}_{mac}$. Since the center of mass $\mathbf{X}/N$ of the electrons is not perfectly localized, the ground state wave function Ψ is not an eigenstate of $\hat{\mathbf{X}}$. Therefore the Cartesian components of the dipole moment undergo quantum fluctuations, having a joint probability distribution $p(\mathbf{d})$ dictated by Ψ. For simplicity we will assume that the nuclei can be treated classically as ''clamped'' point charges; then they only contribute to the average of the distribution, and the quantum fluctuations come solely from the electrons. Hence, in what follows we will neglect the nuclear contribution, focusing on the distribution of the electronic center of mass. If Ψ is the many-electron wave function (parametrized by the nuclear coordinates) with normalization $\langle \Psi | \Psi \rangle = 1$, that distribution is given by

$$p(\mathbf{X}) = \langle \Psi | \delta(\hat{\mathbf{X}} - \mathbf{X}) | \Psi \rangle. \quad (25)$$

Similarly, for a given component, say, X_1, the distribution is

$$p(X_1) = \int_{-\infty}^{+\infty} p(\mathbf{X}) dX_2 dX_3 = \langle \Psi | \delta(\hat{X}_1 - X_1) | \Psi \rangle. \quad (26)$$

The characteristic function $C_{\mathbf{X}}(\boldsymbol{\alpha})$ is obtained by substituting Eq. (25) into Eq. (21):

$$C_{\mathbf{X}}(\boldsymbol{\alpha}) = \langle \Psi | e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | \Psi \rangle. \quad (27)$$

It is clear that if we define $\langle \hat{X}_1^{n_1} \hat{X}_2^{n_2} \hat{X}_3^{n_3} \rangle = \langle \Psi | \hat{X}_1^{n_1} \hat{X}_2^{n_2} \hat{X}_3^{n_3} | \Psi \rangle$ we find, using Eq. (22), $\langle X_1^{n_1} X_2^{n_2} X_3^{n_3} \rangle = \langle \hat{X}_1^{n_1} \hat{X}_2^{n_2} \hat{X}_3^{n_3} \rangle$.[40] The electronic polarization operator is $(\hat{P}_{el})_i = q_e \hat{X}_i / V$, and the moments of its distribution are given by

$$\langle (P_{el})_1^{n_1} (P_{el})_2^{n_2} (P_{el})_3^{n_3} \rangle = \left(\frac{q_e}{V} \right)^n \langle \hat{X}_1^{n_1} \hat{X}_2^{n_2} \hat{X}_3^{n_3} \rangle, \quad (28)$$

where $n = n_1 + n_2 + n_3$.

C. Polarization distribution in extended systems

In the case of a confined system the characteristic function $C_{\mathbf{X}}(\boldsymbol{\alpha})$ was introduced as a purely formal device for obtaining the moments of the distribution, since in practice it was completely equivalent to a direct evaluation of the moments. In the case of an extended system the situation is rather different since, as discussed in the Introduction, the very definition of polarization needs to be reexamined, and a naive generalization of the direct method of calculating the moments does not apply.

Again let us consider a finite system with N electrons in a volume V, but now with twisted boundary conditions [Eq. (4)], which eliminate the surface. The difficulties in defining the macroscopic bulk polarization can be seen from the fact that $\hat{X}_i^n$ is not a valid operator in the Hilbert space defined by Eq. (4): indeed, if $|\Psi_{\mathbf{k}}\rangle$ is a vector in that space, $\hat{X}_i^n | \Psi_{\mathbf{k}} \rangle$ is not (it is not even normalizable), and thus $\langle \hat{X}_i^n \rangle_{\mathbf{k}} = \langle \Psi_{\mathbf{k}} | \hat{X}_i^n | \Psi_{\mathbf{k}} \rangle$ is ill defined. We will now show that although the *operator* $\hat{\mathbf{X}}$ is ill-defined, one can nevertheless define a meaningful joint probability *distribution* $p(\mathbf{X})$ for the electronic center of mass in an extended insulating system. This distribution will have the same physical interpretation as Eq. (25) for a confined system, i.e., the moments of the variables $(P_{el})_i = q_e X_i / V$ are the moments of the distribution of the electronic polarization:

$$\langle (P_{el})_1^{n_1} (P_{el})_2^{n_2} (P_{el})_3^{n_3} \rangle = \left(\frac{q_e}{V} \right)^n \langle X_1^{n_1} X_2^{n_2} X_3^{n_3} \rangle. \quad (29)$$

Our main interest will be in the average $\langle X_i \rangle$ and in the quadratic spread $\langle X_i^2 \rangle_c$ which, according to Ref. 29, measures the electronic localization. More precisely, we will define the localization length $\xi_i(N)$ along the ith direction as

$$\xi_i^2(N) = \frac{1}{N} \langle X_i^2 \rangle_c = \frac{V^2}{q_2^2 N} \langle (\Delta P_{mac})_i^2 \rangle, \quad (30a)$$

and show that for insulators it has a well-defined thermodynamic limit

$$\xi_i^2 = \lim_{N \to \infty} \xi_i^2(N). \quad (30b)$$

The desired distribution $p(\mathbf{X})$ can be obtained via a proper modification of the characteristic function $C_{\mathbf{X}}(\boldsymbol{\alpha})$, defined in Eq. (27) for a confined system. We start by introducing the quantity $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$, which is a *precursor* to the characteristic function for the extended system:

$$\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) = \langle \Psi_{\mathbf{k}} | e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | \Psi_{\mathbf{k}+\boldsymbol{\alpha}} \rangle = \langle \Phi_{\mathbf{k}} | \Phi_{\mathbf{k}+\boldsymbol{\alpha}} \rangle, \quad (31)$$

where in the last equality we used Eq. (5). In the first form the similarity to Eq. (27) is evident;[41] notice, however, that this is not an expectation value over a single ground state $\Psi_{\mathbf{k}}$, since the boundary conditions on the *bra* ($\mathbf{k}$) and on the *ket* ($\mathbf{k}+\boldsymbol{\alpha}$) are different. That is required in order to compensate for the shift by $-\boldsymbol{\alpha}$ in the boundary condition on the *ket* caused by the operator $e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}}$; in this way the states $|\Psi_{\mathbf{k}}\rangle$ and $|\bar{\Psi}_{\mathbf{k}}(\boldsymbol{\alpha})\rangle = e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} |\Psi_{\mathbf{k}+\boldsymbol{\alpha}}\rangle$ obey the same boundary conditions even for $\alpha_i \neq 2\pi n / L_i$, so that their dotproduct $\langle \Psi_{\mathbf{k}} | \bar{\Psi}_{\mathbf{k}}(\boldsymbol{\alpha}) \rangle$ does not vanish and $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ can be chosen to be a differentiable function (we are assuming that the ground state insulating wave function $\Psi_{\mathbf{k}}$ is nondegenerate and is separated in energy from the excited states by a finite gap).

That $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ is not yet the characteristic function for $p(\mathbf{X})$ in the extended system can be seen from the fact that the first ''moment'' that it generates is not gauge invariant, as required from any physical quantity.[42] If $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ were the characteristic function, the first moment would be, according to Eq. (22), $\langle X_i \rangle_{\mathbf{k}} = \imath \partial_{\alpha_i} \mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) |_{\boldsymbol{\alpha}=0} = \imath \langle \Phi_{\mathbf{k}} | \partial_{k_i} \Phi_{\mathbf{k}} \rangle$. But this is nothing other than the *Berry connection*, which is a gauge-dependent quantity (see, for instance, Ref. 16).

D. Gauge-invariant cumulants

Gauge-invariant moments and cumulants for an extended insulating system can be obtained from the following cumulant generating function, which is the central quantity in this work:

$$\ln C(\boldsymbol{\alpha}) = \frac{V}{(2\pi)^3} \int d\mathbf{k} \ln \mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}), \tag{32}$$

where the average is over all twisted boundary conditions (see Appendix D for special cases where the number of electrons per primitive cell is not an integer). The cumulants are then obtained in the same way as in Eq. (23):

$$\langle X_1^{n_1} \cdots X_d^{n_d} \rangle_c = \iota^n \partial_{\alpha_1^{n_1} \cdots \alpha_d^{n_d}}^n \ln C(\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0}, \tag{33}$$

and likewise for the moments. Before we continue, we point out that $C(\boldsymbol{\alpha})$ is in general different from the function

$$C_W(\boldsymbol{\alpha}) = \frac{V}{(2\pi)^3} \int d\mathbf{k}\, \mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}). \tag{34}$$

This is also a characteristic function, but of a different, gauge-dependent distribution, whose interpretation and relation to $C(\boldsymbol{\alpha})$ will be discussed in Sec. VI B.

As mentioned in Sec. II, Eq. (12) is most easily interpreted when using a periodic gauge: $\Psi_{\mathbf{k}+\mathbf{G}} = \Psi_{\mathbf{k}}$. Since we will recover Eq. (12) starting from $\ln C(\boldsymbol{\alpha})$, a periodic gauge will be assumed in what follows; thus what is meant here by gauge invariance is invariance under transformations that preserve the phase in Eq. (10). The most general $\varphi(\mathbf{k})$ in Eq. (9) that complies with this requirement is[12,43]

$$\varphi(\mathbf{k}) = \beta(\mathbf{k}) - \mathbf{k} \cdot \mathbf{R}, \tag{35}$$

where $\mathbf{R}$ is an arbitrary lattice vector and $\beta(\mathbf{k}+\mathbf{G}) = \beta(\mathbf{k})$. The gauge invariance of the cumulants generated by $\ln C(\boldsymbol{\alpha})$ can now be seen as follows: According to Eqs. (9), (23), and (32), the cumulants change under a general gauge transformation as

$$\langle X_1^{n_1} X_2^{n_2} X_3^{n_3} \rangle_c \to \langle X_1^{n_1} X_2^{n_2} X_3^{n_3} \rangle_c + \iota^{n+1} \frac{V}{(2\pi)^3}$$
$$\times \int d\mathbf{k}\, \partial_{k_1^{n_1} k_2^{n_2} k_3^{n_3}}^n \varphi(\mathbf{k}). \tag{36}$$

Substitution of Eq. (35) into Eq. (36) shows that for $n=1$ the cumulants change as $\langle \mathbf{X} \rangle \to \langle \mathbf{X} \rangle + \mathbf{R}$, whereas for $n>1$ they remain unchanged. The change in $\langle \mathbf{X} \rangle$ but not in the higher cumulants (which do not depend on the mean) indicates a rigid shift by $\mathbf{R}$ of the whole distribution $p(\mathbf{X})$, and is related to the quantum of polarization[12,14] (see Appendix C).

The cumulants are also real, as expected: defining $f_{n_1 n_2 n_3}(\mathbf{k}) = \iota^n \partial_{\alpha_1^{n_1} \alpha_2^{n_2} \alpha_3^{n_3}}^n \ln\langle \Phi_{\mathbf{k}} | \Phi_{\mathbf{k}+\boldsymbol{\alpha}} \rangle|_{\boldsymbol{\alpha}=0}$ and $\boldsymbol{\beta} = -\boldsymbol{\alpha}$, we obtain

$$f_{n_1 n_2 n_3}(-\mathbf{k}) = (-1)^n \iota^n \partial_{\beta_1^{n_1} \beta_2^{n_2} \beta_3^{n_3}}^n \ln\langle \Phi_{-\mathbf{k}} | \Phi_{-(\mathbf{k}+\boldsymbol{\beta})} \rangle|_{\boldsymbol{\beta}=0}.$$

Choosing a gauge such that $\Phi_{-\mathbf{k}} = \Phi_{\mathbf{k}}^*$, which can always be done due to time-reversal symmetry,[43] leads to $\langle \Phi_{-\mathbf{k}} | \Phi_{-(\mathbf{k}+\boldsymbol{\beta})} \rangle = \langle \Phi_{\mathbf{k}+\boldsymbol{\beta}} | \Phi_{\mathbf{k}} \rangle$, and thus $f_{n_1 n_2 n_3}(-\mathbf{k}) = [f_{n_1 n_2 n_3}(\mathbf{k})]^*$. That completes the proof, since according to Eqs. (31), (32), and (33), $\langle X_1^{n_1} X_2^{n_2} X_3^{n_3} \rangle_c$ is given by the average over $\mathbf{k}$ of $f_{n_1 n_2 n_3}(\mathbf{k})$. (Since gauge invariance was previously established, the choice $\Phi_{-\mathbf{k}} = \Phi_{\mathbf{k}}^*$ can be made without loss of generality.)

Let us now look at explicit expressions for the first few cumulants. Combining Eqs. (31) and (32) and taking the first derivative, we find

$$\partial_{\alpha_i} \ln C(\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0} = \frac{V}{(2\pi)^3} \int d\mathbf{k} \langle \Phi_{\mathbf{k}} | \partial_{k_i} \Phi_{\mathbf{k}} \rangle. \tag{37}$$

Together with Eq. (23), this gives

$$\langle X_i \rangle_c = \frac{\iota V}{(2\pi)^3} \int d\mathbf{k} \langle \Phi_{\mathbf{k}} | \partial_{k_i} \Phi_{\mathbf{k}} \rangle, \tag{38}$$

which, with the help of Eq. (29), is seen to be precisely the Berry phase expression for the average electronic polarization [Eq. (12)]. Our formulation in terms of $C(\boldsymbol{\alpha})$ therefore agrees with the Berry phase theory of polarization. It is, however, more general, since it also provides the higher moments: for $n=2$, similar steps as before lead to

$$\langle X_i X_j \rangle_c = -\frac{V}{(2\pi)^3} \int d\mathbf{k} [\langle \Phi_{\mathbf{k}} | \partial_{k_i k_j}^2 \Phi_{\mathbf{k}} \rangle$$
$$- \langle \Phi_{\mathbf{k}} | \partial_{k_i} \Phi_{\mathbf{k}} \rangle \langle \Phi_{\mathbf{k}} | \partial_{k_j} \Phi_{\mathbf{k}} \rangle]. \tag{39}$$

Integrating the first term by parts (using the periodic gauge condition) and noting that $\langle \Phi_{\mathbf{k}} | \partial_{k_i} \Phi_{\mathbf{k}} \rangle = -\langle \partial_{k_i} \Phi_{\mathbf{k}} | \Phi_{\mathbf{k}} \rangle$, this becomes $\langle X_i X_j \rangle_c = V/(8\pi^3) \int d\mathbf{k}\, T_{ij}(\mathbf{k})$, where

$$T_{ij}(\mathbf{k}) = \langle \partial_{k_i} \Phi_{\mathbf{k}} | \partial_{k_j} \Phi_{\mathbf{k}} \rangle - \langle \partial_{k_i} \Phi_{\mathbf{k}} | \Phi_{\mathbf{k}} \rangle \langle \Phi_{\mathbf{k}} | \partial_{k_j} \Phi_{\mathbf{k}} \rangle \tag{40}$$

is the gauge-invariant *quantum geometric tensor*[44] [see also Eq. (C9) of Ref. 33]. The real part of $T_{ij}(\mathbf{k})$ is the metric tensor $G_{ij}(\mathbf{k})$ first introduced by Provost and Vallee:[45]

$$G_{ij}(\mathbf{k}) = \text{Re}\langle \partial_{k_i} \Phi_{\mathbf{k}} | \partial_{k_j} \Phi_{\mathbf{k}} \rangle - \langle \partial_{k_i} \Phi_{\mathbf{k}} | \Phi_{\mathbf{k}} \rangle \langle \Phi_{\mathbf{k}} | \partial_{k_j} \Phi_{\mathbf{k}} \rangle, \tag{41}$$

where the second term is automatically real. $G_{ij}(\mathbf{k})$ is the many-body analog of the tensor $g_{ij}(\mathbf{k})$ defined in Eq. (16). Using Eq. (24) and the fact that the cumulants are real, $\langle X_i X_j \rangle_c$ can be rewritten as

$$\langle X_i X_j \rangle_c = \frac{V}{(2\pi)^3} \int d\mathbf{k}\, G_{ij}(\mathbf{k}), \tag{42}$$

which becomes, after taking the trace, the many-body counterpart of Eq. (15). Together with Eq. (30a), the above equation establishes the physical interpretation of the $\mathbf{k}$-averaged metric tensor as the mean-square fluctuation of the macroscopic bulk polarization (see also the next section). The general connection between such ''quantum metrics'' and quantum fluctuations was pointed out already in Ref. 45.

IV. FLUCTUATION-DISSIPATION RELATION

We have seen that the first cumulant agrees with the Berry phase expression for the average macroscopic polarization. Here we will show that the expression for the second cumulant, Eq. (42), is consistent with the fluctuation-dissipation relation[30–32] between the fluctuations in $\mathbf{P}_{\mathrm{mac}}$ and the optical conductivity Im $\chi_{ij}(\omega) = (1/\omega)\mathrm{Re}\ \sigma_{ij}(\omega)$.

For a confined system with open boundary conditions, which is the case discussed by Kudinov,[29] the Kubo-Greenwood formula for the conductivity can be written in terms of the off-diagonal position matrix elements, $X_{nm}^i = \langle \Psi_n | \hat{X}_i | \Psi_m \rangle$:

$$\mathrm{Re}\ \sigma_{ij}^n(\omega) = \frac{\pi q_e^2}{m_e^2 \hbar \omega V} \sum_{m \neq n} \omega_{mn}^2 \mathrm{Re}[X_{nm}^i X_{mn}^j \delta(\omega_{mn} - \omega)$$
$$- X_{nm}^j X_{mn}^i \delta(\omega_{mn} + \omega)], \quad (43)$$

where $\hbar \omega_{mn} = E_m - E_n$. Alternatively, the well-known relation

$$P_{nm}^i = \imath m_e \omega_{nm} X_{nm}^i \quad (m \neq n) \quad (44)$$

can be used to rewrite Eq. (43) in terms of momentum matrix elements. Whereas for a confined system the two formulas are interchangeable, for an extended system the position matrix elements become ill defined, and therefore only the latter form remains valid.

At this point it is convenient to introduce the notation

$$X_{nm,\mathbf{k}}^i = \imath \langle \Phi_{n\mathbf{k}} | \partial_{k_i} \Phi_{m\mathbf{k}} \rangle = (X_{mn,\mathbf{k}}^i)^*. \quad (45)$$

The single-body analog of such quantities is discussed in Refs. 23 and 46. Our motivation for introducing them is the following: if $\hat{\mathbf{P}} = \Sigma_{i=1}^N \hat{\mathbf{p}}(i)$ is the many-body momentum operator for the extended system, and $\mathbf{P}_{nm,\mathbf{k}} = \langle \Psi_{n\mathbf{k}} | \hat{\mathbf{P}} | \Psi_{m\mathbf{k}} \rangle$, in the case of a nonrelativistic Hamiltonian without spin-orbit coupling Eqs. (5) and (6) lead to

$$P_{nm,\mathbf{k}}^i = \frac{m_e}{\hbar} \langle \Phi_{n\mathbf{k}} | [\partial_{k_i}, \hat{H}(\mathbf{k})] | \Phi_{m\mathbf{k}} \rangle$$
$$= \imath m_e \omega_{nm}(\mathbf{k}) X_{nm,\mathbf{k}}^i \quad (m \neq n), \quad (46)$$

which is formally identical to Eq. (44). Notice, however, that we are now dealing with an *extended* system with twisted boundary conditions, for which the proper position matrix elements, $\langle \Psi_{n\mathbf{k}} | \hat{X}_i | \Psi_{m\mathbf{k}} \rangle$, are ill defined; the above relation shows that they should be replaced by the quantities $X_{nm,\mathbf{k}}^i$. Substituting Eq. (46) into the Kubo-Greenwood formula in the form valid for an extended system, we are left with an expression for Re $\sigma_{ij}^{n\mathbf{k}}(\omega)$ formally identical to Eq. (43), with X_{nm}^i replaced by $X_{nm,\mathbf{k}}^i$ and ω_{mn} replaced by $\omega_{mn}(\mathbf{k})$.

Let us now specialize to the ground state ($n=0$), assuming that it is nondegenerate; then $\omega_{m0}(\mathbf{k}) > 0$, and we obtain

$$\int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\ \sigma_{ij}^{0\mathbf{k}}(\omega) = \frac{\pi q_e^2}{\hbar V} \mathrm{Re}\left\{ \sum_{m>0} X_{0m,\mathbf{k}}^i X_{m0,\mathbf{k}}^j \right\}$$

$$= \frac{\pi q_e^2}{\hbar V} \mathrm{Re}\left\{ \langle \partial_{k_i} \Phi_{0\mathbf{k}} | \left(\sum_{m>0} |\Phi_{m\mathbf{k}}\rangle \langle \Phi_{m\mathbf{k}}| \right) \right.$$

$$\left. \times |\partial_{k_j} \Phi_{0\mathbf{k}}\rangle \right\}. \quad (47)$$

Using the completeness relation to eliminate the excited states on the right-hand side (rhs), and comparing with Eq. (40), we find $T_{ij}(\mathbf{k}) = \Sigma_{m>0} X_{0m,\mathbf{k}}^i X_{m0,\mathbf{k}}^j$. Together with $G_{ij}(\mathbf{k}) = \mathrm{Re}\ T_{ij}(\mathbf{k})$, this yields

$$\int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\ \sigma_{ij}^{0\mathbf{k}}(\omega) = \frac{\pi q_e^2}{\hbar V} G_{ij}(\mathbf{k}). \quad (48)$$

Averaging over all twisted boundary conditions and using Eqs. (33) and (42), we arrive at the desired relation:

$$-\frac{1}{N} \partial_{\alpha_i \alpha_j}^2 \ln C(\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0} = \frac{1}{N} \langle X_i X_j \rangle_c$$

$$= \frac{\hbar}{\pi q_e^2 n_0} \int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\ \bar{\sigma}_{ij}(\omega), \quad (49)$$

where $n_0 = N/V$ and $\bar{\sigma}_{ij}(\omega) = (V/8\pi^3) \int d\mathbf{k}\ \sigma_{ij}^{0\mathbf{k}}(\omega)$. Equation (49) is precisely the fluctuation-dissipation relation for more than one variable[31,32] for the extended system at $T=0$ [compare with Eq. (13), using Eq. (30a) and setting $V\mathbf{P}_{\mathrm{mac}} = \mathbf{d}$].

V. LOCALIZATION LENGTH

A. Relation to the conductivity

Here we will generalize to extended systems Kudinov's analysis of the fluctuations in $\mathbf{P}_{\mathrm{mac}}$ as a way to distinghish insulators from conductors[29] and discuss it in terms of a localization length. It is convenient for that purpose to classify solids into three categories, according to the low-frequency behavior of the conductivity at $T=0$ as $V \to \infty$:

$$\lim_{\omega \to 0} \mathrm{Re}\ \sigma(\omega) = 0, \quad \text{insulators}$$

$$\mathrm{Re}\ \sigma(\omega) = (2\pi q_e^2/\hbar^2) D\delta(\omega) + \mathrm{Re}\ \sigma_{\mathrm{reg}}(\omega),$$

$$\text{ideal conductors} \quad (50)$$

$$\lim_{\omega \to 0} \mathrm{Re}\ \sigma(\omega) = \sigma_0, \quad \text{nonideal conductors}.$$

Insulators are characterized by a vanishing dc conductivity, in contrast to conductors. The singular contribution $(2\pi q_e^2/\hbar^2) D\delta(\omega)$ occurs in ideal conductors—those without any scattering mechanism—and D is called the "Drude weight" or "charge stiffness."[39,47] If there is scattering, the δ function peak is smeared out to a Lorentzian, so that $D = 0$ and the dc conductivity of nonideal conductors, such as disordered metallic alloys, has a finite value σ_0; $\sigma_{\mathrm{reg}}(\omega)$ is the regular finite-frequency part of $\sigma(\omega)$ in perfect conductors.

TABLE I. Comparison between the formulas for the Drude weight and for the localization length, their relation to the optical conductivity, and their asymptotic values in the thermodynamic limit for insulators and conductors at $T=0$.

	Drude weight	Localization length
Formula in terms of twisted boundary conditions	$D_i = \dfrac{1}{2V} \left. \dfrac{\partial^2 E(\mathbf{k})}{\partial k_i^2} \right|_{\mathbf{k}=0}$	$\xi_i^2(N) = -\dfrac{1}{N} \left. \dfrac{\partial^2 \ln C(\boldsymbol{\alpha})}{\partial \alpha_i^2} \right|_{\boldsymbol{\alpha}=0}$
Relation to conductivity	$D_i = -\dfrac{1}{2} \lim\limits_{\omega \to 0} \omega \, \mathrm{Im}\, \sigma_{ii}(\omega)$	$\xi_i^2(N) = \dfrac{\hbar}{\pi q_e^2 n_0} \int_0^\infty \dfrac{d\omega}{\omega} \, \mathrm{Re}\, \sigma_{ii}(\omega)$
Asymptotic value $(N, V \to \infty)$		
Insulators	Zero	Finite
Nonideal conductors	Zero	Infinite
Ideal conductors	Finite	Infinite

The only possible divergence of the integral on the rhs of Eq. (49) is around $\omega = 0$,[48] because as $\omega \to \infty$ $\mathrm{Im}\, \chi_{ii}(\omega)$ $\sim \omega^{-3}$.[2] Substituting Eq. (50) into Eq. (49) we see that the quantity ξ_i in Eq. (30b) is finite for insulators and diverges for conductors. By the same token, using the zero-frequency limit of the Kramers-Krönig relation,[1,2] one finds the familiar result that the static susceptibility is finite for insulators and divergent for conductors. Hence, assuming Eq. (50), at $T = 0$ the following three conditions are equivalent: (i) $\mathrm{Re}\, \chi_{ii}(0)$ is finite, (ii) $\lim_{\omega \to 0} \mathrm{Re}\, \sigma_{ii}(\omega) = 0$, and (iii) ξ_i is finite.

The quantity ξ_i has the dimensions of length. Since it is finite for insulators and infinite for conductors, it is natural to interpret it as an electronic localization length along the ith direction. According to Eq. (49), for extended systems with a finite volume $\xi_i(N)$ can be written in the form given in Table I in terms of the cumulant generating function. This formula has a striking similarity to the Drude weight formula derived by Kohn in terms of the total energy $E(\mathbf{k})$,[8,47,49] given also in Table I: both are second derivatives of some quantity with respect to the twisted boundary conditions [in the case of $\xi_i(N)$ the twisting of the boundary condition $\mathbf{k}$ is followed by an averaging over all $\mathbf{k}$, hence the parameter $\boldsymbol{\alpha}$ instead of $\mathbf{k}$].

It is clear from Table I that unlike the localization length, the Drude weight does not provide a universal criterion to discern insulators from conductors.[8,9] However, the combination of the two quantities in principle enables us to distinguish between the three categories. In the same way that the Drude weight measures the ''degree of conductivity'' of an ideal conductor, $1/\xi_i$ measures the degree of localization of the electrons in an insulator. Insofar as localization—in a properly defined sense—is an essential property of the insulating state,[8,9] this can be viewed as a meaningful measure of the ''degree of insulation,'' one which is expected to apply to all types of insulators.

Although we have managed to express ξ_i in terms of the measurable optical conductivity via the fluctuation-dissipation relation, it is not yet clear how it relates to the notion of localization put forward by Kohn, in terms of the localization properties of the insulating wave function in configuration space. That will be discussed in Sec. VI B.

B. Relation to the optical gap

In an insulator optical absorption starts at a threshold energy E_g, below which $\mathrm{Re}\, \sigma_{ii}(\omega) = 0$ (we are neglecting phonon-assisted transitions, so that the gap E_g is the minimum gap for optical transitions). From this it follows that

$$\int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\, \sigma_{ii}(\omega) \le \frac{\hbar}{E_g} \int_0^\infty d\omega \, \mathrm{Re}\, \sigma_{ii}(\omega). \quad (51)$$

With the help of the sum rule for oscillator strengths[1,2,50] $\int_0^\infty d\omega \, \mathrm{Re}\, \sigma_{ii}(\omega) = (1/8)\omega_p^2$ (ω_p is the plasma frequency), together with Eqs. (30) and (49), we conclude that

$$\xi_i^2 \le \frac{\hbar^2}{2m_e E_g}. \quad (52)$$

This inequality shows that the polarization fluctuations [Eqs. (30)] are controlled by the optical gap, lending support to the intuitive notion that the larger the gap, the more localized the electrons. It strongly resembles an inequality previously derived by Kivelson[51] for noninteracting electrons in one dimension, where ξ_i^2 is replaced by the quadratic spread of properly chosen Wannier functions. As discussed in Appendix A, in fact in one dimension ξ_i^2 equals the average spread of the maximally localized Wannier functions.

VI. MANY-BODY WANNIER FUNCTIONS

The expression for the cumulant generating function, Eq. (32), involves an average over all twisted boundary conditions, which was introduced in a somewhat *ad hoc* manner in order to render the resulting distribution $p(\mathbf{X})$ gauge invariant (modulo a rigid shift by a quantum). We will now shed some light on the physical significance of the averaging procedure, by showing how it can be rationalized in terms of the notion of electronic localization in insulators developed by Kohn.[8,9] This will be achieved by introducing many-body Wannier functions and will allow us to tie together the theory of bulk polarization and Kohn's theory of localization.
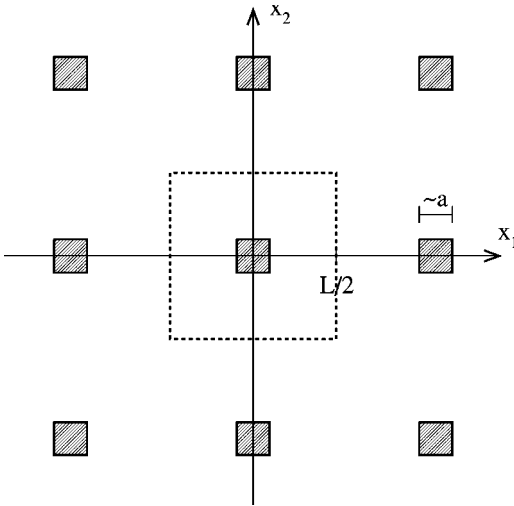
FIG. 1. Schematic representation of the regions $\mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N}$ in the many-electron configuration space where the wave function of a confined system obeying periodic boundary conditions is localized. The system has linear dimensions $\sim a$, and the periodic boundary conditions are over a length $L \gg a$, so that the regions $\mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N}$ are essentially nonoverlapping.

A. Confined system: Localization in real space

It is instructive to start by discussing the case of a confined system of linear dimensions $\sim a$ (e.g., a molecule with N electrons). Instead of the usual open boundary conditions, we can choose to impose periodic boundary conditions on its wavefunction [$\mathbf{k} = 0$ in Eq. (4)], choosing $L \gg a$. The resulting N-electron wave function is periodic in configuration space, as depicted schematically in Fig. 1 for one dimension and $N=2$; it is a sum of partial wave functions that only differ from one another by a translation of the coordinates,

$$\Psi_{\mathbf{k}=0}(\mathbf{x}_1, \ldots, \mathbf{x}_N) = \sum_{\mathbf{m}_1} \cdots \sum_{\mathbf{m}_N} \Psi_{\mathbf{m}_1 \cdots \mathbf{m}_N}(\mathbf{x}_1, \ldots, \mathbf{x}_N),$$
(53)

where the integer vectors $\{\mathbf{m}_i\}$ label the partial wave functions. These are localized in geometrically equivalent regions $\mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N}$ in configuration space (shaded regions in Fig. 1), which for $L \gg a$ are essentially nonoverlapping:

$$\Psi_{\mathbf{m}_1 \cdots \mathbf{m}_N} \Psi_{\mathbf{m}'_1 \cdots \mathbf{m}'_N} \doteq 0$$

$$\text{for } (\mathbf{m}_1, \ldots, \mathbf{m}_N) \neq (\mathbf{m}'_1, \ldots, \mathbf{m}'_N), \quad (54)$$

where, using the notation of Refs. 8 and 9, the symbol $\doteq$ denotes equality apart from exponential small corrections that vanish in a manner such as $e^{-L/\xi}$, where ξ is a localization length (in this example $\xi \sim a$).

Next we switch from periodic to twisted boundary conditions [$\mathbf{k} \neq 0$ in Eq. (4)]. From the confined character of the system it follows, by a simple generalization of the arguments outlined in Sec. 2 of Ref. 8, that the periodic part of the wavefunction, $\Phi_{\mathbf{k}} = e^{-\imath \mathbf{k} \cdot \mathbf{X}} \Psi_{\mathbf{k}}$, can be written with an exponentially small error as

$$\Phi_{\mathbf{k}}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \doteq e^{-\imath \mathbf{k} \cdot \mathbf{Q}} \Phi_{\mathbf{k}=0}(\mathbf{x}_1, \ldots, \mathbf{x}_N). \quad (55)$$

Here we have introduced the quantity

$$\mathbf{Q} = \begin{cases} \displaystyle\sum_{i=1}^{N} (\mathbf{x}_i - \mathbf{L}_{\mathbf{m}_i}) \text{ in } \mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N} \\[12pt] \mathbf{F}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \text{ outside all } \mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N} \end{cases} \quad (56)$$

where $\mathbf{F}$ is a largely arbitrary, periodic function, which joins smoothly with the values of $\mathbf{Q}$ at the boundaries of the regions $\mathcal{R}_{\mathbf{m}_1 \cdots \mathbf{m}_N}$. Let us now look at the precursor characteristic function $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ for such a confined system obeying twisted boundary conditions: substituting the previous expressions into Eq. (31), we find

$$\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) \doteq \langle \Phi_{\mathbf{k}=0} | e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{Q}}} | \Phi_{\mathbf{k}=0} \rangle \doteq \langle \Psi_{0 \cdots 0} | e^{-\imath \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | \Psi_{0 \ldots 0} \rangle, \quad (57)$$

which has the form of Eq. (27), obtained using open boundary conditions. It is clear that as a result of localization of the electrons in real space the choice of boundary conditions is immaterial. In particular, $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ becomes independent of $\mathbf{k}$, and thus from Eqs. (32) and (34) we conclude that

$$\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) \doteq C_W(\boldsymbol{\alpha}) \doteq C(\boldsymbol{\alpha}). \quad (58)$$

Hence, averaging over boundary conditions becomes redundant, and $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ is already the correct, gauge-invariant characteristic function. Finally, it is interesting to note that Eqs. (55) and (57) can be viewed as a particular realization of Eqs. (2.22) and (2.23) of Ref. 45, which were used to relate the quantum metric to the quantum fluctuations for a particular class of manifolds.

B. Extended insulator: Localization in configuration space

Let us now consider an extended system, such as a crystal with twisted boundary conditions over a large but finite volume V. Although in general the charge density will be delocalized in real space, Kohn has argued that if the system is insulating the wave function is localized in configuration space.[8,9] Kohn's notion of localization is weaker than the one implied by Eq. (54) and Fig. 1; however, as we will see, it is sufficient to recover Eq. (58), after making a judicious choice of gauge.

We start by introducing a localized description of the many-electron insulating wave function in configuration space. In the noninteracting case, a localized one-electron description (in real space) is provided by the Wannier functions.[23,26,43] The Wannier function $|\mathbf{R}n\rangle$ associated with band n and centered around the unit cell labeled by the lattice vector $\mathbf{R}$ is related to the Bloch functions by the following unitary transformation:

$$|\mathbf{R}n\rangle = \frac{1}{\sqrt{N_c}} \sum_{\mathbf{k}} e^{-\imath \mathbf{k} \cdot \mathbf{R}} |\psi_{n\mathbf{k}}\rangle, \quad (59)$$

where a periodic gauge is assumed, and the sum is over a uniform grid of N_c points in the Brillouin zone.[52] Due to the discretization of the integral over $\mathbf{k}$, $|\mathbf{R}n\rangle$ is actually periodic in a large cell of volume $N_c v$ (v is the volume of the unit cell). The many-body analog of a periodic Wannier function (many-body Wannier function) can be defined in a similar way in terms of the many-body wave function $\Psi_{\mathbf{k}}$ (again assuming a periodic gauge):

$$|W_\mathbf{M}\rangle = \frac{1}{\sqrt{N_c}} \sum_\mathbf{k} e^{-\iota \mathbf{k} \cdot \mathbf{R_M}} |\Psi_\mathbf{k}\rangle, \qquad (60)$$

so that, using Eq. (5),

$$|\Phi_\mathbf{k}\rangle = \frac{1}{\sqrt{N_c}} \sum_\mathbf{M} e^{-\iota \mathbf{k} \cdot (\hat{\mathbf{X}} - \mathbf{R_M})} |W_\mathbf{M}\rangle. \qquad (61)$$

Here $\mathbf{R_M} = \Sigma_{i=1}^3 M_i \mathbf{L}_i$, where the M_i are integers, and the vectors $\{\mathbf{L}_1, \mathbf{L}_2, \mathbf{L}_3\}$ define the volume V containing N electrons (notice that V is the volume of a supercell, typically a large multiple of the unit cell volume v). $\Phi_\mathbf{k}$ is periodic and $\Psi_\mathbf{k}$ obeys $\mathbf{k}$ boundary conditions over the volume V^N in configuration space, and both $\Psi_\mathbf{k}$ and $W_\mathbf{M}$ are periodic over the volume $(N_c V)^N$. The normalization conventions are the following: $\Phi_\mathbf{k}$ is normalized to one over a volume V^N, and $W_\mathbf{M}$ is normalized to one over a volume $(N_c V)^N$. Similarly to the $|\mathbf{R}n\rangle$, the $|W_\mathbf{M}\rangle$ form an orthonormal set.

From the same type of general considerations which are used to show that Wannier functions can be chosen localized,[26] it follows that we can choose $W_\mathbf{M}$ localized in the variable $\mathbf{X} = \Sigma_{i=1}^N \mathbf{x}_i$, with a distribution

$$p_\mathrm{W}(\mathbf{X}) = \langle W_\mathbf{M} | \delta(\hat{\mathbf{X}} - \mathbf{X}) | W_\mathbf{M} \rangle. \qquad (62)$$

Substituting Eq. (61) in Eq. (31), averaging over $\mathbf{k}$, and comparing with Eq. (34), after discretizing the integral over $\mathbf{k}$, yields

$$C_\mathrm{W}(\boldsymbol{\alpha}) = \frac{1}{N_c} \sum_\mathbf{k} \mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) = \langle W_\mathbf{M} | e^{-\iota \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | W_\mathbf{M} \rangle, \qquad (63)$$

which shows that the function $C_\mathrm{W}(\boldsymbol{\alpha})$ introduced in Sec. III D is the characteristic function of the distribution $p_\mathrm{W}(\mathbf{X})$. It is straightforward to check that the first moment of this distribution equals the first moment of the gauge-invariant polarization distribution $p(\mathbf{X})$ generated by $C(\boldsymbol{\alpha})$; this is similar to the independent-electron case, where the average polarization is given by the sum of the centers of charge of the gauge-dependent Wannier functions.[12] Likewise, the many-body Wannier functions $W_\mathbf{M}$ are gauge dependent, and as a result so are the higher cumulants of $p_\mathrm{W}(\mathbf{X})$, in particular, the quadratic spread. Notice that the gauge dependence of the $W_\mathbf{M}$ implies that in general they overlap with one another,[53] and only in certain gauges are they localized to the point of being essentially nonoverlapping (provided that the system is insulating). This is different from the usual single-body Wannier functions, which in general remain overlapping in any gauge (even though they are orthogonal, so that the overlap integrals vanish); in the many-body case we are free to choose the volume V large enough so that with a judicious choice of gauge the $W_\mathbf{M}$ become nonoverlapping in the high-dimensional configuration space.

We are now in a position to provide the link between the present formalism (and, therefore, the Berry phase theory of polarization) and Kohn's theory of localization.[8,9] A major result of Kohn's work is the conjecture that a general many-body insulating wave function $\Psi_{\mathbf{k}=0}$, with periodic boundary conditions over a large volume V, breaks up into a sum of nonoverlapping parts $\Psi_\mathbf{M}$, localized in disconnected regions $\mathcal{R}_\mathbf{M}$ in configuration space [Eq. (1)]. Upon inspection,
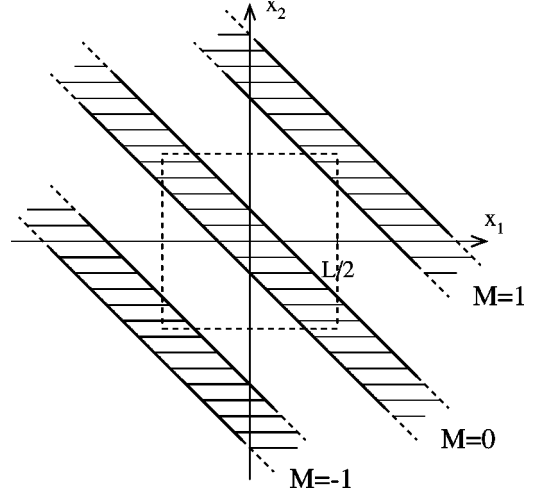

FIG. 2. Schematic representation of the essentially disconnected regions $\mathcal{R}_\mathbf{M}$ in the many-electron configuration space where the wave function of an extended insulator is localized (adapted from Ref. 9). For d real-space dimensions $\mathbf{M} = (M_1, \ldots, M_d)$, and shown is the case of two electrons and $d = 1$, for which the configuration space is (x_1, x_2). The system is composed of possibly strongly overlapping units in real space (e.g., a covalent insulator) and yet, because it is insulating, in configuration space the wave function Ψ breaks up into a sum of partial functions $\Psi_\mathbf{M}$, each localized in a region $\mathcal{R}_\mathbf{M}$, which have an exponentially small overlap with one another, if the system is large.

one finds that the present many-body Wannier functions are nothing other than Kohn's functions, $W_\mathbf{M} = \sqrt{N_c} \Psi_\mathbf{M}$ [compare Eq. (61) with Eq. (6.1) of Ref. 8], except that Kohn only considered gauges where they are localized in such a way that the overlap becomes exponentially small:

$$\Psi_\mathbf{M}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \Psi_{\mathbf{M}'}(\mathbf{x}_1, \ldots, \mathbf{x}_N) \doteq 0 \quad \text{for } \mathbf{M}' \neq \mathbf{M}, \qquad (64)$$

so that the $\Psi_\mathbf{M}$ are uniquely defined apart from exponentially small variations and an overall phase. Transposing the language of Ref. 33 to a many-body framework, Kohn's functions $\Psi_\mathbf{M}$ can be viewed as the maximally localized many-body Wannier functions $W_\mathbf{M}$ (see Appendix B).

In such "nonoverlapping gauges," the precursor characteristic function, Eq. (31), becomes, using Eqs. (61) and (64),

$$\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) \doteq N_c \langle \Psi_\mathbf{M} | e^{-\iota \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | \Psi_\mathbf{M} \rangle \quad \text{(nonoverlapping gauge)}, \qquad (65)$$

which is independent of $\mathbf{k}$, since the $\mathbf{k}$ dependence of $\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$ in a general gauge arises from the cross terms between different $W_\mathbf{M}$. Therefore in such gauges we recover Eq. (58), which for a confined system obeying twisted boundary conditions was valid in any gauge:

$$\mathcal{C}(\mathbf{k}, \boldsymbol{\alpha}) \doteq C_\mathrm{W}(\boldsymbol{\alpha}) \doteq C(\boldsymbol{\alpha}) \quad \text{(nonoverlapping gauge)}. \qquad (66)$$

Finally, from the gauge invariance of $C(\boldsymbol{\alpha})$ we conclude that *in any gauge*

$$C(\boldsymbol{\alpha}) \doteq N_c \langle \Psi_\mathbf{M} | e^{-\iota \boldsymbol{\alpha} \cdot \hat{\mathbf{X}}} | \Psi_\mathbf{M} \rangle, \qquad (67)$$
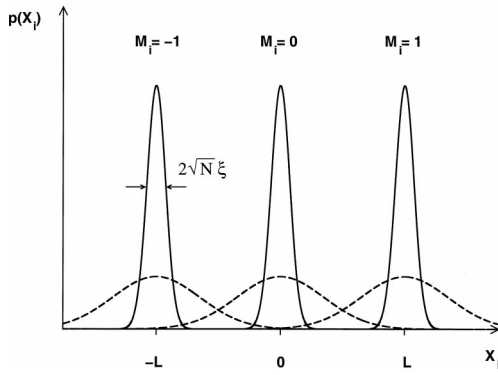
FIG. 3. Localized distribution $p(X_i)$ along the ith direction of N times the electronic center of mass $(\mathbf{X} = \Sigma_{j=1}^N \mathbf{x}_j)$ for a d-dimensional insulator with N electrons in a periodic volume L^d (based on Fig. 6 of Ref. 8). Although each individual electron coordinate $\mathbf{x}_j$, as well as the electronic charge density (not shown), is spread over the whole system, because the system is insulating a localized distribution $p(X_i)$ of width $2\sqrt{N}\xi_i$ can be uniquely defined in terms of the partial wave function $\Psi_{\mathbf{M}}$ in a *single* disconnected region $\mathcal{R}_{\mathbf{M}}$ in the dN-dimensional configuration space (Fig. 2). Choosing a different region $\mathcal{R}_{\mathbf{M}'}$ simply shifts the center of the distribution, if $M'_i \neq M_i$. The solid lines correspond to $d=1$, for which the peaks coming from different $\mathcal{R}_{\mathbf{M}}$ do not overlap with one another for large L, whereas for $d=3$ (dotted lines) they overlap strongly (but the regions $\mathcal{R}_{\mathbf{M}}$ are still essentially disconnected, for large L.)

which is the desired relation. It should be stressed that in deriving this equation we *assumed* Kohn's conjecture regarding the existence of nonoverlapping many-body Wannier functions in the insulating state.

We interpret Eq. (67) as follows: in each region $\mathcal{R}_{\mathbf{M}}$ in the configuration space of the variables $\{\mathbf{x}_i\}$ (Fig. 2), the variable $\mathbf{X} = \Sigma_{i=1}^N \mathbf{x}_i$ takes on a range of values with a distribution generated by $C(\boldsymbol{\alpha})$,

$$p(\mathbf{X}) \doteq N_c \langle \Psi_{\mathbf{M}} | \delta(\hat{\mathbf{X}} - \mathbf{X}) | \Psi_{\mathbf{M}} \rangle, \tag{68}$$

which, together with Eq. (29), gives the bulk polarization distribution. The previous equation can therefore be viewed as the generalization to extended insulating systems of Eq. (25) for the distribution of the electronic center of mass in confined systems.

In Fig. 3 is represented the distribution $p(X_i)$ along the ith direction. According to Eq. (30), its width is $\sqrt{N}\xi_i(N)$. The solid lines describe what happens in a 1D insulator, for which the width of each of the peaks $p(X)$ labeled by M is $\sqrt{N}\xi(N) \propto \sqrt{L}$ and the distance between the centers of consecutive peaks is L, so that for large L they are well separated. In the case of a 3D insulator where all the linear dimensions are similar $(V \sim L^3)$, the peaks $p(X_i)$ overlap for large L, since their width is $\sqrt{N}\xi_i(N) \propto L^{3/2}$ (dashed lines in Fig. 3). The important observation is that, according to Eqs. (64) and (68), even if they are *overlapping*, the distributions $p(\mathbf{X})$ are well-defined ''projections'' into real space of essentially *disconnected* distributions $|\Psi_{\mathbf{M}}|^2$ in configuration space.[54]

At this point we shall reconsider the problem of how to define a meaningful many-body position operator for the electrons in extended systems, an issue which has been re-

cently discussed in Ref. 17. We recall that the source of the problem is that in such systems the usual center-of-mass position operator $\hat{\mathbf{X}}$ is ill-defined (see Sec. III C). Kohn[8,9] proposed the following operator as the substitute for $\hat{\mathbf{X}}$ for taking expectation values over the ground state wave function of an extended *insulator*:

$$\hat{\mathbf{Q}} = \begin{cases} \hat{\mathbf{X}} - \mathbf{R}_{\mathbf{M}} & \text{in } \mathcal{R}_{\mathbf{M}} \\ \mathbf{F}(\mathbf{x}_1, \ldots, \mathbf{x}_N) & \text{outside all } \mathcal{R}_{\mathbf{M}}, \end{cases} \tag{69}$$

where $\mathbf{F}$ is defined in a similar way as in Eq. (56). It is straightforward to check that the moments calculated from $\hat{\mathbf{Q}}$ coincide with those derived from the characteristic function: with the help of Eqs. (1), (67), and (69), we find

$$\langle \Psi_{\mathbf{k}=0} | \hat{Q}_j^n | \Psi_{\mathbf{k}=0} \rangle \doteq N_c \langle \Psi_{\mathbf{M}=0} | \hat{X}_j^n | \Psi_{\mathbf{M}=0} \rangle$$

$$\doteq \iota^n \partial_{\alpha_j^n}^n C(\boldsymbol{\alpha}) |_{\boldsymbol{\alpha}=0}. \tag{70}$$

The connection with Ref. 17 will be made in Sec. VII B.

C. Insensitivity of bulk properties to the boundary conditions

We now have an adequate framework for discussing the implications of localization for the dependence on boundary conditions of the bulk properties of insulators. As a first example, consider the adiabatic current $\mathbf{J}_{\mathbf{k}}^{(\lambda)}$. Since it is gauge invariant, we may evaluate it in any gauge, in particular in a nonoverlapping gauge. Substituting Eq. (61) into Eq. (7) we find, because of the exponential decrease of the overlap as the size L is increased:

$$\mathbf{J}_{\mathbf{k}}^{(\lambda)} \doteq q_e N_c \partial_\lambda \langle \Psi_{\mathbf{M}}^{(\lambda)} | \hat{\mathbf{X}} | \Psi_{\mathbf{M}}^{(\lambda)} \rangle \doteq q_e \partial_\lambda \langle \mathbf{X} \rangle^{(\lambda)}, \tag{71}$$

where for the last equality Eqs. (33) and (67) were used. Thus for a large system size $\mathbf{J}_{\mathbf{k}}^{(\lambda)}$ is essentially $\mathbf{k}$ independent (this was demonstrated in Ref. 25 using a different reasoning, for insulators with an energy gap).

As a second example, let us look at the quantum geometric tensor $T_{ij}(\mathbf{k})$ and its real part, the metric tensor $G_{ij}(\mathbf{k})$. Evaluating Eq. (40) in a nonoverlapping gauge with the help of Eq. (61), we find $T_{ij}(\mathbf{k}) \doteq \langle X_i X_j \rangle_c$, which again is essentially independent of $\mathbf{k}$ for large systems [it is also real, so that $T_{ij}(\mathbf{k}) \doteq G_{ij}(\mathbf{k})$]. Thus we conclude that the $\mathbf{k}$-independent metric tensor gives the mean-square fluctuation of the polarization. This is a stronger statement than the one made at the end of Sec. III D, which pertained to the $\mathbf{k}$-averaged metric [by contrast, the single-electron metric tensor $g_{ij}(\mathbf{k})$ given by Eq. (16) is in general $\mathbf{k}$ dependent, since the one-electron Wannier functions remain overlapping even when they are maximally localized; as a consequence, the quadratic polarization fluctuations are related to its *average* over the Brillouin zone, as shown in Appendix A.]

These two examples illustrate an important point: the physically measurable bulk quantities, such as the polarization current and the polarization fluctuations, are well defined for any single boundary condition $\mathbf{k}$, and the choice of $\mathbf{k}$ becomes immaterial for large system sizes. It may therefore seem bizarre that in the generating function formalism one needs to average over all $\mathbf{k}$ before obtaining gauge-invariant quantities, as shown in Sec. III D. A concrete ex-

ample helps to clarify this state of affairs: in the case of the second cumulant, had we taken the second derivative of the *precursor* cumulant generating function, $\ln \mathcal{C}(\mathbf{k}, \boldsymbol{\alpha})$, *without* afterwards averaging over $\mathbf{k}$, we would be left with a quantity [the integrand of Eq. (39)], which is different from the desired one, $-T_{ij}(\mathbf{k})$, and is gauge dependent. However, the $\mathbf{k}$ average of that gauge-dependent quantity *is* gauge invariant and equals the $\mathbf{k}$ average of $-T_{ij}(\mathbf{k}) \doteq -G_{ij}(\mathbf{k})$ [compare Eqs. (39) and (42)]. A similar situation occurs with the adiabatic polarization current.

The insensitivity of the bulk properties of insulators to the choice of boundary conditions on a large system seems to be a very general property. Indeed, if $\hat{\mathcal{O}}$ is any well-defined operator acting on the ground state of the extended system (e.g., the Hamiltonian $\hat{H}$), then from Eqs. (5), (61), and (64) it follows that $\langle \Psi_{\mathbf{k}} | \hat{\mathcal{O}} | \Psi_{\mathbf{k}} \rangle \doteq \langle \Psi_{\mathbf{k}=0} | \hat{\mathcal{O}} | \Psi_{\mathbf{k}=0} \rangle$, which helps to understand why, for instance, in insulators the Drude weight (see Table I) goes to zero exponentially with the system size.[8] This has been confirmed by numerical simulations[47] and calculations on exactly solvable models.[49] More precisely, in Ref. 49 it was found that the Drude weight for the half-filled Hubbard model in one dimension scales as $\sim L^{1/2} e^{-L/\tilde{\xi}}$, which yields a localization length $\tilde{\xi}$. It would be worthwhile to investigate whether $\tilde{\xi}$ is the same as the localization length ξ used in the present work.

VII. DISCRETIZED FORMULAS

Until now we have dealt with a continuum formalism, in which the cumulants are obtained by differentiating $\ln C(\boldsymbol{\alpha})$ at $\boldsymbol{\alpha}=0$. Heuristically, this can be viewed as a measure of the $\mathbf{k}$-averaged change in $\Phi_{\mathbf{k}}$ as the boundary condition $\mathbf{k}$ is twisted adiabatically [see Eqs. (31), (32), and (33)]. In numerical calculations it is often more natural to perform *independent* calculations on a discrete mesh of $\mathbf{k}$ points and then use the resulting wave functions to estimate the derivatives by finite differences.[12,33] Since the overall phases of the wave functions on the grid are unrelated, useful discretized expressions must remain invariant under arbitrary changes of those phases. The derivation of such discretized formulas becomes particularly transparent in the present formalism, as we will now show.

A. Formulas involving an average over k

We will find it convenient to work with scaled coordinates: let the simulation cell be defined by the vectors $\{\mathbf{L}_1, \mathbf{L}_2, \mathbf{L}_3\}$, to which correspond the reciprocal lattice vectors $\{\mathbf{G}^1, \mathbf{G}^2, \mathbf{G}^3\}$ ($\mathbf{L}_i \cdot \mathbf{G}^j = 2\pi \delta_i^j$). The scaled coordinates S^i are defined by $\mathbf{X} = S^i \mathbf{L}_i$, where a sum over repeated indices is implied; similarly we have $\mathbf{k} = q_i \mathbf{G}^i$ and $\boldsymbol{\alpha} = t_i \mathbf{G}^i$ (and thus $\boldsymbol{\alpha} \cdot \mathbf{X} = 2\pi t_i S^i$). In terms of these variables, Eqs. (31) and (32) become

$$\mathcal{C}(\mathbf{q}, \mathbf{t}) = \langle \Psi_{\mathbf{q}} | e^{-\iota 2\pi t_i S^i} | \Psi_{\mathbf{q}+\mathbf{t}} \rangle = \langle \Phi_{\mathbf{q}} | \Phi_{\mathbf{q}+\mathbf{t}} \rangle, \quad (72)$$

and

$$\ln C(\mathbf{t}) = \int_0^1 dq_1 \int_0^1 dq_2 \int_0^1 dq_3 \ln \mathcal{C}(\mathbf{q}, \mathbf{t}), \quad (73)$$

so that $C(\mathbf{t})$ is the joint characteristic function for the variables $2\pi S^i$.

The average is

$$\langle S^l \rangle = \frac{\iota}{2\pi} \int d\mathbf{q} \, \partial_{t_l} \ln \mathcal{C}(\mathbf{q}, \mathbf{t})|_{\mathbf{t}=0}. \quad (74)$$

Next we discretize the derivative in the integrand as

$$\delta q_l \partial_{t_l} \ln \mathcal{C}(\mathbf{q}, \mathbf{t})|_{\mathbf{t}=0} \simeq \ln \mathcal{C}(\mathbf{q}, \delta \mathbf{q}_l) - \ln \mathcal{C}(\mathbf{q}, 0)$$

$$\simeq \iota \, \mathrm{Im} \ln \mathcal{C}(\mathbf{q}, \delta \mathbf{q}_l), \quad (75)$$

where we made use of Eq. (31). This expression is gauge dependent, just like its continuum counterpart, the Berry connection. As in the continuum case, gauge invariance is recovered by averaging over $\mathbf{k}$; that is done by choosing a row of $J+1$ $\mathbf{k}$ points along the direction of $\mathbf{G}^l$ such that the endpoints are separated by $\mathbf{G}^l$; then we find

$$\langle S^l \rangle \simeq -\frac{1}{2\pi} \int dq_i dq_j \sum_{\gamma=0}^{J-1} \mathrm{Im} \ln \langle \Phi_{\mathbf{k}_\gamma} | \Phi_{\mathbf{k}_{\gamma+1}} \rangle, \quad (76)$$

where (i, j, l) is some permutation of $(1,2,3)$, and the periodic gauge is enforced by setting $\Phi_{\mathbf{k}_J} = e^{-\iota \mathbf{G}^l \cdot \mathbf{X}} \Phi_{\mathbf{k}_0}$. The average polarization is then given by

$$\langle \mathbf{P}_{\mathrm{el}} \rangle \cdot \mathbf{G}^l = \frac{2\pi q_e}{V} \langle S^l \rangle$$

$$\simeq -\frac{q_e}{V} \int dq_i dq_j \, \mathrm{Im} \ln \prod_{\gamma=0}^{J-1} \langle \Phi_{\mathbf{k}_\gamma} | \Phi_{\mathbf{k}_{\gamma+1}} \rangle, \quad (77)$$

which is the many-body analog of the discretized Berry phase formula proposed in Ref. 12. It is straightforward to verify that it is gauge invariant modulo the quantum of polarization. This property hinges upon (i) choosing a periodic gauge, and (ii) having a logarithm in the expression, which in the present derivation appears quite naturally, coming from the cumulant generating function, Eq. (73).[55]

To compute the variance we discretize the second logarithmic derivative:

$$(\delta q_l)^2 \partial_{t_l^2}^2 \ln \mathcal{C}(\mathbf{q}, \mathbf{t})|_{\mathbf{t}=0}$$

$$\simeq \ln \mathcal{C}(\mathbf{q}, \delta \mathbf{q}_l) + \ln \mathcal{C}(\mathbf{q}, -\delta \mathbf{q}_l) - 2 \ln \mathcal{C}(\mathbf{q}, 0)$$

$$= \ln[\mathcal{C}(\mathbf{q}, \delta \mathbf{q}_l) \mathcal{C}(\mathbf{q}, -\delta \mathbf{q}_l)], \quad (78)$$

which is gauge dependent, similarly to Eq. (75).[56] After some manipulations we obtain the following gauge-invariant, $\mathbf{k}$-averaged formula:

$$\langle (S^l)^2 \rangle_c \simeq -\frac{J}{(2\pi)^2} \int dq_i dq_j \ln \left| \prod_{\gamma=0}^{J-1} \langle \Phi_{\mathbf{k}_\gamma} | \Phi_{\mathbf{k}_{\gamma+1}} \rangle \right|^2. \quad (79)$$

The evaluation of the covariance requires taking cross derivatives, which can be discretized as

$$\partial_{xy}^2 f \simeq \frac{1}{2\,\delta x\,\delta y}[f(x+\delta x,y+\delta y)+f(x-\delta x,y-\delta y)$$

$$-f(x+\delta x,y)-f(x-\delta x,y)-f(x,y+\delta y)$$

$$-f(x,y-\delta y)+2f(x,y)], \qquad (80)$$

leading to

$$\langle S^j S^l \rangle_c \simeq -\frac{1}{2(2\pi)^2}\int dq_i \Bigg[\ln \prod_{\nu=0}^{J_j-1}\prod_{\gamma=0}^{J_l-1} |\langle \Phi_{\mathbf{k}_{\nu\gamma}}|\Phi_{\mathbf{k}_{\nu+1,\gamma+1}}\rangle|^2$$

$$-\ln \prod_{\nu=0}^{J_j-1}\prod_{\gamma=0}^{J_l-1} |\langle \Phi_{\mathbf{k}_{\nu\gamma}}|\Phi_{\mathbf{k}_{\nu+1,\gamma}}\rangle|^2$$

$$-\ln \prod_{\nu=0}^{J_j-1}\prod_{\gamma=0}^{J_l-1} |\langle \Phi_{\mathbf{k}_{\nu\gamma}}|\Phi_{\mathbf{k}_{\nu,\gamma+1}}\rangle|^2 \Bigg], \qquad (81)$$

where $\mathbf{k}_{\nu\gamma}=\mathbf{k}_\perp + (\nu/J_j)\mathbf{G}^j + (\gamma/J_l)\mathbf{G}^l$.

In order to convert back to Cartesian coordinates we use the transformation law for a second-rank contravariant tensor, $\langle X_i X_j\rangle_c = H_{il}\langle S^l S^m\rangle_c H_{jm}$, where $H=\{\mathbf{L}_1,\mathbf{L}_2,\mathbf{L}_3\}$. Equations (77), (79), and (81) are all that is needed to calculate the average polarization and its quadratic fluctuations, as well as the correlations along different Cartesian directions (if the cell symmetry is orthorhombic or higher, Eq. (81) is not needed for computing the fluctuations.) In Appendix D we give the required modifications to deal with fractional filling.

B. Formulas involving a single k

We now turn to a different kind of discretized formulas, the so-called ''single-point'' formulas, such as Eq. (17). In the case of localization they were previously given for one dimension only.[34] In the present formalism the generalization to higher dimensions becomes straightforward. We will arrive at the ''single-point'' formulas starting from the expressions derived in the previous section, which involve averages over a grid of **k** points. The basic idea is to perform the many-body analog of a ''Brillouin zone folding.''[16] Let us start by discretizing the remaining integrals in Eqs. (77), (79), and (81); the expression for the average polarization, for example, becomes

$$\langle \mathbf{P}_{\mathrm{el}}\rangle \cdot \mathbf{G}^l \simeq -\frac{q_e}{V}\frac{1}{J_i J_j}\mathrm{Im}\,\ln \prod_{\mu=0}^{J_i-1}\prod_{\nu=0}^{J_j-1}\prod_{\gamma=0}^{J_l-1} \langle \Phi_{\mathbf{k}_{\mu\nu\gamma}}|\Phi_{\mathbf{k}_{\mu\nu,\gamma+1}}\rangle. \qquad (82)$$

In the following we will assume a uniform spacing along all the reciprocal lattice directions: $\mathbf{k}_{\mu\nu\gamma}=\tilde{\mathbf{k}}+(\mu/J_i)\mathbf{G}^i +(\nu/J_j)\mathbf{G}^j+(\gamma/J_l)\mathbf{G}^l$ and $\tilde{\mathbf{k}}$ is fixed (usually $\tilde{\mathbf{k}}=0$). Next we build an *ansatz* wave function[34] $\tilde{\Psi}_{\tilde{\mathbf{k}}}$ containing $\tilde{N}=JN$ electrons as the antisymmetrized product of the $J=J_1 J_2 J_3$ separate N-electron wave functions $\Psi_{\mathbf{k}_{\mu\nu\gamma}}$:

$$\tilde{\Psi}_{\tilde{\mathbf{k}}}=\frac{1}{\sqrt{\tilde{N}!N!^J}}\sum_P (-1)^P P\Psi(1,\ldots,\tilde{N}), \qquad (83)$$

where the sum is over all the permutations P of the $\tilde{N}$ particle coordinates, $(-1)^P$ is the parity of the permutation, and

$\Psi(1,\ldots,\tilde{N})=\Pi_{\mu\nu\gamma}\Psi_{\mathbf{k}_{\mu\nu\gamma}}$. It is easy to verify that $\tilde{\Psi}_{\tilde{\mathbf{k}}}$ obeys $\tilde{\mathbf{k}}$ boundary conditions on the larger cell containing $\tilde{N}$ electrons. Using the fact that the grid is uniform, it can be shown that for even N this leads to [compare with Eq. (16) in Ref. 34, which deals with the 1D case]:

$$\prod_{\mu\nu\gamma} \langle \Phi_{\mathbf{k}_{\mu\nu\gamma}}|\Phi_{\mathbf{k}_{\mu\nu,\gamma+1}}\rangle = \langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle, \qquad (84)$$

where $\tilde{\mathbf{G}}^l=\mathbf{G}^l/J_l$ is a basis vector of the reciprocal of the larger cell, and $\hat{\tilde{\mathbf{X}}}$ is the center of mass position operator of the $\tilde{N}$-electron system. Comparison with Eq. (82) yields

$$\langle \mathbf{P}_{\mathrm{el}}\rangle \cdot \tilde{\mathbf{G}}^l \simeq -\frac{q_e}{\tilde{V}}\mathrm{Im}\,\ln\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle. \qquad (85)$$

Similarly, discretizing the integrals in Eq. (79) and using Eq. (84), we find

$$\langle (S^l)^2\rangle_c \simeq -\frac{1}{(2\pi)^2}\frac{J_l}{J_i J_j}\ln|\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle|^2. \qquad (86)$$

The quantity on the lhs pertains to the N-particle system, whereas those on the rhs pertain to the $\tilde{N}$-particle system. However, if we choose $J_1=J_2=J_3$ and use $\langle X_i X_j\rangle_c/N \simeq \langle \tilde{X}_i \tilde{X}_j\rangle_c/\tilde{N}$ together with the transformation law between Cartesian and scaled coordinates, we find $\langle \tilde{S}^i\tilde{S}^j\rangle_c =J_1\langle S^i S^j\rangle_c$. Equation (86) then becomes

$$\langle (\tilde{S}^l)^2\rangle_c \simeq -\frac{1}{(2\pi)^2}\ln|\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle|^2, \qquad (87)$$

where all the quantities are now explicitly written for the $\tilde{N}$-particle system.

The following relation can be derived along the same lines as Eq. (84):

$$\prod_{\mu\nu\gamma} \langle \Phi_{\mathbf{k}_{\mu\nu\gamma}}|\Phi_{\mathbf{k}_{\mu\nu+1,\gamma+1}}\rangle = \langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^j\cdot\hat{\tilde{\mathbf{X}}}}e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle. \qquad (88)$$

Discretizing the remaining integral in Eq. (81) and again setting $J_1=J_2=J_3$, we obtain, using Eqs. (84) and (88),

$$\langle \tilde{S}^j\tilde{S}^l\rangle_c \simeq -\frac{1}{2(2\pi)^2}[\ln|\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^j\cdot\hat{\tilde{\mathbf{X}}}}e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle|^2$$

$$-\ln|\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^j\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle|^2$$

$$-\ln|\langle \tilde{\Psi}_{\tilde{\mathbf{k}}}|e^{-\iota\tilde{\mathbf{G}}^l\cdot\hat{\tilde{\mathbf{X}}}}|\tilde{\Psi}_{\tilde{\mathbf{k}}}\rangle|^2]. \qquad (89)$$

Equations (85), (87), and (89) are the desired ''single-point'' formulas. In Appendix D we modify them for the case of fractional filling.

In one dimension the above formulas become particularly simple, and we recover the results of Refs. 17 and 34. In Cartesian coordinates ($\tilde{X}=\tilde{S}\tilde{L}$ and $\tilde{G}=2\pi/\tilde{L}$) they are

$$\langle \tilde{X}\rangle \simeq -\frac{\tilde{L}}{2\pi}\,\mathrm{Im}\,\ln\langle \tilde{\Psi}_{\tilde{k}}|e^{-\iota(2\pi/\tilde{L})\hat{\tilde{X}}}|\tilde{\Psi}_{\tilde{k}}\rangle, \qquad (90)$$

which is equivalent to Eq. (18), and

$$\langle \Delta \tilde{X}^2 \rangle \simeq -\frac{\tilde{L}^2}{(2\pi)^2} \ln |\langle \tilde{\Psi}_{\tilde{k}} | e^{-\iota (2\pi/\tilde{L}) \hat{\tilde{X}}} | \tilde{\Psi}_{\tilde{k}} \rangle|^2, \quad (91)$$

which, together with Eq. (30), gives Eq. (19).

C. Interpretation of the ''single-point'' formulas

In the previous section we derived ''single-point'' formulas for the first two moments of the polarization, starting from discretized expressions involving an average over boundary conditions. In order to switch between the two descriptions, an *ansatz* wave function $\tilde{\Psi}_{\tilde{k}}$ given by Eq. (83) was used. However, this is strictly valid only if the $\tilde{N}$ particles are not correlated with each other. In that case $\tilde{\Psi}_{\tilde{k}}$ becomes a Slater determinant of one-electron orbitals, and the procedure leading to the ''single-point'' formulas (''Brillouin zone folding'') does not involve any further approximations (see Refs. 16 and 17).

For a correlated state of many particles the situation is rather different. In that context the single-point formulas were originally proposed in one dimension for an *arbitrary* correlated $\tilde{N}$-electron wave function $\tilde{\Psi}_{\tilde{k}=0}$ with periodic boundary conditions over a cell of size $\tilde{L}$;[17,34] however, such a wave function in general will not obey Eq. (83). The derivation of the previous section allows us to assess the approximations involved: these have to do with the extent to which a wave function given by Eq. (83) differs from the fully correlated wave function. The key quantity to consider is the correlation length L_{corr}, which quantifies the range over which the particles are correlated (in addition to any long range order which is included in the mean field potentials ''seen'' by each particle). The fact that the correlations are short ranged has been termed ''nearsightedness'' by Kohn in a recent paper;[57] in fact, in an insulator, for example, one expects the longest range correlations to be of the van der Waals type, which decay as $1/r^6$ in the energy and as $1/r^3$ in the wave function.[39,58] The basic assumption underlying Eq. (83), namely that the $\tilde{N}$ electrons only correlate in groups of N at a time,[34] is consistent with the principle of nearsightedness. This is the justification for applying the single-point formulas to a correlated insulating wave function, provided that the cell is large enough.

From this perspective it becomes clear that the two types of formulas derived in Secs. VII A and VII B constitute different approximations to the same continuum expressions, which involve an average over all twisted boundary conditions. In both approaches one must choose cells with linear dimensions greater than L_{corr}: in the latter approach one must use a single large cell with size JV in order to have the same level of accuracy as J independent calculations each of size V using the former approach.

From a practical point of view, there are two possible ways to proceed: either perform several calculations with different twisted boundary conditions on a smaller cell and then use the formulas of Sec. VII A, or perform a single calculation on a large cell and then use the formulas of Sec. VII B. Although the two approaches are comparable in terms of accuracy, the former approach should be more efficient, because the available many-body algorithms do not scale linearly with the number of particles. On the other hand, the averaging over boundary conditions may be more cumbersome to implement in practice.

VIII. CONCLUSIONS

We have presented in this work a unified theory of electronic polarization and localization in bulk insulators. The central quantity in the formalism is the cumulant generating function $\ln C(\boldsymbol{\alpha})$ defined in Eq. (32); it provides a systematic procedure for extracting from the **k**-dependence of the ground state insulating wave function $\Psi_{\mathbf{k}}$ the moments of a properly defined distribution $p(\mathbf{X})$ for the electronic center of mass. In complete analogy with the case of a confined system, this distribution is simply related to the quantum distribution of the ground state polarization, via Eq. (29).

Several seemingly disparate ideas regarding electronic polarization and localization in insulators[8,29,34] are brought together quite naturally using the generating function approach. In particular, it shows the connection between the Berry phase theory of polarization,[12–16] as well as the related approach to localization,[33,34,36] and Kohn's theory of localization in the insulating state.[8,9] A key quantity is the electronic localization length ξ_i, which is defined in terms of the experimentally measurable mean-square fluctuation of the polarization [Eqs. (30) and Table I], making contact with the work of Kudinov.[29] In the thermodynamic limit it agrees with the localization length defined in Ref. 34 for 1D systems [Eq. (19)]. Furthermore, the generating function formalism also reveals a very close formal analogy between Kohn's localized many-electron functions $\Psi_{\mathbf{M}}$ and the maximally localized one-electron Wannier functions defined by Marzari and Vanderbilt:[33] the former can be viewed as maximally localized many-body Wannier functions (see Sec. VI B and Appendix B). Moreover, in the same way that the quadratic spread of the functions $\Psi_{\mathbf{M}}$ is a measure of the mean-square fluctuations of the bulk polarization, the gauge-invariant part of the spread of the Wannier functions (which in 1D systems equals the spread of the maximally localized Wannier functions) measures the same quantity for uncorrelated insulators. The fluctuation-dissipation relation can be used to derive an inequality [Eq. (52)] between the polarization fluctuations and the minimum energy gap for optical absorption in an insulator. The present approach also provides some extra insight into the appearence of a ''quantum of polarization'' in periodic insulators,[12] which is seen to be related to the localized nature of the insulating wave function (see Appendix C).

The localization length seems to play a role in the theory of insulators similar to that of the Drude weight in the theory of ideal conductors: the latter measures how ''free'' the ''free charges'' in a perfect conductor are, whereas the former measures how localized the ''bound charges'' in an insulator are. Interestingly, both quantities can be expressed as second derivatives with respect to the twisted boundary conditions (see Table I), which play a crucial role in the formalism. As discussed in Sec. VI C, *bulk properties* of insulators are rather insensitive to the boundary conditions, unlike the properties of conductors. Nevertheless, the insulating *wave function* itself is not insensitive to the boundary

conditions;[25] on the contrary, the derivatives of $\ln C(\boldsymbol{\alpha})$, which measure the **k**-averaged change with **k** in $\Phi_{\mathbf{k}} = e^{-\imath \mathbf{k} \cdot \mathbf{X}} \Psi_{\mathbf{k}}$ as the boundary condition on $\Psi_{\mathbf{k}}$ is twisted, contain quantitative information about basic properties of the insulator: in particular, the first derivative gives the average macroscopic polarization, and the second derivative gives its mean-square fluctuation. Both quantities have a geometrical interpretation: the former is a Berry phase on a manifold of quantum states parametrized by the twisted boundary conditions[12,14] [Eq. (38)], and the latter is a metric on the same manifold [Eq. (42)].

The generating function approach also leads naturally to discretized formulas that can be used to compute the polarization and the localization in many-body numerical calculations in any number of dimensions. Two alternative kinds of expressions exist: those involving wave functions computed on a uniform grid of **k** points [Eqs. (77), (79), and (81)], and those involving a single wavefunction with a fixed boundary condition $\widetilde{\mathbf{k}}$ [Eqs. (85), (87), and (89)]. The present derivation clearly shows how the two types of formulas are related to one another.

APPENDIX A: LOCALIZATION LENGTH AND SPREAD OF THE ONE-ELECTRON WANNIER FUNCTIONS

In Sec. VI B it was shown that the width of the electronic center of mass distribution $p(X_i)$, arising from the maximally-localized many-body Wannier function $\Psi_{\mathbf{M}}$, is $\sqrt{N} \xi_i$. In this Appendix we work in the independent-electron framework, in which the usual one-electron Wannier functions are defined, and investigate the relation between their spread and the localization length ξ_i (i.e., the root-mean-square fluctuation of the polarization). It is clear that since the spread [Eq. (14)] is gauge-dependent, it cannot relate directly to any measurable quantity. One might have guessed that, as happens with the many-body Wannier functions, the physically meaningful quantity would be the spread of the maximally localized Wannier functions. Building upon the results of Ref. 33, we show here that the gauge-invariant part Ω_{I} of the spread of the occupied Wannier functions [Eq. (15)] measures the mean-square fluctuation of the bulk polarization.

The proof follows from the fluctuation-dissipation theorem: for a crystalline insulator in the independent-electron approximation, the real part of the optical conductivity due to interband vertical transitions is given by[50]

$$\mathrm{Re}\,\sigma_{ij}(\omega) = \frac{\pi f q_e^2}{m_e^2 \hbar \omega} \sum_{n=1}^{M} \sum_{m=M+1}^{\infty}$$

$$\times \int_{\mathrm{BZ}} \frac{d\mathbf{k}}{(2\pi)^3} p_{nm,\mathbf{k}}^i p_{mn,\mathbf{k}}^j \delta(\omega_{mn}^{\mathbf{k}} - \omega), \quad (A1)$$

where f is the occupation number of states in the valence band (in spin-degenerate systems $f=2$), $\mathbf{p}_{nm,\mathbf{k}} = -\imath \hbar \langle \psi_{n\mathbf{k}} | \boldsymbol{\nabla} | \psi_{m\mathbf{k}} \rangle$, and $\hbar \omega_{mn}^{\mathbf{k}} = \epsilon_{m\mathbf{k}} - \epsilon_{n\mathbf{k}}$ is the difference between single-particle energies. Instead of Eq. (46) we now have $\mathbf{p}_{nm,\mathbf{k}} = m_e \omega_{mn}^{\mathbf{k}} \langle u_{n\mathbf{k}} | \partial_{\mathbf{k}} u_{m\mathbf{k}} \rangle$, and following similar steps as in Sec. IV for the many-body case we find, using Eq. (16),

$$\frac{\hbar}{\pi f q_e^2} \int_0^\infty \frac{d\omega}{\omega} \mathrm{Re}\,\sigma_{ij}(\omega) = \frac{1}{(2\pi)^3} \int_{\mathrm{BZ}} d\mathbf{k}\, g_{ij}(\mathbf{k}). \quad (A2)$$

Combining with Eqs. (15) and (49), using $v/(fV) = M/N$, and taking the trace, we obtain the fluctuation-dissipation relation:[59]

$$\Omega_{\mathrm{I}} = \frac{\hbar v}{\pi f q_e^2} \int_0^\infty \frac{d\omega}{\omega} \mathrm{Tr}\,\mathrm{Re}\,\sigma(\omega) = M \lim_{N \to \infty} \frac{\langle \Delta \mathbf{X}^2 \rangle}{N}. \quad (A3)$$

Comparison with Eqs. (30) shows that $\Omega_{\mathrm{I}}/M = \Sigma_{i=1}^3 \xi_i^2$, which is the multidimensional generalization of a result obtained in Ref. 34. It has been found that typically Ω_{I} accounts for most of the spread Ω; for instance, in semiconductors it usually accounts for more than 90%.[33] Thus it is justifiable to view ξ_i^2 as an estimate of the average spread along the ith direction of the occupied Wannier functions (in one dimension it is actually the average spread of the maximally localized functions[33]). This conclusion agrees with the analysis in Appendix 1 of Ref. 8: there, the distribution $p(X)$ of the electronic center of mass of a 1D noninteracting insulating system was studied, with the result that X is localized in an interval of width $\sqrt{N}b$, where ''b is a length of the order of the range of the Wannier function.'' It is clear from the results of Sec. VI B and the previous discussion that the gauge-invariant ξ is precisely that length b.

It is straightforward to check that for noninteracting crystalline insulators, Eq. (52) becomes, after summing over all d Cartesian directions, $\Omega_{\mathrm{I}}/M \leq d\hbar^2/(2m_e E_{\mathrm{g}})$, where E_{g} is the minimum direct gap over the Brillouin zone. Hence in general the inequality involves the gauge-invariant part of the spread. Since in one dimension this equals the average spread of the maximally localized Wannier functions, we recover Kivelson's original result.[60]

APPENDIX B: SPREAD OF THE MANY-BODY WANNIER FUNCTIONS

In Sec. VI B we introduced the functions $W_{\mathbf{M}}$, which play a role in the many-body theory of polarization similar to that of the Wannier functions in the independent-electron theory. Here we will show that this formal analogy carries over to considerations about the spread of those functions. As mentioned in Sec. II, in the independent-electron framework the quadratic spread Ω of the occupied Wannier functions can be decomposed into a sum of two positive terms, one of them (Ω_{I}) gauge-invariant;[33] hence $\Omega_{\mathrm{I}} \leq \Omega$ in any gauge. Follow-

ing similar steps as in Ref. 33, here we will derive the corresponding result for the many-body functions $W_\mathbf{M}$. First we define their gauge-dependent spread by analogy with Eq. (14):

$$\Omega^{(\mathrm{W})} = \langle W_\mathbf{0} | \hat{\mathbf{X}}^2 | W_\mathbf{0} \rangle - \langle W_\mathbf{0} | \hat{\mathbf{X}} | W_\mathbf{0} \rangle^2 = - \partial_{\boldsymbol{\alpha}^2}^2 \ln C_\mathrm{W}(\boldsymbol{\alpha}) |_{\boldsymbol{\alpha}=0},$$
(B1)

where $C_\mathrm{W}(\boldsymbol{\alpha})$ is given by Eq. (63). Next we show that the role of Ω_I is played by $\langle \Delta \mathbf{X}^2 \rangle = - \partial_{\boldsymbol{\alpha}^2}^2 \ln C(\boldsymbol{\alpha}) |_{\boldsymbol{\alpha}=0}$: taking this derivative inside the integral in Eq. (32), discretizing that integral,[52] and comparing with Eq. (63), this becomes

$$\langle \Delta \mathbf{X}^2 \rangle = \langle W_\mathbf{0} | \hat{\mathbf{X}}^2 | W_\mathbf{0} \rangle - \frac{1}{N_c} \sum_\mathbf{k} (\imath \langle \Phi_\mathbf{k} | \partial_\mathbf{k} \Phi_\mathbf{k} \rangle)^2. \quad \text{(B2)}$$

Now we use the relation

$$\imath \langle \Phi_\mathbf{k} | \partial_\mathbf{k} \Phi_\mathbf{k} \rangle = \sum_\mathbf{M} e^{-\imath \mathbf{k} \cdot \mathbf{R}_\mathbf{M}} \langle W_\mathbf{M} | \hat{\mathbf{X}} | W_\mathbf{0} \rangle, \quad \text{(B3)}$$

which is the many-body analog of Eq. (5) of Ref. 33 and can be derived in the same way. Substituting into Eq. (B2) gives

$$\langle \Delta \mathbf{X}^2 \rangle = \langle W_\mathbf{0} | \hat{\mathbf{X}}^2 | W_\mathbf{0} \rangle - \sum_\mathbf{M} | \langle W_\mathbf{M} | \hat{\mathbf{X}} | W_\mathbf{0} \rangle |^2, \quad \text{(B4)}$$

and comparison with Eq. (B1) yields the desired result: $\langle \Delta \mathbf{X}^2 \rangle \leqslant \Omega^{(\mathrm{W})}$.

There is, however, an important difference with respect to the single-electron Wannier functions: in Ref. 33 it was shown that it is only in one dimension that the maximally localized one-electron Wannier functions have a spread $\min[\Omega] = \Omega_\mathrm{I}$, whereas in higher dimensions $\min[\Omega] > \Omega_\mathrm{I}$, i.e., Ω_I is strictly a lower bound to Ω. As discussed in Sec. VI B, according to Kohn[8,9] a gauge can be chosen where the functions $W_\mathbf{M}$ have an exponentially small overlap with one another in the high-dimensional configuration space, in which case only the term $\mathbf{M} = 0$ survives in the sum on the rhs of Eq. (B4). Therefore we conclude that for an insulator in any number of dimensions we have $\min[\Omega^{(\mathrm{W})}] \doteq \langle \Delta \mathbf{X}^2 \rangle$, where for any finite size the corrections due to the exponentially small overlaps make the lhs slightly larger than the rhs. Thus Kohn's functions $\Psi_\mathbf{M}$ are the maximally localized many-body Wannier functions $W_\mathbf{M}$, since as $V \to \infty$ the spread $\langle \Delta \mathbf{X}^2 \rangle$ of the gauge-invariant distribution $p(\mathbf{X})$ obtained from $\Psi_\mathbf{M}$ [Eq. (68)] becomes the minimum of the spread $\Omega^{(\mathrm{W})}$ of the gauge-dependent distribution $p_\mathrm{W}(\mathbf{X})$ obtained from $W_\mathbf{M}$ [Eq. (62)]. This point can be further elucidated by noting, in the spirit of Refs. 33 and 51, that in the limit of $V \to \infty$ the functions $\Psi_\mathbf{M}$ become eigenfunctions of the operator $\hat{\mathbf{X}}$ projected onto the occupied manifold.[61]

APPENDIX C: THE QUANTUM OF POLARIZATION

Here we will discuss the relation between the quantum of polarization[12–16] and the localized nature of a bulk insulating wave functions [8,9] (see Fig. 2) (a related discussion, regarding the closely related phenomenom of quantized charge transport in insulators, can be found in Ref. 62.) Let us consider a periodic system of volume L^3, which for simplicity we take to be cubic. If the external potential acting on the

electrons is changed adiabatically along an insulating closed path parametrized by λ ($\hat{H}^{(\lambda=1)} = \hat{H}^{(\lambda=0)}$), then, since the hamiltonian comes back to itself, the net effect, as far as the wave function is concerned, has to be either (i) each $\Psi_\mathbf{M}$ returns to itself or (ii) there is a rigid translation of all the $\Psi_\mathbf{M}$ in configuration space by the same amount, which can be described by a uniform shift of their indices: $\mathbf{M} \to \mathbf{M} + (n_1, n_2, n_3)$.

As far as charge transport is concerned, the important observation is that, since the system remains insulating throughout the path, the regions $\mathcal{R}_\mathbf{M}$ remain disconnected, so that no charge can flow between them during the adiabatic motion. The resulting integrated current flowing through the system during the cycle [which, according to Eqs. (2) and (3), measures the change in polarization] can then be inferred from Figs. 2 and 3, which show that the regions $\mathcal{R}_\mathbf{M}$ are labeled by the center of mass of the electrons, and that one can go from one region to the next by moving any one electron across the length L of the periodic system. The smallest nonzero change in the average polarization along the ith direction is given by the smallest nonzero shift in the distribution $p(X_i)$ ($n_i = \pm 1$), and is seen to equal $|q_e| L / V = |q_e| / L^2$, which is the quantum.[12,14]

It should be noted that strictly speaking the exact quantization of charge transport in an insulating system with periodic boundary conditons only occurs in the thermodynamic limit.[24,25] In fact, the quantization was established for a finite system only after averaging over all twisted boundary conditions; when using periodic boundary conditions, there are exponentially small corrections.[25] This is consistent with Kohn's picture that for any finite V the regions $\mathcal{R}_\mathbf{M}$ are not completely disconnected but have an exponentially small overlap, which allows for a correspondingly small charge flow between neighboring regions, thus destroying the exact quantization.

APPENDIX D: THE CASE OF FRACTIONAL FILLING

The formulas given in the text need to be generalized in order to deal with correlated systems which have in the ground state a noninteger number of electrons per primitive cell, and yet are insulating. This can happen in many 1D models,[36,63] but it appears that in higher dimensions the Mott transition to an insulating state is usually accompanied by a breaking of the symmetry (e.g., a charge-density wave) that restores integer filling.[64] Therefore we shall restrict the ensuing analysis to one dimension, as done in Refs. 28 and 36.

Aligia[28] has shown that in cases where there is a fractional number of electrons per primitive cell, the limits of integration over k in the Berry phase formula for the polarization difference [Eq. (12)] need to be modified, from which it follows that the "single-point" formulas for the polarization and localization derived in Refs. 17 and 34 also need to be changed. For the purposes of the present paper it is straightforward to modify the integral over k in the definition of $\ln C(\alpha)$, and from that derive the required modifications to the discretized numerical formulas.

Let us consider a 1D system with a simulation cell of size L and n/l electrons per unit cell, where n/l is an irreducible fraction. Following Ref. 28, we modify the cumulant gener-

ating function, Eq. (32), as follows:

$$\ln C(\alpha) = \frac{1}{l} \int_0^{2\pi l/L} dk \, \ln \mathcal{C}(k, \alpha). \tag{D1}$$

In order to obtain the discretized formulas, we just need to retrace the steps taken in Secs. VII A and VII B, with the above modification. Discretizing the interval $0 \leq k \leq 2\pi l/L$ into a uniform row of $J+1$ points k_γ, we find that Eq. (77) changes to

$$\langle X \rangle \simeq -\frac{L}{2\pi l} \text{Im} \ln \prod_{\gamma=0}^{J-1} \langle \Phi_{k_\gamma} | \Phi_{k_{\gamma+1}} \rangle, \tag{D2}$$

which agrees with the result quoted in Ref. 28. Similarly, Eq. (79) becomes:

$$\langle \Delta X^2 \rangle \simeq -\frac{JL^2}{(2\pi l)^2} \ln \left| \prod_{\gamma=0}^{J-1} \langle \Phi_{k_\gamma} | \Phi_{k_{\gamma+1}} \rangle \right|^2. \tag{D3}$$

As for the modified single-point formulas,[36] it is straightforward to verify, applying the approach of Sec. VII B to the previous two equations, that they are the same as Eqs. (90) and (91), except for the substitution $\tilde{L} \to \tilde{L}/l$.

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[41] The second form of Eq. (31) will be the most useful in what follows. We note that it is the many-body analog of a quantity introduced in the single-particle context in Ref. 33 [see Eq. (25) of Ref. 33].

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finite V the fluctuations are in general finite even for a conductor, at any given $\mathbf{k}$. It is also clear that the crossing of low-lying levels at a finite number of $\mathbf{k}$ points in a conductor at finite V (Ref. 47) does not affect our conclusion regarding the divergence of Eq. (49) as $V \rightarrow \infty$.

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[52] N_c equals the number of cells in the periodic crystal; when that number goes to infinity the sum is replaced by an integral: $(1/N_c) \Sigma_{\mathbf{k}} \rightarrow [v/(2\pi)^3] \int d\mathbf{k}$ (in the correlated case the unit cell volume v is replaced by the supercell volume V).

[53] According to Eq. (61) $\Psi_{\mathbf{k}=0} \propto \Sigma_{\mathbf{M}} W_{\mathbf{M}}$. Since $\Psi_{\mathbf{k}=0}$ is gauge invariant apart from an overall phase, if the $W_{\mathbf{M}}$ were nonoverlapping, they would also have to be gauge invariant, apart from the same overall phase.

[54] In Appendix 1 of Ref. 8 a definition for the center-of-mass distribution of a periodic system was proposed, which amounts to replacing the δ function in Eq. (25) by a suitably defined "periodic δ function." For a 1D insulator such definition yields the same distribution (for large L) as the characteristic function $C(\alpha)$. However, it does not give an individual peak, since the resulting distribution is periodic by construction; as a consequence, in three dimensions it yields a delocalized center of mass even for insulators, because the contributions from different $\Psi_{\mathbf{M}}$ overlap with one another (this is the "strange situation" alluded to in Appendix 1 of Ref. 8); our definition in terms of the characteristic function, on the other hand, generates one localized distribution $p(\mathbf{X})$ from each $\Psi_{\mathbf{M}}$ separately (see overlapping dotted curves in Fig. 3), which retains the desired physical interpretation in any number of dimensions.

[55] Notice that Eq. (74) can be rewritten without the logarithm: $\langle S^l \rangle = (\iota/2\pi) \int d\mathbf{q} \; \partial_{t_l} C(\mathbf{q},\mathbf{t})|_{\mathbf{t}=0}$. However, if we discretize the derivative in the integrand in the straightforward way, the resulting expression will no longer be gauge invariant.

[56] However, it is easy to see that $-\partial^2_{\alpha_i \alpha_j} \text{Re} \ln C(\mathbf{k},\boldsymbol{\alpha})|_{\boldsymbol{\alpha}=0} = G_{ij}(\mathbf{k})$, so that taking the real part of Eq. (78) yields a gauge-invariant approximation to the metric tensor in scaled coordinates.

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[59] Here all the filled bands are being treated as a *composite band* (in the terminology of Ref. 33), to which corresponds a single Ω_I; in general different Ω_I's can be ascribed to each separate group of composite bands (Ref. 33), but then the connection to the polarization fluctuations, which involve all the valence electrons, appears to be lost.

[60] In Ref. 51 the Wannier functions were obtained as the eigenfunctions of the position operator projected into a given band. As shown in Ref. 33, in 1D this choice minimizes the quadratic spread. Notice also that Kivelson considered disordered insulators; in that case E_g is the absolute gap.

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