

## Macroscopic polarization as a geometric quantum phase: Many-body formulation

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During past decades, concepts about the electrostatics of infinite systems have been a challenge for theoretical physicists. In particular, the question of whether the absolute macroscopic polarization or the difference between the polarizations of two states of an insulating crystal is a well-defined bulk property has remained a controversial one. Recently, King-Smith and Vanderbilt, and Resta have provided an approach in terms of the geometric Berry phase of electronic orbitals in an independent-particle approximation. Here we extend the derivation of Niu and Thouless for quantized charge transport to the case where the quantum adiabatic evolution is noncyclic, and we show how this polarization difference can be written in terms of a Berry phase for a system with many-body interactions. We also discuss the origin and magnitude of the “quantum uncertainty” that appears when a path-independent gauge is used to compute those geometric quantum phases. This geometric viewpoint not only helps us understand the issues raised above but provides a mathematical method to compute polarizations in a many-body framework.

### I. INTRODUCTION

The electric polarization is a quantity which plays a central role in the dielectric phenomena of matter.<sup>1-4</sup> The macroscopic polarization  $\mathbf{P}$  is the average value of the polarization density field  $\mathcal{P}(\mathbf{r})$ , which satisfies the equation

$$\nabla \cdot \mathcal{P}(\mathbf{r}) = -\rho(\mathbf{r}), \quad (1)$$

where  $\rho(\mathbf{r})$  is the charge density of the system. In a finite system, if  $\mathcal{P}(\mathbf{r})$  vanishes outside the sample, it is straightforward to show that  $\mathbf{P}$  is the dipole moment of  $\rho(\mathbf{r})$

$$\mathbf{P} = \frac{1}{\Omega} \int_{\Omega} d^3r \mathcal{P}(\mathbf{r}) = \frac{1}{\Omega} \int_{\Omega} d^3r \mathbf{r} \rho(\mathbf{r}). \quad (2)$$

Thus  $\mathbf{P}$  is uniquely determined by  $\rho(\mathbf{r})$  and the condition  $\mathcal{P}(\mathbf{r}) = 0$  outside the volume of the sample  $\Omega$ .

In an extended system, however, the interpretation of this integral suffers from difficulties. In particular, there has been much debate and discussion<sup>5,6</sup> about whether or not it is possible to uniquely define a “macroscopic bulk polarization” which is a property only of the interior of the material independent of surface termination. The difficulty originates as a result of the position vector  $\mathbf{r}$  in the definition above: a charge distribution on the boundary of the finite system can lead to a contribution to the polarization per unit volume  $\mathbf{P}$  which does not vanish in the thermodynamic limit in which the finite system is taken to infinite size. This corresponds to the fact that the total polarization of a finite sample depends upon the charge state of the surfaces; therefore it cannot be a bulk property. Nevertheless, in an insulating system one can define the change in polarization  $\Delta\mathbf{P}$ , e.g., due to a phonon mode or a ferroelectric displacement.

In quantum insulators a charge disturbance confined to the surface region cannot propagate its effects into the bulk region provided the macroscopic electric field  $\mathbf{E}_{\text{mac}}$  is kept vanishing (this concept is developed in more detail in Sec. II where the meaning of bulk and surface regions is cleared up). Since the surface charge cannot flow in an insulator, the change  $\Delta\mathbf{P}$  must be associated only with the bulk (in null macroscopic electric field  $\mathbf{E}_{\text{mac}}$ ).

One can attempt to derive expressions for quantities such as the polarization directly from information on a single periodic cell in an infinite insulating crystal. Such attempts have led to much controversy and disagreements. It is easily demonstrated<sup>5,7</sup> that it is impossible to derive the polarization strictly from the charge density in the unit cell, since the dipole moment depends upon the choice of the cell. However, the change in polarization between any two physical states  $\Delta\mathbf{P}$  is uniquely given by the change in dipole moment of a unit cell plus a surface integral which physically is the contribution of the polarization current which flows through the cell.<sup>5</sup> This quantity ( $\Delta\mathbf{P}$ ) was shown to be independent of the choice of cell; however, no prescription for calculating the surface integral was given.<sup>5</sup>

Another approach is to consider an infinite system with finite wavelength variations, i.e., electric fields, currents, and charge densities which are periodically varying in space. The properties are well known and can be described in terms of longitudinal and transverse electromagnetic phenomena. If one carries out perturbation expansions about a known equilibrium state, the dielectric phenomena can be described in terms of correlation functions of currents and densities at different time and position.<sup>8,9</sup> In quantum mechanics there are well known expressions for linear response tensors and it is possible, in principle, to take into account any order in perturbation theory. In this formalism it is essential to con-

sider finite wavelengths and take the proper limits as the wavelength is increased to infinity, in order to describe macroscopic phenomena.<sup>3,10</sup>

Recently<sup>11,12</sup> there has been a breakthrough in the calculation of polarization in crystalline dielectrics. An approach has been developed within the context of one-body theory with a formal structure which relies on concepts of topology and differential geometry.<sup>13</sup> In any single-body mean-field theory of an infinite periodic crystal, all physical quantities can be written as integrals over matrix elements as a function of  $\mathbf{k}$  in the Brillouin zone. The authors of Refs. 11 and 12 consider insulators where the integrals over filled bands are over the complete Brillouin zone and they take advantage of periodicity in  $\mathbf{k}$  space. Following them we define  $\xi = (\xi_1, \xi_2, \xi_3)$  to be the dimensionless reduced  $\mathbf{k}$  vector. The electronic contribution to the polarization difference  $\Delta\mathbf{P}_{\text{el}}$  ( $\Delta\mathbf{P} = \Delta\mathbf{P}_{\text{el}} + \Delta\mathbf{P}_{\text{ion}}$ , where  $\Delta\mathbf{P}_{\text{ion}}$  is the ionic contribution) between two states which are connected through a continuous adiabatic transformation characterized by the parameter  $\xi_4$  (in null macroscopic electric field  $\mathbf{E}_{\text{mac}}$ ) can be expressed in terms of a geometric quantum phase  $\gamma(C_i)$  (Berry's phase<sup>14</sup>)

$$\Omega_0 \mathbf{G}_i \cdot \Delta\mathbf{P}_{\text{el}} = 2e |\varepsilon_{ijk}| \int d\xi_j d\xi_k \gamma(C_i). \quad (3)$$

The  $(\xi_j; \xi_k)$  double integral is over the unit square  $[0,1] \times [0,1]$ ,  $\mathbf{G}_i$  are reciprocal unit vectors,  $\Omega_0$  is the unit cell volume,  $\varepsilon_{ijk}$  is the Levi-Civita tensor with indices running from 1 to 3 (no summation is assumed),  $e$  the electron charge, and

$$\gamma(C_i) = \oint_{C_i} \mathcal{A}(\xi) \cdot d\xi, \quad (4)$$

whose nonzero value reflects the fact that the periodic boundary conditions of the single-particle wave functions (Bloch condition) induce a nonsimply connected configuration space. The connection  $\mathcal{A}$  ("gauge field") is given by<sup>15</sup>

$$\mathcal{A}(\xi) = i \sum_{n=1}^{\bar{n}} \langle u_n(\xi) | \nabla_{\xi} u_n(\xi) \rangle. \quad (5)$$

$C_i$  is the contour of the unit square  $[0,1] \times [0,1]$  in the plane parallel to  $(\xi_i; \xi_4)$  for fixed  $(\xi_j; \xi_k)$ . The sum is performed over the  $\bar{n}$  filled bands, i.e., the lowest single-particle eigenstates whose periodic parts are given by  $u_n(\xi)$ . In the periodic gauge<sup>11</sup> Eq. (3) can be written in terms of the initial ( $\xi_4 = 0$ ) and final ( $\xi_4 = 1$ ) states ( $d^3\xi = d\xi_1 d\xi_2 d\xi_3$ )

$$\Omega_0 \mathbf{G}_i \cdot \Delta\mathbf{P}_{\text{el}} = 2e i \int d^3\xi \sum_{n=1}^{\bar{n}} \{ \langle u_n^1(\xi) | \partial_{\xi_i} u_n^1(\xi) \rangle - \langle u_n^0(\xi) | \partial_{\xi_i} u_n^0(\xi) \rangle \}, \quad (6)$$

where  $\partial_{\xi_i}$  stands for  $\frac{\partial}{\partial \xi_i}$ . These authors<sup>11,12</sup> showed that a two-point formula such as Eq. (6) determines  $\Delta\mathbf{P}_{\text{el}}$  to within a factor of  $\frac{2e}{\Omega_0} \mathbf{R}$ , where  $\mathbf{R}$  is any lattice vector.

There are several unsatisfactory aspects of the formulas as stated above. One is that they are restricted to single-body mean-field descriptions. A second one is the fact that the formulas for  $\mathbf{P}$  are subject to addition of any integral multiple of the quantum  $\frac{2e}{\Omega_0} \mathbf{R}$ . In a crystal with a large cell, or if one arbitrarily considers a cell larger than the primitive one, then the "quantum" is reduced. In particular, in an amorphous system it becomes infinitesimal, and the resulting polarization becomes completely arbitrary.

The purpose of this paper is to give full many-body expressions for the polarizations which define the polarization change in terms of a Berry's phase with no added "quantum," i.e., no uncertainty. The latter expressions require specification of the path in defining  $\Delta\mathbf{P}$ . When an expression is derived involving only the end points of the path ("two-point" formula), then the "quantum" arises. This is important in that it shows that the change in polarization itself is well defined and that it is only the particular expressions that are problematic. Finally, we show that the many-body two-point expressions, which intrinsically involve a large cell to treat particle correlations, have the same "quantum" as that for the primitive cell. Our many-body approach follows the key ideas of Niu and Thouless<sup>16</sup> for the quantization of charge transport in an insulator. Our derivation is based on two main concepts. The first regards the observation that for a quantum insulator charge cannot flow between the bulk and surface of the system, while the second is the theoretical technique of averaging over boundary conditions (i.e., over momentum vectors  $\mathbf{k}$ ). Actually the fact that for a given observable averaging or picking a particular ( $\mathbf{k}$ ) boundary condition leads to the same result (in the thermodynamic limit) can be considered the condition for this observable to be a bulk property.

## II. GEOMETRIC PHASE APPROACH: THE MANY-BODY FORMULATION

We start by defining our general model of the infinite crystal: First consider a finite system with a boundary surface whose macroscopic polarization  $\mathbf{P}$  is unambiguously given by Eq. (2), with the condition  $\mathbf{E}_{\text{mac}} = 0$  in the bulk (see Fig. 1); then we perform the thermodynamic limit. Our finite crystal has a perfectly periodic bulk region in the interior, and surface regions (see Fig. 2).

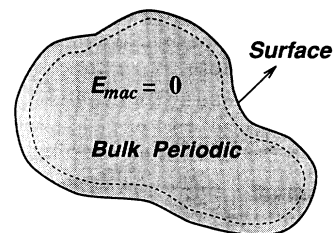


FIG. 1. Schematic of bulk plus surface regions for a general sample shape. The macroscopic electric field  $\mathbf{E}_{\text{mac}}$  is kept zero in the interior of the sample by *external* means.

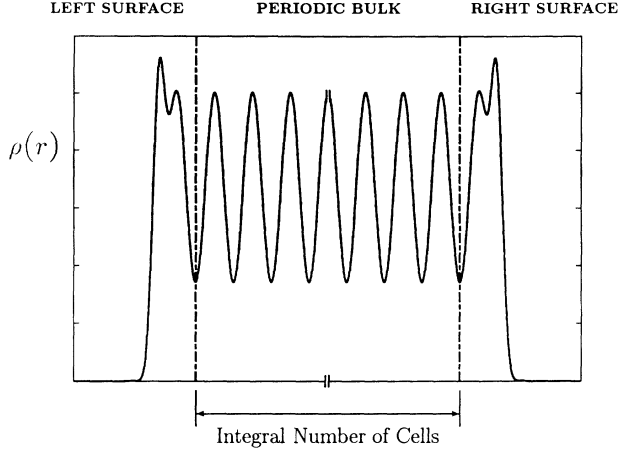


FIG. 2. 1D crystal (or parallel geometry) showing a possible partition (by imaginary dash-dotted lines) of the total charge into surface and bulk regions. In this case, from the density alone it is impossible to define an intrinsic bulk polarization.

Note that we do not make any assumptions about the surfaces; the only hypothesis is that the bulk region is insulating and has periodic density  $\rho(\mathbf{r})$ . Contrary to what happens in metals, the magnitude of  $\mathbf{E}_{\text{mac}}$  inside a dielectric is not in general zero; we impose this condition because we want the electrostatic potential to be periodic in the bulk. This is accomplished either grounding the surfaces (metallic surface) or adding *external* charges on the surfaces.

Suppose that we separate the surface from the bulk region by imaginary lines (as in Fig. 2). For the sake of clarity we illustrate the concepts with a one-dimensional (1D) crystal of linear dimension  $L$  and such that  $\int dr \rho(r) = 0$  (the dipolar moment of  $\rho$  is origin independent). Then if one attempts to divide the total density into bulk and surface parts,

$$\rho(r) = \rho_{\mathcal{L}}(r) + \rho_{\text{bulk}}(r) + \rho_{\mathcal{R}}(r), \quad (7)$$

$$\mathbf{P} = \frac{1}{L} \int_{-\frac{L}{2}}^{\frac{L}{2}} dr r \rho(r) = D_{\mathcal{L}} + \frac{1}{L} \int dr r \rho_{\text{bulk}}(r) + D_{\mathcal{R}}, \quad (8)$$

where  $D_{\mathcal{L},\mathcal{R}}$  is the left (right) surface region contribution of charge density  $\rho_{\mathcal{L},\mathcal{R}}(r)$  to the dipolar moment. It is clear that *both* surface and bulk contributions to  $\mathbf{P}$  depend upon the arbitrary definition of the boundaries so that neither is invariant. With this prescription for the partition, it is impossible to define an intrinsic bulk polarization from the density alone ( $\rho_{\text{bulk}}$ ) as shown before.<sup>5,7</sup> As can be easily realized, this argument also holds for differences  $\Delta\mathbf{P}$ , since a change in  $\mathbf{P}$  cannot be uniquely related to the bulk or surface terms. Of course,  $\Delta\mathbf{P}$  is definitely defined for the finite crystal, but additional arguments are needed to specify the change in  $\Delta\mathbf{P}$  as a bulk property.<sup>7</sup>

For an insulator we can define the bulk polarization

$\mathbf{P}_{\text{bulk}}$  of the finite crystal in a different way. The essential step is to realize that in an insulator one can make a separation of the charge density into bulk and surface parts *uniquely*. This is then sufficient to derive a unique bulk polarization  $\mathbf{P}_{\text{bulk}}$ . The crux of the proof is that *charge cannot flow* between the “bulk” and “surface” parts. This is the idea used by Kallin and Halperin<sup>17</sup> to show that piezoelectricity is a bulk effect; this division is also used in the derivations of King-Smith and Vanderbilt.<sup>11,18</sup>

For the finite crystal the bulk density can be defined as

$$\tilde{\rho}_{\text{bulk}}(r) = 2e \sum_{n,j} |W_n(r - ja)|^2 + \rho_{\text{ion}}(r), \quad (9)$$

where  $W_n$  are localized Wannier functions centered at  $ja$  (the sum over  $n$  extends over the occupied bands), and  $\rho_{\text{ion}}(r)$  represents the density of positive charges necessary to neutralize the electronic bulk charges [ $\int dr \tilde{\rho}_{\text{bulk}}(r) = 0$ ]. The surfaces are described by functions which are not periodic. They can be arbitrarily complex, but since the bulk is insulating, each surface region (denoted  $\mathcal{L}, \mathcal{R}$  in Fig. 3) has wave functions which are localized (exponentially) to one surface region. This can be proven to be the case based only upon the insulating character of the bulk. Therefore the total charge density is defined to be  $\rho(r) = \tilde{\rho}_{\mathcal{L}}(r) + \tilde{\rho}_{\text{bulk}}(r) + \tilde{\rho}_{\mathcal{R}}(r)$  (see Fig. 3). Now, for the finite crystal each distribution has well-defined dipole moments.

The generalization of the previous arguments to 3D is straightforward: in the following we will use the 3D notation. Once we have performed the aforementioned charge partition, we can find the polarization difference  $\Delta\mathbf{P}_{\text{bulk}}$  between two crystal states in the following way

$$\Delta\mathbf{P}_{\text{bulk}} = \frac{2e}{\Omega_0} \sum_{n=1}^{\tilde{n}} \int_{\Omega} d^3r \mathbf{r} [ |W_n^1(\mathbf{r})|^2 - |W_n^0(\mathbf{r})|^2 ] + \frac{1}{\Omega_0} \int_{\Omega_0} d^3r \mathbf{r} [ \rho_{\text{ion}}^1(\mathbf{r}) - \rho_{\text{ion}}^0(\mathbf{r}) ], \quad (10)$$

where superscripts 1 and 0 label each of the crystal states. The statements concerning charge partitioning are gen-

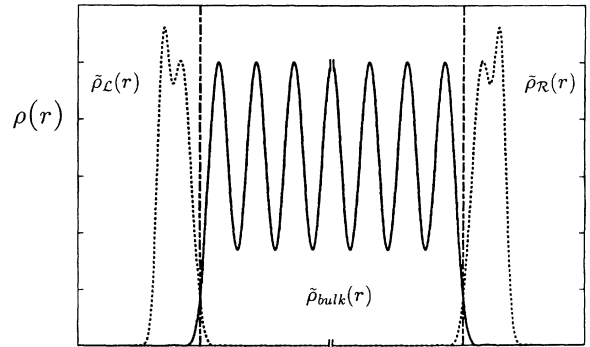


FIG. 3. Same geometry as in Fig. 2 but now the partition of the charge density is different. The charge cannot flow from the bulk to the surface regions. Under this assumption it is possible to define an intrinsic bulk polarization (up to a “quantum”).

eral and not restricted to single-body theory. In a milestone paper,<sup>19</sup> Kohn developed a characterization of the insulating state of matter from a many-body point of view. The essential property of its wave function is that it can be written as a sum of functions which are localized in disconnected regions of the multiparticle configuration space and have vanishing overlap. This is exactly the property needed to make a consistent generalization of the previous arguments to the many-body case.

Now we want to show, using elementary arguments from macroscopic electrostatics, that the change in bulk polarization due to a transformation performed in an insulating crystal is identical to the integrated bulk current. The starting point is the continuity equation which, because charge cannot flow between surface and bulk regions, is satisfied for each domain separately (with the condition that  $\mathbf{E}_{\text{mac}}$  is kept zero). After Eq. (1), the continuity equation for the bulk domain reads

$$\nabla \cdot \left( \mathbf{j}_{\text{bulk}} - \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} \right) = 0, \quad (11)$$

where  $\mathbf{j}_{\text{bulk}}$  is the bulk current density, and whose solution is

$$\mathbf{j}_{\text{bulk}} = \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} + \nabla \wedge \mathcal{M}_{\text{bulk}}; \quad (12)$$

$\mathcal{M}_{\text{bulk}}$  is the bulk magnetization vector which is a periodic function of the vector position  $\mathbf{r}$ . Then, the total integrated current per unit cell volume transported during the time  $T$  is given by

$$\begin{aligned} \mathbf{Q}_{\text{bulk}} &= \frac{1}{\Omega_0} \int_0^T dt \int_{\Omega_0} d^3r \mathbf{j}_{\text{bulk}} \\ &= \int_0^T dt \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} + \frac{1}{\Omega_0} \int_0^T dt \int_{\Omega_0} d^3r \nabla \wedge \mathcal{M}_{\text{bulk}}. \end{aligned} \quad (13)$$

We can transform the volume integral of the last term to a surface integral over the cell boundary  $\partial\Omega_0$  which vanishes (given the periodicity condition on  $\mathcal{M}_{\text{bulk}}$ )

$$\begin{aligned} \mathbf{Q}_{\text{bulk}} &= \int_0^T dt \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} \\ &\quad + \frac{1}{\Omega_0} \int_0^T dt \oint_{\partial\Omega_0} dS [\hat{\mathbf{n}} \wedge \mathcal{M}_{\text{bulk}}], \end{aligned} \quad (14)$$

where  $\hat{\mathbf{n}}$  is outward surface-normal unit vector. Then,

$$\mathbf{Q}_{\text{bulk}} = \Delta \mathcal{P}_{\text{bulk}} = \int_0^T dt \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} = \int_0^1 d\lambda \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial \lambda}, \quad (15)$$

where in the last equality we assume that during time  $T$  our system has continuously evolved from a state characterized by  $\lambda(0)=0$  to a different (in principle) state with  $\lambda(T)=1$ ,  $\lambda(t)$  being a scalar which parametrizes a path in the space of Hamiltonians. The previous relation is quite general provided one chooses carefully the boundary con-

ditions (zero  $\mathcal{P}$  and  $\mathcal{M}$  outside the sample) and assumes that the system remains insulating during the evolution (otherwise we cannot uniquely disentangle bulk from surface charges). We note that it is possible to define the change in polarization needed in Eq. (15) in terms of the dipolar moment of the charge inside a cell plus a contribution which represents the charge transferred across the cell boundary

$$\begin{aligned} \Delta \mathcal{P}_{\text{bulk}} &= \frac{1}{\Omega_0} \int_0^T dt \int_{\Omega_0} d^3r \mathbf{r} \frac{\partial \rho}{\partial t} \\ &\quad + \frac{1}{\Omega_0} \int_0^T dt \oint_{\partial\Omega_0} dS \mathbf{r} \left[ \hat{\mathbf{n}} \cdot \frac{\partial \mathcal{P}_{\text{bulk}}}{\partial t} \right]. \end{aligned} \quad (16)$$

Although it was shown in Ref. 5 that this is formally a bulk property independent of the choice of cell, no explicit expressions were given and no calculations have been attempted using this formalism to our knowledge.

At this point it is clear the close connection between the problem of adiabatic charge transport addressed by Thouless<sup>20</sup> and the issue of the change in polarization between two states of the same crystal connected through an adiabatic transformation. This connection is a fundamental one independent of the quantum mechanical formulation, that is, whether it is a single-body or a many-body theory. To be more precise, Thouless focused on cyclic adiabatic changes of the insulating system, that is, changes such that the Hamiltonian returns to itself in a period of time  $T$ . He found that the bulk contribution to the integrated current  $\mathbf{Q}_{\text{bulk}}$  (equal to  $\Delta \mathcal{P}_{\text{bulk}}$ ) across a boundary is quantized after such a cyclic quantum adiabatic evolution. Niu and Thouless<sup>16</sup> have generalized this result to the case where substrate disorder and particle interactions are present. Although Niu and Thouless considered only 1D cases, their quantum is readily generalized to be the same as that defined by King-Smith and Vanderbilt,<sup>11</sup> and Resta.<sup>12</sup> In the present case, unlike Niu and Thouless, we are interested not only in the quantum but also in noncyclic adiabatic changes of the Hamiltonian operator which induce a polarization difference between two states that do not belong to the same ray.<sup>21</sup> However, because of Eq. (15),  $\Delta \mathcal{P}_{\text{bulk}}$  is exactly equal to the integrated current whether or not the evolution is adiabatic and/or cyclic. Hence we can readily extend<sup>22</sup> Niu and Thouless's many-body derivation for  $\mathbf{Q}_{\text{bulk}}$  to the case where the quantum adiabatic evolution is noncyclic. This scheme provides the many-body generalization of the geometric phase approach<sup>11,12</sup> to macroscopic polarization.

Let us start by writing the  $N$ -body Hamiltonian which describes our electron system

$$\hat{\mathbf{H}}(\lambda) = \sum_{i=1}^N \frac{p_i^2}{2m} + \sum_{1 \leq i < j \leq N} v(|x_i - x_j|) + \hat{V}_{\text{ext}}(\{x_i\}, \lambda), \quad (17)$$

where  $\hat{V}_{\text{ext}}(\{x_i\}, \lambda)$  is the "external" potential which changes adiabatically as a function of  $\lambda$  and  $v(r)$  denotes the repulsive interacting potential. We consider only non-

relativistic Hamiltonians which do not include spin-orbit interactions.<sup>23</sup> The approach of Niu and Thouless is to define a modified Hamiltonian  $\widehat{H}$ , which corresponds to enforcing modified boundary conditions in the original real Hamiltonian. We can study the ground state of  $\widehat{H}$ , in which  $k$  is an additional parameter.  $\Phi_0$  is the ground state solution of the snapshot Schrödinger equation with the many-electron Hamiltonian

$$\widehat{H}(k, \lambda) = \sum_{i=1}^N \frac{(p_i + \hbar k)^2}{2m} + \sum_{1 \leq i < j \leq N} v(|x_i - x_j|) + \widehat{V}_{\text{ext}}(\{x_i\}, \lambda), \quad (18)$$

which satisfies strict periodic boundary conditions over the *supercell* of linear dimension  $L$ , i.e.,  $\Phi_0(x_1, \dots, x_i + L, \dots, x_N) = \Phi_0(x_1, \dots, x_i, \dots, x_N)$  (Ref. 24) and depends parametrically upon  $k$  and  $\lambda$  [from now on we will denote this dependence by  $\Phi_0(k, \lambda)$ ].

For a 1D  $N$ -fermion system enclosed in a box of dimension  $L$  the electronic contribution to  $\Delta \mathbf{P}_{\text{bulk}}$  turns out to be given (in the thermodynamic limit) by

$$\frac{2\pi}{e} \Delta \mathbf{P}_{\text{el}} = \lim_{\substack{N, L \rightarrow \infty \\ N/L \rightarrow \text{const}}} \gamma(C), \quad (19)$$

with a geometric phase ( $k = \pi \kappa / L$ )

$$\gamma(C) = i \int_{-1}^1 d\kappa \int_0^1 d\lambda \left\{ \langle \partial_\lambda \Phi_0 | \partial_\kappa \Phi_0 \rangle - \langle \partial_\kappa \Phi_0 | \partial_\lambda \Phi_0 \rangle \right\}, \quad (20)$$

which after applying Stokes's theorem<sup>25</sup> reads [ $\xi = (\kappa, \lambda)$ ]

$$\gamma(C) = \oint_C \mathcal{A}(\xi) \cdot d\xi, \mathcal{A}(\xi) = i \langle \Phi_0 | \nabla_\xi \Phi_0 \rangle. \quad (21)$$

This is the many-body generalization of Eqs. (4) and (5).

In the expression above for the connection  $\mathcal{A}$  we have assumed that the ground state  $\Phi_0$  is nondegenerate ( $\langle \Phi_0 | \Phi_0 \rangle = 1$ ). The line integral is performed along the closed path  $C$  defined as the contour of the rectangle  $[-1, 1] \times [0, 1]$  in the plane  $\kappa, \lambda$ . Notice that if one can choose  $\Phi_0(k, \lambda)$  to be always real, then  $\gamma(C) = 0$  (assuming  $C$  does not encircle a degeneracy); in other words, the Hermitian Hamiltonian  $\widehat{H}$  must be complex in order to have a nontrivial  $\gamma(C)$ . Since the external potential  $\widehat{V}_{\text{ext}}$  is real, this complex character is induced by the boundary conditions through the  $k$ -dependent kinetic energy operator in Eq. 18.

Berry's phase  $\gamma(C)$  depends on the path and not on the rate at which it is transversed. Moreover,  $\gamma(C)$  is independent of the choice of phases for  $\Phi_0$  (i.e., gauge independent): Suppose that one makes a different choice, say  $\Phi_0 \rightarrow \widetilde{\Phi}_0 = \exp[i \Theta(k, \lambda)] \Phi_0$ . Then, from Eq. (20) and noticing that the normalization condition  $\langle \Phi_0 | \Phi_0 \rangle = 1$  implies that  $\langle \Phi_0 | \nabla_\xi \Phi_0 \rangle = -\langle \nabla_\xi \Phi_0 | \Phi_0 \rangle$ , it is straightforward to prove that the geometric phase is *invariant*, that

is,  $\gamma(C) = \widetilde{\gamma}(C)$ . In that sense  $\Delta \mathbf{P}_{\text{el}}$  is an observable (modulo “nothing”).

One can extend the analysis and realize that the periodic boundary conditions determine a global topology such that

$$\Phi_0(k, \lambda) = \exp \left[ i \left( \Theta(k, \lambda) + G \sum_{i=1}^N x_i \right) \right] \Phi_0(k + G, \lambda), \quad (22)$$

where  $\Theta(k, \lambda)$ , which fixes the gauge, is an arbitrary function of  $k$  and  $\lambda$  and  $G = \frac{2\pi}{L} n$  is a “supercell” reciprocal vector ( $n$  is any integer). If one insists on working in a *path independent* gauge ( $\Theta$  is a function of  $k$  only),<sup>26</sup> it is simple to show that

$$\gamma(C) = i \int_{-\pi/L}^{\pi/L} dk \left\{ \langle \Phi_0(k, 1) | \partial_k \Phi_0(k, 1) \rangle - \langle \Phi_0(k, 0) | \partial_k \Phi_0(k, 0) \rangle \right\}, \quad (23)$$

which would mean that  $\gamma(C)$  depends only upon the initial and final states; the proof of this relation goes along the same lines as Ref. 11. However, because Berry's phase  $\gamma(C)$  is intrinsically a function of the path, a path-independent expression such as Eq. (23) turns out to be defined modulo “something.” This is precisely the indeterminacy of the quantum which is related to the lack of history in Eq. (23). In order to show what is the minimum uncertainty one has to deal with in using Eq. (23) we consider a cyclic evolution. In this case the final state can differ at most by a phase:  $\Phi_0(k, 1) = \exp[-i\gamma_k] \Phi_0(k, 0)$ . Under such circumstances and recalling that we work in a path independent gauge, simple algebraic manipulations show that  $\exp[i\gamma_k] = \exp[i\gamma_{k+G}]$ , which implies that  $\gamma_k = \phi_k + \sum_{i=1}^N k n_i L$  (with  $\phi_k$  periodic in  $k$  and  $n_i L$  an arbitrary supercell vector) and therefore  $\gamma(C) = 2\pi \sum_{i=1}^N n_i$ . In this case  $\Delta \mathbf{P}_{\text{el}}$  will be determined modulo ( $e$ ). This result is basically the same as that of Refs. 11 and 12; however, we have clarified that it is a gauge independent result in the sense that it is due to the “lack of history” in *any* two-point formula.

So far we have been discussing the 1D crystal. One can easily generalize the many-body formulation of macroscopic polarization to 3D.<sup>22</sup> In this case, our system consists of  $N$  fermions enclosed in a box whose symmetry is consistent with the symmetry of the crystal (that is, of the external potential). For the sake of clarity, let us assume that the crystal is cubic, in which case we can choose as *supercell* a cube of side  $L$  without loss of generality. Then, the change in polarization along a Cartesian direction  $\alpha$  is given by

$$\frac{(2\pi)^3}{e} (\Delta \mathbf{P}_{\text{el}})_\alpha = |\varepsilon_{\alpha\beta\gamma}| \int_{-\pi/L}^{\pi/L} dk_\beta \int_{-\pi/L}^{\pi/L} dk_\gamma \gamma(C_\alpha), \quad (24)$$

where Berry's phase  $\gamma(C_\alpha)$  is formally given by Eq. (20) with  $\kappa$  replaced by  $\kappa_\alpha$ . Again, no summation is assumed over the indices of the Levi-Civita tensor  $\varepsilon_{\alpha\beta\gamma}$ .

At this point, it is appropriate to ask what the size of the quantum is if one uses the path independent formula for  $\gamma$  [Eq. 23]. To answer this question we study again a cyclic evolution. Following the same arguments as in 1D we arrive to the conclusion that  $\gamma_{\mathbf{k}} = \phi_{\mathbf{k}} + \sum_{i=1}^N \mathbf{k} \cdot \mathbf{L}_i$ , where  $\mathbf{L}_i = (n_1, n_2, n_3)L$ . Hence  $(\Delta \mathbf{P}_{el})_{\alpha}$  will be determined modulo  $(\frac{e}{L^2})$ . Qualitatively, one needs only to move one electron charge  $e$  per supercell area in order to go back to the original system Hamiltonian, and this is the quantum uncertainty. It is clear that the path independent formula is unsuitable whenever the physical change in polarization is larger than the quantum. The reason for the failure is the assumptions that led to the path-independent form Eq. (23):  $\gamma$  *does* depend on the path (history), as the system evolves  $\gamma$  changes in a deterministic manner and Eq. (20) is the appropriate expression giving  $(\Delta \mathbf{P}_{el})_{\alpha}$  uniquely.

Since many-body calculations intrinsically involve large cells, this may appear to be a problem in the many-body formulation which was not present in the single-body one. However, this is not the case. First, if the single-body formulation Eq. (6) is applied directly to a supercell the same shortcomings show up (i.e., the uncertainty is  $e/L^2$ ). Of course, in the noninteracting case, if the underlying periodicity of the system (i.e., of the external potential) is smaller than the one of the supercell, one can cure this problem by using single-particle states which display this property. That is, the quantum is larger and equal to  $e/L_0^2$  ( $2e/L_0^2$  if there is spin degeneracy), where  $L_0^2$  is the area of the primitive cell (small unit cell). Similar requirements on the wave function can be used in actual many-body calculations for interacting electrons. In fact, there already exists an explicit way to overcome the problem for a system with a smaller underlying periodicity. The first important thing to remark is that, apart from single-particle translations over the supercell  $[\mathbf{t}_i(\mathbf{L}) \Phi_0(\mathbf{r}_1, \dots, \mathbf{r}_i, \dots, \mathbf{r}_N) = \Phi_0(\mathbf{r}_1, \dots, \mathbf{r}_i + \mathbf{L}, \dots, \mathbf{r}_N) = \Phi_0(\mathbf{r}_1, \dots, \mathbf{r}_i, \dots, \mathbf{r}_N)]$ , there is the center of mass symmetry which leaves the Hamiltonian of the system invariant

$$\mathbf{T}(\mathbf{L}_0) \Phi_0(\{\mathbf{r}_i\}) = \Phi_0(\{\mathbf{r}_i + \mathbf{L}_0\}) = \Phi_0(\{\mathbf{r}_i\}), \quad (25)$$

where  $\mathbf{T}(\mathbf{L}_0) = \prod_{i=1}^N \mathbf{t}_i(\mathbf{L}_0)$  is the center of mass translation operator. The relevant matrix elements involved in the computation of  $(\Delta \mathbf{P}_{el})_{\alpha}$  are of the form

$$\langle \Phi_0 | \partial_{k_{\alpha}} \Phi_0 \rangle = \int_{\Omega} d\mathbf{r}_1 \cdots \int_{\Omega} d\mathbf{r}_N \Phi_0^*(\{\mathbf{r}_i\}) \partial_{k_{\alpha}} \Phi_0(\{\mathbf{r}_i\}), \quad (26)$$

where  $\Omega$  is the volume of the supercell. Suppose we make the following coordinate transformation:  $\bar{\mathbf{r}}_1 = \sum_{i=1}^N \mathbf{r}_i$ ;  $\bar{\mathbf{r}}_j = \mathbf{r}_j$  ( $j = 2, \dots, N$ ). Then, the domain of integration in Eq. (26) changes to

$$\int_{\Omega} d\bar{\mathbf{r}}_2 \cdots \int_{\Omega} d\bar{\mathbf{r}}_N \int_{-\frac{L_0}{2} + \bar{\mathbf{r}}_2 + \cdots + \bar{\mathbf{r}}_N}^{\frac{L_0}{2} + \bar{\mathbf{r}}_2 + \cdots + \bar{\mathbf{r}}_N} d\bar{\mathbf{r}}_1 \Phi_0^*(\{\bar{\mathbf{r}}_i\}) \partial_{k_{\alpha}} \times \Phi_0(\{\bar{\mathbf{r}}_i\}), \quad (27)$$

but because of the periodicity of the integrand with re-

spect to the center of mass coordinate the last expression reduces to

$$N_{\text{cell}} \int_{\Omega} d\bar{\mathbf{r}}_2 \cdots \int_{\Omega} d\bar{\mathbf{r}}_N \int_{-\frac{L_0}{2} + \bar{\mathbf{r}}_2 + \cdots + \bar{\mathbf{r}}_N}^{\frac{L_0}{2} + \bar{\mathbf{r}}_2 + \cdots + \bar{\mathbf{r}}_N} d\bar{\mathbf{r}}_1 \Phi_0^*(\{\bar{\mathbf{r}}_i\}) \times \partial_{k_{\alpha}} \Phi_0(\{\bar{\mathbf{r}}_i\}), \quad (28)$$

where  $N_{\text{cell}} = \frac{\Omega}{\Omega_0}$  is the number of elementary cells of volume  $\Omega_0$  contained in the supercell. One can go back to the original coordinate system and write the matrix element as

$$N_{\text{cell}} \int_{\Omega_0} d\mathbf{r}_1 \int_{\Omega_0} d\mathbf{r}_2 \cdots \int_{\Omega_0} d\mathbf{r}_N \Phi_0^*(\{\mathbf{r}_i\}) \partial_{k_{\alpha}} \Phi_0(\{\mathbf{r}_i\}). \quad (29)$$

In this way we exploited the additional symmetry and, if we now consider a cyclic evolution, it is not difficult to realize that the uncertainty turns out to be  $e/L_0^2$ . Therefore, we arrive at the conclusion that the quantum uncertainty is related to the center of mass translational symmetry and, independent of whether the system is interacting or noninteracting, it is given by  $e/L_0^2$ .

In the case of a system where the minimum cell must be very large (e.g. a disordered system) there appears to be a problem. However, this is at least in principle solvable by specifying the path. If one specifies that the final state is arrived at by a series of steps  $i$  in which  $\Delta \mathbf{P}_i$  is smaller than the quantum, then the sum  $\sum_i \Delta \mathbf{P}_i = \Delta \mathbf{P}$  can be arbitrarily large. In other words, the specification of the path leads to a unique answer, not limited by the quantum, which must be possible since the path-dependent phase [Eq. (21)] is well defined as shown previously.

### III. SIMPLE APPLICATION: THE HUBBARD DIMER

In this section we apply previous ideas to compute the electric dipole moment (polarization) of a diatomic molecule with a Hubbard on-site interaction. This *Hubbard dimer* is a very simple model which displays the essential features we wish to emphasize here, i.e., it is a system with many-body interactions and an analytic solution. Its second quantized Hamiltonian is

$$\begin{aligned} \hat{\mathbf{H}} = & \epsilon_a \sum_{\sigma=\uparrow,\downarrow} \hat{c}_{a,\sigma}^{\dagger} \hat{c}_{a,\sigma} + \epsilon_b \sum_{\sigma=\uparrow,\downarrow} \hat{c}_{b,\sigma}^{\dagger} \hat{c}_{b,\sigma} \\ & + t \sum_{\sigma=\uparrow,\downarrow} \left( \hat{c}_{a,\sigma}^{\dagger} \hat{c}_{b,\sigma} + \hat{c}_{b,\sigma}^{\dagger} \hat{c}_{a,\sigma} \right) \\ & + U_a n_{a,\uparrow} n_{a,\downarrow} + U_b n_{b,\uparrow} n_{b,\downarrow}, \end{aligned} \quad (30)$$

where  $\hat{c}_{a,\sigma}^{\dagger}$  is a fermion operator which creates an electron in the atomic state  $\psi_a$  (of energy  $\epsilon_a$ ) with spin  $\sigma$ ,  $t$  is the hopping matrix element between atoms  $a$  and  $b$ , and  $U_{a(b)}$  is the on-site Coulomb repulsion. The number operator  $n_{a,\sigma} = \hat{c}_{a,\sigma}^{\dagger} \hat{c}_{a,\sigma}$ .

On the other hand, the Hamiltonian of a *lattice* of Hubbard dimers is given by

$$\widehat{\mathbb{H}}_\ell = \sum_j \left[ \widehat{\mathbb{H}}(j) + \tilde{t} \sum_{\sigma=\uparrow,\downarrow} [\hat{c}_{a,\sigma}^\dagger(j+1) \hat{c}_{b,\sigma}(j) + \hat{c}_{b,\sigma}^\dagger(j) \hat{c}_{a,\sigma}(j+1)] \right], \quad (31)$$

where  $j$  is the dimer index and  $\tilde{t}$  is the hopping matrix element connecting neighboring dimers. The isolated-dimer limit is recovered by letting  $\tilde{t} \rightarrow 0$ .

In order to implement the geometric approach, we will assume that the system evolves adiabatically from the state where the two constituent atoms are equal (zero dipole) to the situation where they are different, keeping the internuclear separation  $\delta$  fixed. Instead of working with the real Hamiltonian of Eq. (31), we introduce the modified complex Hamiltonian, which is a lattice version of Eq. (18), in the gauge where all the phase change is in the hopping matrix element  $\tilde{t}$

$$\widehat{H}(k) = \sum_j \left[ \widehat{\mathbb{H}}(j) + \tilde{t} \sum_{\sigma=\uparrow,\downarrow} [\hat{c}_{a,\sigma}^\dagger(j+1) e^{-ikL_0} \hat{c}_{b,\sigma}(j) + \hat{c}_{b,\sigma}^\dagger(j) e^{ikL_0} \hat{c}_{a,\sigma}(j+1)] \right]. \quad (32)$$

Its spectrum and ground state  $\Phi_0(k)$  depend explicitly on  $k$ , the momentum label of the *generalized* Bloch functions.<sup>24</sup>  $L_0$  is the linear size of the primitive cell which holds a single dimer.

Following the analysis of the preceding section and applying the “two-point” expression Eq. (23), we arrive at the conclusion that the electronic contribution to the change in polarization is given (up to a quantum) as the limit of Berry’s phase

$$\Delta \mathbf{P}_{\text{el}} = \lim_{N,L \rightarrow \infty} i \frac{e}{2\pi} \int_{-\pi/L}^{\pi/L} dk \langle \Phi_0(k) | \partial_k \Phi_0(k) \rangle, \quad (33)$$

where  $\Phi_0(k) = \exp[-ik \sum_{i=1}^N x_i] \Psi_0(\{x_i\})$  and  $\Psi_0$  is the ground state solution of  $\widehat{\mathbb{H}}_\ell$  ( $\langle \Psi_0 | \Psi_0 \rangle = 1$ ), which satisfies the generalized Bloch condition on the boundary.<sup>24</sup> The full solution of the Hubbard dimer lattice is beyond the scope of this paper. However, we can illustrate the solution of the many-body problem in the isolated dimer case where there is a simple analytic solution.

Now, we want to compute the electric dipole moment of the isolated molecule  $\mathbf{d}$  using both the direct method (dipolar moment of the charge distribution) and the geometric phase approach. In the direct method the physical quantity  $\mathbf{d}$  is

$$\mathbf{d} = e \langle \tilde{\Psi}_0 | \hat{x} | \tilde{\Psi}_0 \rangle, \quad (34)$$

where  $\tilde{\Psi}_0$  is the ground state of the Hamiltonian  $\widehat{\mathbb{H}}$  which satisfies free boundary conditions ( $\langle \tilde{\Psi}_0 | \tilde{\Psi}_0 \rangle = 1$ ) and

$\hat{x} = \sum_{i=1}^N x_i$  (here  $N$  is the number of electrons which compose the dimer). The comparison of both approaches (which, of course, give the same answer) will let us appreciate the essence and origin of Berry’s phase in this class of problems. It is important to stress that, since the system is finite, there will be no quantum uncertainty if a two-point formula is applied (the center of mass is unambiguously defined). So, not only the dipole moment difference but the absolute dipole of the dimer are well defined (modulo nothing). We show that Berry’s phase, which turns out to be the dipole moment in this context, appears as a consequence of using a topologically different configuration space than the one of the original problem [Eq. (30)].

The geometric phase procedure consists in changing the original simply connected space to a multiply connected one (1D torus or ring). This procedure seems artificial when applied to a finite system, but in fact this is nothing else than a restatement of the Bloch condition in the infinite system (lattice). The modified complex Hamiltonian of the dimer is the one written in Eq. (32) but specialized to a single cell (see Fig. 4). The isolated-dimer limit is characterized by letting  $\tilde{t} \rightarrow 0$  (as the ring linear dimension  $L_0 \rightarrow \infty$ ). Then, following the same analysis which led to Eq. (33), the electric dipole moment is given by

$$\mathbf{d} = \lim_{L_0 \rightarrow \infty} i \frac{e L_0}{2\pi} \int_{-\pi/L_0}^{\pi/L_0} dk \langle \Phi_0(k) | \partial_k \Phi_0(k) \rangle. \quad (35)$$

Notice that we have multiplied Eq. (35) by  $L_0$  because the relevant observable is the “total polarization” (electric dipole moment); otherwise  $\mathbf{d}$  would vanish in the limit  $L_0 \rightarrow \infty$  ( $N$  is finite). By explicit construction of the ground states  $\tilde{\Psi}_0$  and  $\Psi_0$  we will show that both expressions, Eqs. (34) and (35), give the same answer.

We study the simplest dimer, i.e., the homonuclear neutral molecule ( $Z_a = Z_b = |e|$  nuclear charge) with  $N=2$  electrons (in the sector of total spin  $S=0$ ) for certain values of the Hamiltonian parameters. The ground state of the two-particle system ( $\tilde{\Psi}_0, \Psi_0$ ) will be given by a symmetric function of the particle coordinates  $x_i$  (the

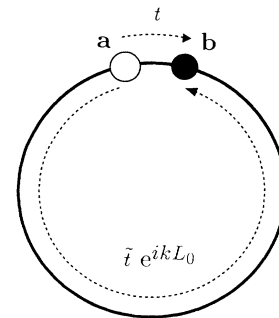


FIG. 4. Schematic plot of a Hubbard dimer on a ring (1D torus) of perimeter  $L_0$ . In the picture we display the two atoms (**a** and **b**) and the hopping matrix elements which enter in the modified complex Hamiltonian  $\widehat{H}(k)$ . Notice that the time-reversal symmetry is broken.

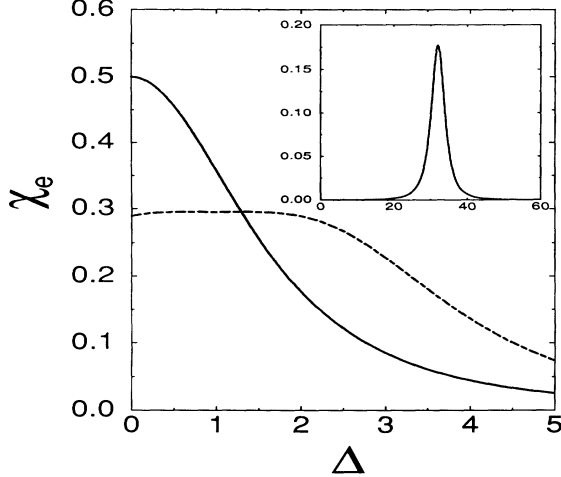


FIG. 5. Charge susceptibility  $\chi_e$  as a function of the atomic energy difference  $\Delta = \epsilon_a - \epsilon_b$  for three different physical regimes. The solid line in the main figure corresponds to the single-body approximation ( $U_a = U_b = 0$ ). The dash-dotted line mimics the intermediate valence situation ( $U_a = 0; U_b/t \sim 1$ ). The curve in the inset corresponds to the strong-coupling limit ( $U_{a(b)}/t \gg 1$ ).

complete antisymmetry is restored by the spin part of the wave function<sup>23</sup>). Under these assumptions, the ground state  $\Phi_0(k)$  will be given by

$$\Phi_0(k) = \alpha_k \psi_a(x_1)\psi_a(x_2) + \beta_k \psi_b(x_1)\psi_b(x_2) + \frac{\gamma_k}{\sqrt{2}} [\psi_a(x_1)\psi_b(x_2) + \psi_a(x_2)\psi_b(x_1)] , \quad (36)$$

where the functions  $\alpha_k, \beta_k, \gamma_k$  depend parametrically upon  $k$  and the Hamiltonian parameters. The atomic states  $\psi_{a(b)}$  are defined on a closed manifold satisfying strict periodic boundary conditions, i.e.,  $\psi_{a(b)}(x + L_0) = \psi_{a(b)}(x)$  (single-valued functions on the ring). After Eq. (35)

$$\mathbf{d} = \lim_{L_0 \rightarrow \infty} \frac{e L_0}{2\pi} \int_{-\pi/L_0}^{\pi/L_0} dk [\langle \Psi_0 | (x_1 + x_2) \Psi_0 \rangle + i \langle \Psi_0 | \partial_k \Psi_0 \rangle] . \quad (37)$$

It is very simple to prove (assuming that the orbital states  $\psi_{a(b)}$  are orthogonal) that the first term of Eq. (37) in the limit  $L_0 \rightarrow \infty$  is indeed equal to  $e \langle \tilde{\Psi}_0 | \hat{x} \tilde{\Psi}_0 \rangle$ , while the second vanishes provided that  $\tilde{t}$  goes to zero faster than  $1/L_0$ .

In Fig. 5 we plot the *charge susceptibility*  $\chi_e$ , a result which has been obtained using the previous calculation for the dipolar moment  $\mathbf{d}$ . Specifically,  $\chi_e$  is defined as

$$\chi_e = \frac{1}{e\delta} \frac{\partial \mathbf{d}}{\partial \Delta} , \quad (38)$$

where  $\Delta = \epsilon_a - \epsilon_b$ . In particular, we consider three physical situations: zero coupling ( $U_{a(b)} = 0$ ), strong cou-

pling ( $U_{a(b)}/t \gg 1$ ), and intermediate valence limits. The physical picture that emerges from this toy model is clear: Whenever the on-site two-body interaction is turned off (single-body approximation),  $\chi_e$  is a monotonically decreasing function of  $\Delta$ . As soon as the repulsive Hubbard interaction is turned on, more interesting situations arise. In the intermediate valence regime ( $U_a = 0; U_b/t \sim 1$ )  $\chi_e$  is large whenever  $\Delta \sim U_b$ , while in the strong-coupling limit ( $U_{a(b)}/t \gg 1$ ) the system hardly responds (Heisenberg limit) up to the moment  $\Delta \sim U_b$  where a resonance starts to develop. In the case of the Hubbard lattice [Eq. (31)] we would expect a similar physical behavior in  $\chi_e$  for small  $\tilde{t}$ .

#### IV. CONCLUSIONS

In this paper we have developed full many-body expressions for the change in bulk polarization  $\Delta \mathbf{P}_{\text{bulk}}$  between two quantum insulating states in terms of a Berry phase. Our derivation is based upon two main concepts: The first regards the observation that for a quantum insulator charge cannot flow between the bulk and surface of the system, where the division between bulk and surface is discussed in the text (illustrated in Fig. 3) and follows the work of Kallin and Halperin. The second is the method of averaging over boundary conditions (i.e., over momentum vectors  $\mathbf{k}$ ) following the approach of Niu and Thouless. The geometric phase formulas depend on the path which connects the two states and it turns out that  $\Delta \mathbf{P}_{\text{bulk}}$  defined in this way is a well-defined bulk property. If one attempts to consider *only* the dependence of  $\Delta \mathbf{P}_{\text{bulk}}$  upon the initial and final configurations (two-point formula), then a quantum of uncertainty arises. We showed that this uncertainty is related to the center of mass translational symmetry and the quantum is the one that corresponds to the primitive cell. This geometric viewpoint provides a mathematical method to compute macroscopic polarizations in a many-body framework.

We have illustrated such concepts using a simple model with an analytic solution: a neutral Hubbard dimer. Since this is a finite system, both the electric dipole difference and the absolute dipole are well-defined quantities (modulo nothing). However, this simple toy model let us emphasize the origin of Berry's phase in the context of polarization: Since the original Hamiltonian of the system is a real symmetric operator, the nontrivial geometric quantum phase is due to the  $D$ -dimensional torus topology of the manifold induced by the periodic boundary conditions.

#### ACKNOWLEDGMENTS

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- <sup>1</sup> J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975).
- <sup>2</sup> L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, London, 1960).
- <sup>3</sup> M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford University Press, Oxford, 1954).
- <sup>4</sup> *The Dielectric Function of Condensed Systems*, edited by L. V. Keldysh, D. A. Kirzhnits, and A. A. Maradudin (North-Holland, Amsterdam, 1989).
- <sup>5</sup> R. M. Martin, Phys. Rev. B **9**, 1998 (1974).
- <sup>6</sup> A. K. Tagantsev, Phys. Rev. Lett. **69**, 389 (1992); A. Baldereschi, M. Posternak, and R. Resta, *ibid.* **69**, 390 (1992).
- <sup>7</sup> R. M. Martin, Phys. Rev. B **6**, 4874 (1972).
- <sup>8</sup> R. Kubo, M. Toda, and N. Hashitsume, *Statistical Physics II* (Springer-Verlag, Berlin, 1985).
- <sup>9</sup> P. C. Martin, *Measurement and Correlation Functions* (Gordon and Breach, New York, 1968).
- <sup>10</sup> R. Pick, M. H. Cohen, and R. M. Martin, Phys. Rev. B **1**, 910 (1970).
- <sup>11</sup> R. D. King-Smith and D. Vanderbilt, Phys. Rev. B **47**, 1651 (1993).
- <sup>12</sup> R. Resta, *Ferroelectrics* **136**, 51 (1992); *Europhys. Lett.* **22**, 133 (1993).
- <sup>13</sup> For an excellent introduction to such concepts see Ch. Nash and S. Sen, *Topology and Geometry for Physicists* (Academic, London, 1983).
- <sup>14</sup> *Geometric Phases in Physics*, edited by A. Shapere and F. Wilczek (World Scientific, Singapore, 1989).
- <sup>15</sup> Basically, a connection is a quantity which let us compare two tangent spaces at a pair of points infinitesimally separated in a curved manifold. See, for instance, Ref. 13, Chap. 7.
- <sup>16</sup> Q. Niu and D. J. Thouless, J. Phys. A **17**, 2453 (1984).
- <sup>17</sup> C. Kallin and B. I. Halperin, Phys. Rev. B **29**, 2175 (1984).
- <sup>18</sup> D. Vanderbilt and R. D. King-Smith, Phys. Rev. B **48**, 4442 (1993).
- <sup>19</sup> W. Kohn, Phys. Rev. **133**, A171 (1964).
- <sup>20</sup> D. J. Thouless, Phys. Rev. B **27**, 6083 (1983).
- <sup>21</sup> Elements of the Hilbert space  $\mathcal{L}^2$  of a quantum mechanical system (i.e, wave functions) which differ *only* by a phase are equivalent and said to belong to the same ray.
- <sup>22</sup> For the detailed many-body formulation and application see G. Ortiz, P. Ordejon, and R. Martin (unpublished).
- <sup>23</sup> Since only spin-free Hamiltonians are considered here, the many-body wave function  $\Xi$  can be written as a product of a coordinate and a spin function:  $\Xi(\{x_i\}, \Sigma) = \Phi(\{x_i\}) \otimes \chi(\Sigma)$ , where  $\Sigma = (\sigma_1, \dots, \sigma_i, \dots, \sigma_N)$  are discrete spin variables. The configuration part  $\Phi$  must have the right symmetry in order to account for the Pauli principle.
- <sup>24</sup> The many-body wave functions  $\Psi_m$  of the original real Hamiltonian  $\hat{H}$  are related to the states of the modified Hamiltonian  $\hat{H}_m$  by a simple gauge transformation:  $\Psi_m = \exp[ik \sum_{i=1}^N x_i] \Phi_m$  and hence satisfy  $\Psi_m(x_1, \dots, x_i + L, \dots, x_N) = \exp[ikL] \Psi_m(x_1, \dots, x_i, \dots, x_N)$  (*the generalized Bloch condition*).
- <sup>25</sup> Y. Choquet-Bruhat, C. DeWitt-Morette, and M. Dillard-Bleick, *Analysis, Manifolds and Physics* (North-Holland, Amsterdam, 1982), p. 216.
- <sup>26</sup> This is not allowed if the external potential itself  $\hat{V}_{\text{ext}}$  induces a holonomy, i.e., a nontrivial geometric phase.  $\Theta(k) = 0$  corresponds to the *periodic* gauge used in Refs. 11 and 12. We find it more suitable to call the general  $\Theta(k)$  gauge the *path independent* ( $\lambda$  independent) gauge.



FIG. 1. Schematic of bulk plus surface regions for a general sample shape. The macroscopic electric field  $\mathbf{E}_{mac}$  is kept zero in the interior of the sample by *external* means.