

A fresh look at the position operator (as in polarization, magnetization, and more)

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Macroscopic polarization, orbital magnetization, and related quantities have a trivial expression for a molecule or for a finite crystallite, where the wavefunction is square-integrable: so-called “open” boundary conditions (OBCs). In condensed matter physics it is instead necessary to adopt periodic boundary conditions (PBCs): what makes life difficult is the unbounded position operator \mathbf{r} , which is *not* a legitimate operator within PBCs.

Solutions to this problem are known since the early 1960s, when the first expressions for computing the dielectric polarizability of a solid were proposed. Other solutions, to be applied to different problems, appeared over the years. Invariably, bulk properties of a crystalline system are cast as Brillouin zone (BZ) integrals, where the integrand is expressed in terms of Bloch orbitals. We have therefore two expressions—one a \mathbf{k} -space integral within PBCs, the other an \mathbf{r} -space integral within OBCs—for the same physical property. What is disturbing is that the two expressions do not bear any apparent relationship.

Here I am going to show that the OBCs and PBCs expressions for a given property are essentially the same when expressed in terms of the *projected* position operator $\tilde{\mathbf{r}}$, defined below. While some of the results presented here are known, some are not: in particular, the expression for orbital magnetization given below is new.

For noninteracting electrons (either Hartree-Fock or Kohn-Sham) the ground state is uniquely determined by the projector P over the occupied electronic states; in the large system limit, P is the same within either OBCs or PBCs. We define as Q the complementary projector over the unoccupied states, and we define the *Hermitian* operator $\tilde{\mathbf{r}} = P\mathbf{r}Q + Q\mathbf{r}P$, i.e. \mathbf{r} minus its occupied-occupied and unoccupied-unoccupied components. In a crystalline system $\tilde{\mathbf{r}}$ is a lattice-periodical operator, and is expressed as a BZ integral. The physical properties of interest will be expressed as traces per unit volume involving $\tilde{\mathbf{r}}$. The novel compact formulation applies to both OBCs and PBCs; it even applies to noncrystalline systems, where $\tilde{\mathbf{r}}$ is no longer lattice-periodical, but is nonetheless a regular operator.

In the following the symbol “Tr” means trace per unit volume; it becomes the trace per cell in the crystalline case. The expressions are given for “spinless electrons”; trivial factors of 2 are not included.

Polarization. When an insulating system is adiabatically transformed between an initial and a final state the (electronic) polarization difference is

$$\Delta \mathbf{P}_{\text{electronic}} = -e \int_0^1 d\lambda \text{Tr} \left\{ \frac{\partial P}{\partial \lambda} \tilde{\mathbf{r}} \right\}.$$

Quadratic spread. The gauge-invariant quadratic spread, as it enters the Marzari-Vanderbilt theory of maximally localised Wannier functions, is

$$\Omega_{\text{I}} = V_{\text{cell}} \text{Tr} \{ P \tilde{\mathbf{r}} \cdot \tilde{\mathbf{r}} \},$$

where V_{cell} is the cell volume. When used for a molecule within OBCs, the analogous formula yields the gauge-invariant spread of Boys’ orbitals.

Orbital magnetization. The new formula for insulating materials is

$$M_{\gamma} = -\frac{ie}{2\hbar c} \epsilon_{\gamma\alpha\beta} \text{Tr} \{ |H - \mu| \tilde{r}_{\alpha} \tilde{r}_{\beta} \},$$

where μ is the Fermi level. We stress that this applies to both a finite crystallite within OBCs, and to a crystalline system within PBCs. When the operator $\tilde{\mathbf{r}}$ is expressed as a BZ integral, we recover the known \mathbf{k} -space formula of the modern theory of magnetization. The present formula, as given, holds for Chern insulators as well.

Chern number. We switch here to a two-dimensional system, and we redefine “Tr” as the trace per unit area. The Chern number is

$$C = -2\pi i \text{Tr} \{ P[\tilde{x}, \tilde{y}] \}.$$

If the formula is adopted for a finite flake within OBCs the total trace vanishes; nonetheless the trace per unit area evaluated in an inner region of the flake provides the Chern number.

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