Drude weight in systems with open boundary conditions

Gabriele Bellomia\textsuperscript{1} and Raffaele Resta\textsuperscript{2,3,∗}

\textsuperscript{1}International School for Advanced Studies (SISSA), Via Bonomea 265, 34136 Trieste, Italy
\textsuperscript{2}Istituto Officina dei Materiali IOM-CNR, Strada Costiera 11, 34151 Trieste, Italy
\textsuperscript{3}Donostia International Physics Center, 20018 San Sebastián, Spain

A many-electron conducting system undergoes free acceleration in response to a macroscopic field. The Drude weight $D$—also called charge stiffness—measures the adiabatic (inverse) inertia of the electrons; the $D$ formal expression requires periodic boundary conditions. When instead a bounded sample is addressed within open boundary conditions, no current flows and a constant (external) field only polarizes the sample: Faraday cage effect. Nonetheless a low-frequency field induces forced oscillations: we show here that the low-frequency linear response of the bounded system is dominated by the adiabatic inertia and allows an alternative evaluation of $D$. Simulations on model one-dimensional systems demonstrate our main message.

I. INTRODUCTION

Irrelevance of the boundary conditions in the thermodynamic limit is a basic tenet of statistical mechanics and condensed matter physics. Among the possible choices of boundary conditions two are prominent: Born-von-Kármán periodic boundary conditions (PBCs) and the so-called “open” boundary conditions (OBCs). Insofar as an intensive physical observable is computed from finite realizations of a given system, the two choices yield somewhat different results. Yet one postulates that the large-system limit yields the same value for any intensive physical observable.

To be more specific, we will consider below the ground state of a (macroscopically homogeneous) system of $N$ electrons and a neutralizing background of nuclei in a cubic box of volume $L^d$ ($d$ is the dimension). The choice PBCs vs. OBCs amounts to choosing two different Hilbert spaces for describing our system: within PBCs the many-body wavefunction is periodic with periodic boundary conditions (PBCs) and the so-called “open” boundary conditions (OBCs). Insofar as an intensive physical observable is computed from finite realizations of a given system, the two choices yield somewhat different results. Yet one postulates that the large-system limit yields the same value for any intensive physical observable.

Some intensive physical observables are non-problematic: this is e.g. the case of spectral properties. At finite size the spectra are discrete within both OBCs and PBCs, and different between themselves. In the large-system limit the two spectra become continuous and coincide, yielding the same density of states. Indeed, it is a standard exercise to verify this in the special case of a free-electron gas, which can be worked out analytically. Some other properties are more problematic, and were understood relatively recently: in this class are electrical conductivity, which can be worked out analytically. Some other properties are more problematic, and were understood relatively recently: in this class are electrical conductivity, which can be worked out analytically.

II. PHENOMENOLOGY

The conductivity tensor $\sigma_{\alpha\beta}(\omega)$ yields the current density linearly induced by a macroscopic electric field at frequency $\omega$ (Greek subscripts are Cartesian indices); for the sake of simplicity we assume time-reversal symmetry, in which case the transverse conductivity vanishes and $\sigma_{\alpha\beta}(\omega)$ is a symmetric tensor.

In a metal, in absence of dissipation, the electrons in a dc field undergo free acceleration and $\sigma_{\alpha\beta}(\omega)$ is divergent for $\omega = 0$. The most general form for longitudinal
conductivity is:

\[ \sigma_{\alpha \beta}(\omega) = D_{\alpha \beta} \left[ \delta(\omega) + \frac{i}{\pi \omega} \right] + \sigma_{\alpha \beta}^{(\text{regular})}(\omega), \] (1)

where the constant \( D_{\alpha \beta} \) goes under the name of Drude weight (or charge stiffness) and accounts for the inertia of the many-electron system in the adiabatic limit.\(^{4,5} \) The Drude weight can also be defined as:\(^2 \)

\[ D_{\alpha \beta} = \pi \lim_{\omega \to 0} \omega \Im \sigma_{\alpha \beta}(\omega). \] (2)

Longitudinal conductivity obeys the f-sum rule

\[ \int_0^\infty d\omega \ \Re \sigma_{\alpha \alpha}(\omega) = \frac{D_{\alpha \alpha}}{2} + \int_0^\infty d\omega \ \Re \sigma_{\alpha \beta}^{(\text{regular})}(\omega) \]

\[ = \frac{\omega_p^2}{8} \delta_{\alpha \beta} = \frac{\pi e^2 n}{2m} \delta_{\alpha \beta}, \] (3)

where \( n = N/L^d \) is the electron density and \( \omega_p \) is the plasma frequency. For free electrons \( \sigma_{\alpha \beta}^{(\text{regular})}(\omega) \) vanishes and \( D_{\alpha \beta} \) assumes the same value as in classical physics,\(^{10,11} \) i.e. \( D_{\alpha \beta} = D_{\text{free}} \delta_{\alpha \beta} \), with \( D_{\text{free}} = \pi e^2 n/m \). Given Eq. (3), switching the periodic potential on has the effect of transferring some spectral weight from the Drude peak into the regular term; for band insulators the Drude peak vanishes and \( \Re \sigma_{\alpha \beta}^{(\text{regular})}(\omega) \) is zero for \( \omega < \epsilon_{\text{gap}}/\hbar \). In the special case of a band metal considered here \( \sigma_{\omega \beta}^{(\text{regular})}(\omega) \) is a linear-response property which accounts for interband transitions, and is nonvanishing only at frequencies higher than a finite threshold; in more general cases of a noncrystalline and/or correlated many-electron system this selection rule breaks down and \( \sigma_{\alpha \beta}^{(\text{regular})}(0) \) may be nonzero.\(^5 \)

### III. DRUDE WEIGHT

When applied to a band metal within PBCs, Kohn’s general expression\(^2-4 \) becomes the Fermi-volume integral\(^5 \)

\[ D_{\alpha \beta} = 2\pi e^2 \sum_j \int_{BZ(2\pi)^d} \frac{dk}{2\pi^d} \theta(\mu - \epsilon_{j k}) m_{j \alpha \beta}^{-1}(k), \] (4)

where \( BZ \) is the Brillouin zone, \( \mu \) is the Fermi level, and the effective inverse mass tensor of band \( j \) is

\[ m_{j \alpha \beta}^{-1}(k) = \frac{1}{\hbar^2} \partial_{\alpha \beta}^2 f(\epsilon_{j k}) \] (5)

For insulators, the integral in Eq. (4) trivially vanishes; for metals, the contribution of the core bands to \( D_{\alpha \beta} \) vanishes as well. \( D_{\alpha \beta} \) can be equivalently expressed as a Fermi-surface integral, by means of an integration by parts: it acquires then the meaning of an “intraband” term:\(^5 \)

\[ D_{\alpha \beta} = -2\pi e^2 \sum_j \int_{BZ(2\pi)^d} \frac{dk}{2\pi^d} f'(\epsilon_{j k}) v_{j \alpha}(k) v_{j \beta}(k), \] (6)

where \( v_{j \alpha}(k) = \partial_k \epsilon_{j k}/\hbar \) and at zero temperature the Fermi occupation function is \( f(\epsilon) = \theta(\mu - \epsilon) \). Eq. (6) is in explicit agreement with the spirit of Landau’s Fermi-liquid theory, which holds that charge transport in metals involves only quasiparticles with energies within \( k_B T \) of the Fermi level; Eq. (6) is in fact at the root of the semiclassical theory of transport.\(^{11} \)

Notice that so far we have not explicitly invoked the Kubo formula for conductivity; this is a virtue of Kohn’s approach, where they remain implicit. The above results can be equivalently formulated via Kubo formula; it is essential then to adopt the vector-potential gauge and to set \( \mathbf{q} = 0 \) (not \( \mathbf{q} \to 0 \)).\(^4,9 \)

When a bounded crystallite (cut from a bulk metal) is addressed within OBCs there is no Drude peak at finite size. It also follows that the \( \omega > 0 \) region of the spectrum saturates the f-sum rule.\(^{12} \)

### IV. KUBO FORMULA

The Kubo formulae can be cast in several equivalent ways; here it is expedient to adopt the form\(^5 \)

\[ \sigma_{\alpha \beta}(\omega) = \frac{2ie^2\hbar}{L^d} \sum_{m,n} \left( f_n - f_m \right) \frac{\langle n | v_\alpha | m \rangle \langle m | v_\beta | n \rangle}{\epsilon_m - \epsilon_n} \] (7)

where the velocity is \( \mathbf{v} = i[H, \mathbf{r}]/\hbar \), the positive infinitesimal \( \eta \) enforces causality, and \( f_n = 1/(e^{\epsilon_n/k_B T} + 1) \) is the Fermi occupation factor.

The expression of Eq. (7) is quite general, and applies both within OBCs and PBCs; in the latter case \( n \) must be identified with the band and Bloch index \( j \mathbf{k} \), and the volume \( L^d \) with the cell volume. We perform the \( T \to 0 \) limit first, thus getting for an isotropic system and at \( \omega > 0 \):

\[ \Re \sigma(\omega) = \frac{2\pi e^2}{\hbar L^d} \sum_{\epsilon_{n \leq \mu} \epsilon_{m > \mu}} \frac{|\langle n | v_x | m \rangle|^2}{\omega_{nm}} \delta(\omega - \omega_{nm}), \] (8)

where \( \omega_{nm} = (\epsilon_m - \epsilon_n)/\hbar \). Eq. (8) obeys the f-sum rule in the OBCs case, but instead does not saturate it in the PBCs metallic case.\(^{12} \) in fact Eq. (8) in the thermodynamic limit yields the regular term \( \sigma_{\alpha \beta}^{(\text{regular})}(\omega) \) only. We stress that the matrix elements and the selection rules are quite different in the two cases. In the special case of free electrons the PBCs orbitals are plane waves and all matrix elements vanish because of an obvious selection rule.

We observe that, by adopting PBCs, we may even perform the limits in Eq. (7) in the reverse order (thanks to the Bloch theorem): first the thermodynamic limit—from discrete to continuous \( \mathbf{k} \)—and afterwards \( T \to 0 \). In this case Eq. (7) is endowed with an intraband piece, where the diagonal elements \( \langle n | v | m \rangle \) are identified with \( v_j(k) = \partial_k \epsilon_{j k}/\hbar \), and the factor \( (f_n - f_m)/(\epsilon_m - \epsilon_n) \) with \(-f'(\epsilon_{j k})\); one thus gets the Drude term exactly in
the same form as in Eq. (6), besides the \( \omega > 0 \) interband term.\(^5\)

At any finite size all poles in Eq. (8) occur at positive energies, within both PBCs and OBCs. There is an outstanding difference, though: the PBCs poles are gapped, while the OBCs poles converge to zero frequency. The selection rules forbid intraband PBCs contributions to Eq. (8), while instead within OBCs the intraband transitions originate low-frequency poles, which contribute with extra spectral weight to the \( f \)-sum rule. We expect these low-energy poles to coalesce, for \( L \to \infty \), into a single pole at \( \omega = 0 \), whose residue yields \( D \). In the following of this paper we are going to study this process in detail for a few one-dimensional test cases.

V. SOUZA-WILKENS-MARTIN SUM RULE

Souza, Wilkens, and Martin (hereafter quoted as SWM)\(^8\) proposed in 2000 to characterize the metallic/insulating behavior of a material by means of the integral (for isotropic systems)

\[
I^{(\text{SWM})} = \int_0^\infty \frac{d\omega}{\omega} \text{Re} \sigma(\omega),
\]

(9)

which diverges for all metals and converges for all insulators. We adopt here the SWM approach, but we stress that—at finite size—its PBCs features are quite different from the OBCs ones.

In a band metal \( I^{(\text{SWM})} \) diverges within PBCs because of the \( \delta \)-like Drude peak, which exists even at finite size; equivalently, it diverges because a dc field induces free acceleration (again, even at finite size). Within OBCs, instead, all of the poles of \( \sigma(\omega) \) occur at nonzero frequency; \( I^{(\text{SWM})} \) is finite at any size, and diverges in the large-system limit. Our simulations will show that such divergence is due to the low-frequency poles which are the fingerprint of \( D \) within OBCs: the system cannot undergo free acceleration, but when the size is increased the forced oscillation decrease in energy and couple to the field with nonvanishing oscillator strength.

In a band insulator \( I^{(\text{SWM})} \) is finite both within PBCs and OBCs; while we expect the integrated values to converge towards the same large-system limit. From Eq. (8) we get the sum

\[
I^{(\text{SWM})} = \frac{2\pi e^2}{\hbar L^2} \sum_{\epsilon_n \leq \mu, \epsilon_m > \mu} \frac{|(n \mid v \mid m)|^2}{\omega_{nm}^2};
\]

(10)

when evaluated within PBCs vs. OBCs its terms differ in energies, matrix elements, and selection rules. The lowest PBCs transition energy is gapped, while no selection rule forbids low-energy transitions within OBCs: this fact is at the root of the divergence of the OBCs SWM integral, Eq. (10), in the metallic case.

By exploiting completeness, \( I^{(\text{SWM})} \) can be transformed into a ground-state property, having the meaning of second cumulant moment of the electron distribution.\(^8,13,14\) We are not going to exploit such transformation here, implementing instead Eq. (10), and focusing on the behavior of its contributions.

VI. SIMULATIONS

From now on we address \( D \) in units of \( \pi e^2 n/(2m) = \omega_p^2/8 \), such that the \( f \)-sum rule yields 1. Therefore \( D_{\text{free}}/2 = 1 \) for free electrons, and \( D/2 < 1 \) for a generic metal; the conductivity will be displayed in units of \( \omega_p/2 \) throughout.

A. Free electrons

As said above, the free-electron case in PBCs is trivial: \( \sigma^{(\text{regular})}(\omega) \equiv 0 \) and all of the spectral weight goes into the \( \delta(\omega) \) term. In the OBCs case we have computed Eq. (8) using the (analytical) eigenvalues and eigenfunctions of a 1d infinite potential well of length \( L \), at a linear density \( n = N/L = 0.2 \) bohr\(^{-1} \), with double orbital occupancy. The result is shown in Fig. 1 for \( N = 162 \), for a cutoff of 1.4 Ha; the \( \delta \)-singularities have been plotted, as customarily, as narrow Gaussians. The figure perspicuously show that the poles of Eq. (8) accumulate at very low energy; the value of \( \sigma(0) \) does not carry any physical meaning, since it depends on the (arbitrary) Gaussian smearing. We emphasize instead the accurate integrated (smearing-independent) value of \( \sigma(\omega) \): the \( f \)-sum rule is satisfied here at 99.99%.

From Fig. 1 it is seen that the poles carrying non-negligible residues occur at frequencies lower than about 0.15 Ha; this value is clearly size-dependent, since the spacing of the levels goes like \( 1/L \). One would expect that all poles converge towards zero in the large-\( L \) limit; but the situation, illustrated in Fig. 2, is much less triv-
FIG. 2. $L$-dependence of the poles in Eq. (8), at a low cutoff $\epsilon_{\text{cut}} = 0.01$ Ha. The top panel shows the pole frequencies: the number of poles increases with $L$, there are families of poles, each family following a $1/L$ law. The bottom panel shows the corresponding residues in units of $\pi e^2 n/(2m) = \omega_p^2/8$, exponentially vanishing with the family index. At any given cutoff, there are several families of poles, whose number increases with size. Within a given family the frequency follow a $1/L$ law, as shown in Fig. 2, top panel; the cutoff has been lowered for the sake of clarity. We also find (Fig. 2, bottom) that the pole residues are essentially $L$-independent and that they are exponentially vanishing with the family index; therefore only a small number of low-frequency poles carry significant residues. The message of both panels altogether is therefore that, despite a complex pole pattern, the spectral weight is confined in a frequency region proportional to $1/L$.

### B. Periodic potential

Next we switch on a potential in the form of a periodic array of Gaussians:

$$U(x) = \sum_{m=-\infty}^{\infty} V(x - ma), \quad V(x) = V_0 e^{-x^2/b^2};$$

we set $a = 5$ and $b = 1$ bohr. We get a model metal with 1 electron/cell and a model insulator with 2 electrons/cell.

FIG. 3. Conductivity of the model insulator in units of $\omega_p/2$, after Eq. (8). The gap is $\epsilon_{\text{gap}} = 0.35$ Ha; the cutoff is shown as a vertical dashed line. Left panel: OBCs; Right panel: PBCs.

FIG. 4. Drude weight as a function of the periodic potential strength $V_0$.

in the former case the density is the same as for the free-electron case, discussed above. By choosing $V_0 = 0.8$ Ha the first gap in the spectrum is $\epsilon_{\text{gap}} = 0.35$ Ha.

### C. Model insulator

We start with showing the results of the (almost trivial) insulating case. With $V_0 = 0.8$ Ha and a cutoff of 2.2 Ha we are very close to completeness: $f$-sum = 99.93% and 99.99% within OBCs and PBCs, respectively. The conductivity plots evaluated from Eq. (8) in the two cases are basically undistinguishable (Fig. 3); the SWM integrals, Eq. (10), differ by 0.3%.

### D. Model metal

We start showing in Fig. 4 the value of $D$ as a function of the periodic potential strength $V_0$, computed within PBCs, i.e. with the the 1d version of Eq. (6). Starting from the free-electron $V_0 = 0$ case, $D$ decreases and
converges to zero in the flat-band limit. All of the following simulations are performed with a cutoff of 2.2 Ha: f-sum = 99.97% and 99.70% within OBCs and PBCs, respectively. The conductivity plots evaluated from Eq. (8) in the two cases are shown in Fig. 5. As explained above, Eq. (8) within PBCs yields the regular (interband) term $\sigma^{\text{regular}}(\omega)$ only; the Drude (intra-band) term must be evaluated separately from Eq. (6).

Within OBCs $\sigma(\omega)$, evaluated from Eq. (8), saturates the $f$-sum. It shows two well separated contributions, which clearly originate from the intraband (low-frequency) and interband (high-frequency) transitions. The spectral weight of the intraband transitions is accounted for, within PBCs, by the $\delta(\omega)$ Drude term. The same spectral weight is retrieved, within OBCs, in the low-frequency poles. Previous considerations, based on the results in Fig. 2, also show that such spectral weight accumulates at $\omega = 0$ in the large-system limit. The low-frequency peak in the OBCs conductivity is indeed the main focus of the present work; we are going to closely investigate it in the following.

To start with, it is expedient to compare Fig. 5, left panel, to the free-electron case at the same density, Fig. 1. We clearly see that the effect of switching the periodic potential on is essentially a rescaling of the Drude peak: part of its spectral weight is transferred to the regular conductivity term.

In the flat region between the two OBCs contributions the conductivity $\sigma(\omega)$ is (exponentially) vanishing. By choosing $\tilde{\omega}$ in the middle of this region, we partition the OBCs $f$-sum as:

$$\int_0^\infty d\omega \text{ Re } \sigma(\omega) = \frac{\tilde{D}}{2} + \int_{\tilde{\omega}}^\infty d\omega \text{ Re } \sigma(\omega),$$

VII. CONCLUSIONS

The real part of conductivity in bounded metallic systems within OBCs exhibits a qualitatively different behavior from that of analogous systems within PBCs.\textsuperscript{6,12} Such difference stems from the response of the many-electron system to a dc field, which in metals induces free acceleration within PBCs but not within OBCs. Here we have thoroughly investigated the issue at the independent-electron level. The adiabatic inverse inertia of the electrons is measured by the Drude weight $\tilde{D}$,
which has a well known PBCs expression (even beyond independent electrons\textsuperscript{2}), while instead it is formally zero within OBCs.

Upon general grounds, one expects that both kinds of boundary conditions should produce a given intensive observable in the thermodynamic limit: we solve here the apparent paradox, and we show how to actually evaluate $D$ from the OBCs Kubo formula for conductivity. Simulations on simple paradigmatic model 1$d$ systems validate our theory to a very high numerical accuracy.

An oscillating low-frequency field induces in a metal forced oscillations, which are dominated by the many-electron inertia: the response carries therefore the same essential information as the response to a constant field within PBCs. Both frameworks allow therefore the evaluation of $D$ in two formally different ways. We conjecture that such general principle applies in general to any metallic many-electron system, well beyond the simple models thoroughly addressed in this work.

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\textsuperscript{*} resta@iom.cnr.it

\textsuperscript{1} D. Vanderbilt, \textit{Berry Phases in Electronic Structure Theory} (Cambridge University Press, Cambridge, 2018).

\textsuperscript{2} W. Kohn, Phys. Rev. \textbf{133}, A171 (1964).

\textsuperscript{3} W. Kohn, in \textit{Many–Body Physics}, edited by C. DeWitt and R. Balian (Gordon and Breach, New York, 1968), p. 351.

\textsuperscript{4} D. J. Scalapino, S. R. White, and S. C. Zhang, Phys. Rev. \textbf{47}, 7995 (1993).

\textsuperscript{5} P. B. Allen, in: \textit{Conceptual foundations of materials: A standard model for ground- and excited-state properties}, S.G. Louie and M.L. Cohen, eds. (Elsevier, 2006), p. 139.

\textsuperscript{6} M. Rigol and B. S. Shastry, Phys. Rev. B \textbf{77}, 161101(R) (2008).

\textsuperscript{7} M. Graf and P. Vogl, Phys. Rev. B \textbf{31}, 4940 (1995).

\textsuperscript{8} I. Souza, T. Wilkens, and R. M. Martin, Phys. Rev. B \textbf{62}, 1666 (2000).

\textsuperscript{9} R. Resta, J. Phys. Condens. Matter \textbf{30}, 414001 (2018).

\textsuperscript{10} P. Drude, Annalen der Physik. \textbf{306}, 566 (1900).

\textsuperscript{11} N. W. Ashcroft and N. D. Mermin, \textit{Solid State Physics} (Saunders, Philadelphia, 1976), Ch. 1 and Ch.13.

\textsuperscript{12} E. Akkermans, J. Math. Phys. \textbf{38}, 1781 (1997).

\textsuperscript{13} R. Resta, J. Chem. Phys. \textbf{124}, 104104 (2006).

\textsuperscript{14} R. Resta, Riv. Nuovo Cimento \textbf{41}, 463 (2018).

\textsuperscript{15} A. Marrazzo and R. Resta, Phys. Rev. Lett. \textbf{122}, 166602 (2019).