Nearsightedness of electronic matter

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In an earlier paper, W. Kohn had qualitatively introduced the concept of “nearsightedness” of electrons in many-atom systems. It can be viewed as underlying such important ideas as Pauling’s “chemical bond,” “transferability,” and Yang’s computational principle of “divide and conquer.” It describes the fact that, for fixed chemical potential, local electronic properties, such as the density \( n(r) \), depend significantly on the effective external potential only at nearby points. Changes of that potential, no matter how large, beyond a distance \( R \) have limited effects on local electronic properties, which rapidly tend to zero as a function of \( R \). In the present paper, the concept is first sharpened for representative models of uncharged fermions moving in external potentials, and then the effects of electron–electron interactions and of perturbing external charges are discussed.

Let it be a conventional qualitative wisdom among physicists and chemists that, in the absence of long-range ionic interactions, if an atom A in a solid is exchanged for another atom B, the change of the total energy of the system is largely determined by atoms A and B and their near neighbors. Similarly for a molecule M adsorbed on a surface. These are qualitative examples of “nearsightedness.”

Understanding the physics and chemistry of large molecules and solids would have been practically impossible if not for the principle of transferability (1, 2). It is generally accepted that, in the absence of long-range ionic interactions, large molecules or materials systems can be studied and understood one neighborhood at a time, without the necessity of studying the entire system at once. The useful computational method of “divide and conquer” takes advantage of this fact (3).

We can argue that Pauling’s concept (4) of the chemical bond has a well-defined meaning because, to a good approximation, its properties depend only on the relative positions of the bonded atoms and their near neighbors (5). Anything beyond them has little influence on the properties of the chemical bond.

These important concepts, based on decades of empirical and computational work, point to a property of matter that we call “nearsightedness.”

NEMS frequently reminds one of other well-known and well-accepted concepts but, in fact, it is different. To avoid “deadly sins,” let us present a list of what NEM is not:

(i) NEM is not an aspect of linear or higher-order nonlinear response to external perturbations (but does not exclude these).

(ii) NEM is not screening of charges, which renders long-range Coulomb potentials short-range (NEM applies also to neutral fermions).

(iii) NEM does not apply to systems of few electrons or to noninteracting bosons below their condensation temperature (interacting bosons are beyond the scope of this paper).

(iv) NEM is not limited to electrons at \( T = 0^\circ \) but carries over to finite \( T \), including the classical (high \( T \)) limit.

(v) NEM is not limited to macroscopically homogeneous systems. E.g., it applies to a point \( r \) on an interface.

Abbreviations: NEM, nearsightedness of electronic matter; CPU, central processing unit.

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In this article, we communicate the first quantitative results on NEM for 1D, 2D, and 3D noninteracting, periodic electrons and preliminary results for nonperiodic and interacting electrons. We shall see that, no matter how complicated or strong $w(r')$ is, far away from the perturbation the change of electron density has a universal form, which is completely determined by the reflection coefficient, in 1D, or elements of the scattering matrix, in 2D and 3D, evaluated at certain energies. NEM follows from the fact that these coefficients cannot exceed a certain upper bound. Based on these asymptotic estimates, we discuss the nearsightedness range and present an application to linear scaling electronic algorithms.

**Noninteracting Fermions**

We emphasize again that NEM, as a general principle, does not require interactions or screening. It is due to the destructive interference of density (not wave) amplitudes $n_j$ associated with the occupied single-particle eigenstates $\psi_j$.

**One Dimension.** We begin with a model of 1D electrons in a periodic potential $v(x)$ with inversion symmetry, at $T = 0^+$.

The unperturbed Hamiltonian is $[\hbar = 2m = 1]$

$$H_0 = -d^2/dx^2 + v(x), \quad v(x + b) = v(x).$$

[2]

We first restrict the perturbing potential $w(x)$ to vanish for $x > 0$. The density change is given by

$$\Delta n(x) = \frac{1}{\pi i} \int_{C} [G_E(x, x) - G^0_E(x, x)] dE,$$

[3]

where $G^0_E$ and $G_E$ are the unperturbed and perturbed Green’s functions, respectively, and $C$ is a contour surrounding the eigenvalues below $\mu$. The integral in Eq. 3 can be mapped into the complex $k$-plane,

$$\Delta n(x) = 2 \int_{C} R(k) \psi_k(x)^2 dk, \quad x > 0,$$

[4]

where $C$ corresponds to $C$ for $n(x)$ is the reflection coefficient from right to left, and $\psi_k(x)$ are the normalized unperturbed Bloch functions.

**Asymptotics.** For insulators, we can and shall restrict ourselves to $1m k \geq 0$ and to the first Brillouin zone. We denote by $\kappa$ the branch point that connects the highest occupied and the lowest unoccupied bands (13). For the case when $w(x)$ generates no bound states in the insulating gap, we can choose $C$ in Eq. 4 to be the contour $C_1$ in Fig. 2. Because $\psi_k(x) = u_k(x) e^{ikx}$, with $u_k(x)$ periodic of $x$, $\psi_k(x)$ decays exponentially with $x (e^{-\Delta n(x) x})$ for Im $k > 0$ and the asymptotic behavior of $\Delta n(x)$ comes from the points of $C_1$ in the immediate vicinity of $\kappa$. Using the behavior of the Bloch functions near the branch point (13), we find

$$\Delta n(x) \rightarrow 2R(\kappa) \left(\frac{2\pi}{x}\right)^{1/2} s_n(x)^2 e^{-2\pi x},$$

[5]

where $q = \Im \kappa$ and $s_n(x) = \left[\left|k - \kappa\right| u_k(x)\right]_{x \rightarrow \kappa}$ is a real, periodic or antiperiodic function, $s_n(x + b) = s_n(x)$, depending on the band index. $w(x)$ enters in this asymptotic form only through $R(\kappa)$.

The sign of this $\Delta n(x)$ is independent of $x$ and is given by the sign of $R(\kappa)$. The exponential decay constant $q$ in Eq. 5 equals that of the Wannier function of the highest occupied band (14–16), of the density matrix (17). In the limit of a sufficiently small insulating gap $G$ (in units of the width $W$ of the highest occupied band), $q = \sqrt{\alpha} W / G$, where $\alpha$ is the effective mass at the top of the last occupied band. Because $G \sim \sqrt{\alpha}$, $q$ is proportional to $G$, in line with ref. 18.

One can show that $|R(\kappa)|_{\max} = 1$, with the maximum taken over all functions $w(x)$ that generate no bound states in the insulating gap. Thus, $\Delta n(x)$ cannot exceed an upper bound, independent of $w$. If there are bound states, the asymptotic behavior of $\Delta n(x)$, Eq. 5, may change, but NEM remains (unpublished results).

For metals, we integrate Eq. 4 along the contour $C_2$ in Fig. 2. For large $x$,

$$\Delta n(x) \rightarrow \frac{2}{\pi} \Im [R(k_F)\psi_{k_F}(x)^2],$$

[6]

the slowly decaying Friedel oscillations (19). Again, NEM follows from the fact that $|R(k_F)|$ cannot exceed 1, for any $w(x)$.

For perturbed Kronig–Penney models (20), we found that the asymptotic expressions Eqs. 5 and 6 set in after one or two lattice parameters.

**Nearsightedness range $R$.** For a given $\Delta n$, the nearsightedness range $R(r_0, \Delta n)$ at $r_0$ was introduced as the smallest distance such that any scalar perturbation $w(r')$, lying entirely outside this range, produces a density change at $r_0$, $\Delta n(r_0)$, smaller than $\Delta n$. Fully characterized, $R$ is a function of $r_0$, chemical potential $\mu$, and $\Delta n$, and a function of $w(r)$

$$R = R(r_0, |v(r')|, \mu, \Delta n).$$

[7]

From this definition, it follows that, at every fixed $r_0$, $\partial R(r_0, \Delta n)/\partial \Delta n \leq 0$.

To calculate $R$ at a point $x_0$, we need to simultaneously consider all perturbing potentials $w_{L,R}$ to the left and right of $x_0$. In this case, the density change at $x_0$ is given by the individual contributions $\Delta n_{L,R}(x_0)$ of $w_{L,R}$, Eqs. 5 and 6, plus multiple reflection corrections. In the limit when the distance from $x_0$ to $w_{L,R}$ is large, these corrections were found exponentially small for insulators and comparable to $\Delta n_{L,R}(x_0) + \Delta n_{R,L}(x_0)$ for metals.

For insulators, the asymptotic behavior of $R$ in the limit $\Delta n \rightarrow 0$, as derived from the upper bound of Eq. 5, from the above remark and from a cell-averaging of $\Delta n(x)$, is

$$R(\Delta n) \rightarrow \frac{1}{2q} \Im \frac{\tilde{n}}{\Delta n},$$

[8]

where

$$\tilde{n} = \frac{8}{\sqrt{2r}} \int_{x_0}^{b} s_n(x)^2 dx.$$  

[9]

In the small gap and thick binding limits, $\tilde{n}$ is completely determined by the exponential decay constant $q$, $\tilde{n} \rightarrow 4\sqrt{\pi}/q$, and by $\tilde{n} \rightarrow 4\sqrt{q}/\pi b$, respectively.

For metals, the upper bound of Eq. 6, with inclusion of multiple reflections and cell-averaging, leads to the following asymptotic behavior.
The density change for \( y \) is given by

\[
v_y(x) = \lambda \sum_n \delta(x - x_n),
\]

where \( \lambda \) is the density of states at the Fermi energy. Thus, a small random potential increases \( R \) for insulators and decreases it for metals. We conjecture that this remains true for disorder, more generally.

**Higher Dimensions.** We first consider, as an example, 2D fermions in a periodic potential with square symmetry. We present only the cases when the first band, assumed isolated, nondegenerate, and with convex isenergetic lines in \( k \)-space, is partially or completely filled. We first restrict the perturbing potential \( w(x, y) \) to vanish for \( y > 0 \) and the periodicity along the \( x \) direction to be preserved (so that \( k_x \) remains a good quantum number).

The density change for \( y \to \infty \) is given by

\[
\Delta n(\vec{r}) \to 2 \int_{|\vec{k}'| < |\vec{k}|} S(\vec{k}, \vec{k}') \psi_{\vec{k}}(\vec{r}) \psi_{\vec{k}'}(\vec{r}) d\vec{k},
\]

with \( \vec{k}' = (k_y, -k_x) \) and \( S(\vec{k}, \vec{k}') \) the scattering matrix element between \( \vec{k} \) and \( \vec{k}' \).

**Asymptotics.** For fixed \( k_x \), the analytic structure relative to \( k_y \) of the band energy \( E_k \) and Bloch function \( \psi_k \) of the first band is completely analogous to that in 1D (13): \( E_k \) and \( \psi_k \) have branch points of order 1 and 3, respectively, at \( k_y = \pm \pi + iq(k_x) \), connecting the first band with a higher band (we restrict ourselves to \( k_x \geq 0 \) and to the first Brillouin zone). Their behavior near these points is the same as in 1D, namely \( E_k \) behaves as a square root and \( \psi_k \) diverges as \( (k_y - k_y)^{-1/4} \) (unpublished work).

If the band is completely filled and \( w(\vec{r}) \) does not generate bound states in the insulating gap, for a given \( k_x \), the integral over \( k_y \) in Eq. 15 can be taken over a contour surrounding the branch point (as in Fig. 2) and its asymptotic behavior can be determined as in the 1D case. Thus, the integral over \( k_y \) decays exponentially, as function of \( y \), with a rate \( 2q(k_x) \). There will be two values, \( \pm k_x \), of \( k_x \) where \( q(k_x) \) reaches its lowest value \( q_0 \). The asymptotic behavior of \( \Delta n(\vec{r}) \), for \( y \to \infty \), comes from the immediate vicinity of these points. Defining \( \beta = (\partial^2 q/\partial k_x^2)_{k_x = k_x} \) and \( k_0 = (k_x, \pi + iq_0) \), \( k_0 = (k_x, \pi - iq_0) \), gives

\[
\Delta n(\vec{r}) \to \frac{4\pi}{y} \sqrt{\frac{2}{\beta}} \text{Re} \{ S(k_0, k_0) \psi_{k_0}^*(\vec{r}) \psi_{k_0}(\vec{r}) \} e^{-2\beta y},
\]

where \( \psi_{k_0}(\vec{r}) = \sqrt{[k_y - k_x]^{1/4} e^{-ik_y q_y}} \psi(\vec{r}) e^{-ik_x y} \) is a quasiperiodic function in \( x \) and \( y \). Again, \( w(\vec{r}) \) enters in this asymptotic form only through \( S(k_0, k_0) \).

The exponential decay in Eq. 16 is twice as fast as the exponential decay, in the \( y \) direction, of the density matrix or of the Wannier function of the first band. \( |S(k_0, k_0)| = 1 \) for a hard wall and, in general, we expect it to be of order 1.

If the band is partially filled, the asymptotic behavior of \( \Delta n(\vec{r}) \) is determined by the two points on the Fermi surface, denoted by \( k = (k_x, k_y) \) and \( k_0 = (k_x, k_y) \), where the tangent to the Fermi surface is along the \( k_x \) direction:

\[
\Delta n(\vec{r}) \to 2\text{Im} \sqrt{-\frac{i}{\pi y}} S(k_0, k_0) \psi_{k_0}^*(\vec{r}) \psi_{k_0}(\vec{r}),
\]

with \( \eta \) the curvature of the Fermi surface at these points. From the unitarity of the scattering matrix, one can immediately find that \( |S(k_0, k_0)| = 1 \), with the maximum taken over all \( \psi(\vec{r}) \). Thus, the asymptotic density change cannot exceed an upper bound, no matter how large the perturbing potential is.

The 3D case is analogous.

**Nearbandgap range R.** For metals, the simplest model is jellium enclosed by a spherical hard wall, for which one easily finds

\[
R(\Delta n) \to \left\{ \begin{array}{ll}
2.2 r_\eta / 2 \Delta n = 2.2 r_\eta / \Delta n & (2D) \\
2.5 r_\eta / 2 \Delta n = 2.5 r_\eta / \Delta n & (3D),
\end{array} \right.
\]

where \( r_\eta = 1.25 \) is the density of the uniform gas and \( r_\eta = 1.3 \) is the Wigner-Seitz radius. For metals in periodic potentials, the \( \Delta n \) dependence remains unchanged, but \( k_0 \) is replaced by a \( k_{eff} \) depending on the band structure and the filling.

For an insulator with square symmetry, we calculated \( \Delta n(\vec{r}) \) due to enclosing the point \( r_0 \) in four hard walls along the symmetry axes, at a distance \( D \) from \( r_0 \). The density change near \( r_0 \) is given by the sum of the changes due to each individual, infinitely extended wall, Eq. 16, plus multiple reflection corrections, which were found to be exponentially negligible in the limit \( D \to \infty \). Similarly for a 3D insulator with cubic symmetry. From Eq. 16, its 3D analog, and the previous remark, we find that the cell-averaged density change at \( r \) becomes less than a given \( \Delta n \) for \( D \approx D(\Delta n) \),

\[
D(\Delta n) \to \frac{1}{2q_{eff}} \ln \frac{\hat{n}}{\Delta n},
\]

where \( \hat{n} \) can be easily calculated from the band structure. Finding an analytic expression of the nearbandgap range \( R \) for a general 2D or 3D insulator is clearly a next-to-impossible task. However, on the basis of the above calculations, the proof of exponential localization of 2D and 3D Wannier functions (22, 23) and of 1D generalized Wannier functions (14, 15), as well as our 1D result, Eqs. 5 and 8, we expect results of the following form for 2D and 3D insulators:

\[
R(\Delta n) \to \left\{ \begin{array}{ll}
\frac{1}{2q_{eff}} \ln \frac{\hat{n}}{\Delta n} & (d = 2, 3),
\end{array} \right.
\]
is the error due to the hard walls around $B_n$ of each $V_n$, primarily from the nearest walls. For metals, Eqs. \ref{1} and \ref{2} define the physical basis of linear scaling. Here, we exemplify this for “cubic” periodic systems in 1D, 2D, and 3D, respectively. Similarly, for insulators, $b \to (2\pi)^{-1}\ln[\sqrt{n}/2\Delta n]$, with $n$ defined in Eqs. \ref{1} and \ref{2}.

The central processing unit (CPU) time for electronic structure calculations of a system consisting of many ($N_a$) atoms grows very rapidly with $N_a$ if the calculations are performed for the entire system at once. It has been pointed out (3, 24) that the dependence on $N_a$ can be made linear for large $N_a$ by dividing the system into $N_s$ suitable subsystems, where $N_s \approx N_a$. In ref. 6, NEM was identified as the physical basis of linear scaling. Here, we quantify this idea.

The procedure is illustrated in Fig. 3 [a detailed discussion (including self-consistency) can be found in ref. 24]. The system of volume $V$ is divided into segments $V_n$, with overlapping buffer zones $B_n$. The density $n(r)$ and contribution to the total energy of each $V_n$ are obtained from calculations, including its buffer zone. For a required accuracy $\Delta n$, the thickness $b$ of $B_n$ is chosen so that, when $r$ is on the boundary of $V_n$, $\Delta n(r) \leq \Delta n$, where $\Delta n(r)$ is the error due to the hard walls around $B_n$. An upper bound for $b$ is given by the maximum of $R(\Delta n)$ on the boundary of $V_n$. Finally, the size of each $V_n$ is chosen to minimize the total CPU time.

We exemplify this for “cubic” periodic systems in 1D, 2D, and 3D, where the $V_n$ are all identical “cubes” of edge size $a$. The largest density change occurs at the “corners” of $V_n$ and comes primarily from the nearest walls. For metals, Eqs. \ref{3}, \ref{4}, and their 3D analog, plus the inclusion of the multiple reflections, lead to $b \to \chi a \left(\ln(\sqrt{n}/\Delta n)\right)^{1/2}$. For a 5% accuracy, $b = 6.2, 6.7$, and 4.5$a$, respectively. For insulators, $b \to (2\pi)^{-1}\ln[\sqrt{n}/2\Delta n]$, with $n$ defined in Eqs. \ref{1} and \ref{2}.

The total CPU time is given by $t \approx N_a r$, where $N_s = (V/a^d)$ is the number of segments, and $r \approx (2b + a)^{-d}$ is the CPU time for the electronic structure calculation of one segment plus its buffer zone ($r \approx 2–3$ for DFT (density functional theory) (24), and higher for other methods). Minimizing the total CPU time with respect to $a$, we obtain the optimal size, $a = 2b/(\nu - 1)$. With this optimization, and from our estimates of $b$, we obtain the following dependence of the total CPU time on the desired accuracy and the total number of atoms:

$$t \approx N_a \times \left\{ \begin{array}{ll}
(\Delta n)^{2(1-\nu d)/(d+1)} & \text{(ungapped)} \\
(\ln(\sqrt{n}/2\Delta n))^{1-1/d} & \text{(gapped)}
\end{array} \right. \quad \begin{array}{l}
[21]
\end{array}
$$

For metals, $b$ can be greatly reduced by averaging the wall-induced Friedel oscillations over two or more values of $b$.

Interacting Fermions

In considering the response of charged fermions to distant disturbances it is necessary to distinguish between two cases:

(i) Distant perturbing potentials, $w(r)$, with $|r_0 - r| \geq R$. The simplest description of many-body interaction effects is the random phase approximation (RPA). Within RPA, we found that, in all dimensions, the many-body interaction leads to a decrease of $R$ in typical metals but an increase of $R$ in typical insulators due to a reduction in the gap.

(ii) Distant perturbing charge densities $\rho(r)$. In analogy with $R(r_0, \Delta n)$, we define a charge-nearsightedness range, $R_c(r_0, \Delta n)$ as the smallest distance such that any charge perturbation $\rho(r)$ lying entirely outside this range produces a density change at $r_0$, $\Delta n(r_0)$, smaller than $\Delta n$.

As is well known, the long-range Coulomb potential, because of perturbing electric charges, is screened out by metallic electrons. Preliminary model calculations for metallic electrons, in the Thomas–Fermi approximation, indicate that they are charge-nearsighted, i.e., have a finite $R_c$. However, charged insulating fermions are “classically farsighted,” in the sense that, at sufficiently large distances, the fermions “see” the classical long-range total potential $\int \rho(r)/|r_0 - r|dr'$, where $\rho$ is the total perturbing charge density, including depolarization. Thus, for example, in metals, replacing a neutral atom or ion by another atom or ion always has short-range electronic consequences, whereas in an insulator ions lead to classical long-range electronic effects.

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