Locality properties and Wannier functions for interacting systems

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Abstract

We define Wannier functions for interacting systems, and show that the results on the localization of the Wannier functions for non-interacting systems carry over to the Wannier functions for interacting systems. In addition we demonstrate that the characterization of metals and insulators by the decay properties of their respective density matrices does not only apply to non-interacting, but also to interacting systems. As a prototypical example of a correlated system we investigate the one-dimensional Hubbard model. We propose an expression for the density matrix of that model, and derive a relation between the decay constant of the density matrix and the gap.

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The locality properties of solids, their so called nearsightedness, have recently moved to the focus of much attention [1–7]. This is largely due to the intense efforts going into the development of electronic structure methods that scale linearly with system size [5,6]. These methods depend crucially on the locality of the density matrix. The closely related issue of the locality of the Wannier functions [8] has also attracted renewed interest, largely due to the development of methods for the practical construction of localized Wannier functions [4]. The uses of Wannier functions are well proven. First, they are fundamental in the theory of electron dynamics in the presence of weak external fields [9].

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Second, they allow for an intuitive interpretation of the bonding properties in solids [4]: Localized Wannier functions correspond to either bonds or lone electron pairs. Third, they are at the center of the modern theory of polarization [10]. Finally, they are important for some linear scaling algorithms [5,6].

General results on the localization of the Wannier functions are quite difficult to obtain. The problem has actually been called “one of the few basic questions of the quantum theory of periodic solids in the one-electron approximation which is not completely solved” [11]. The most important results so far are the proofs of the existence of exponentially localized Wannier functions for isolated, simple bands in any dimension [11], and for complex bands in the tight-binding limit, and in perturbation theory [12]. For practical applications knowledge of the decay constants is of considerable importance. First results were obtained in [13], considerably more general results for the density matrix were given in [2].

Trying to extend the results to interacting systems we are faced with a fundamental problem: The Hamiltonian is no longer a one-body operator. Hence it seems to be impossible to define meaningful single-electron Bloch waves and the corresponding Wannier functions. This is true already in the limit of weak interactions, although there one would expect that the non-interacting Wannier functions still could be useful. We can, however, avoid the problem of having to deal with a many-body operator, by changing perspective. Looking at the one-body density matrix, we realize that it can replace the (single-electron) Hamiltonian in the standard construction of Wannier functions. We are thus led to Wannier functions made from natural orbitals [15], which we christen natural Wannier functions. Instead of the energy bands $\varepsilon_j(k)$ we now consider occupation bands $n_j(k)$. We prove that the projection operator $P(k)$ onto an isolated set of occupation bands is analytic in $k$. Once this result is established, the results [11,12] obtained for the standard Wannier functions immediately carry over to the natural Wannier functions. So the natural Wannier functions can be considered as the natural generalization of the concept of Wannier functions to interacting systems. Being constructed from natural orbitals, they are an optimal basis, meaning that it is sufficient to consider only the bands $n_j(k)$ with high occupation to obtain a good description of the interacting system [15]. Being Wannier functions, they are localized, thus allowing to take advantage of $O(N)$ methods in evaluating, e.g., Coulomb matrix elements. Moreover, they can be expected to represent the chemical bonding in the correlated system, as do the ordinary Wannier functions in the independent-particle case. We furthermore show that the characterization of metals and insulators in terms of the decay properties of the density-matrix [14], also applies to correlated systems. Thus the decay constant $\gamma$ may play a similar role as the localization length defined in [7]. Finally, we consider the one-dimensional Hubbard model as an explicit example of a correlated system, deriving a relation giving the decay constant $\gamma$ as a function of the gap, thus
extending the results of [2] to an interacting system.

We start our construction from the many-body wavefunction \( \Psi(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_N) \). For simplicity we consider spinless electrons. The one-body density matrix \( D \) is then given by

\[
D(\mathbf{r}, \mathbf{r}') = \int d\mathbf{r}_2 \cdots d\mathbf{r}_N \Psi^*(\mathbf{r}', \mathbf{r}_2, \ldots, \mathbf{r}_N) \Psi(\mathbf{r}, \mathbf{r}_2, \ldots, \mathbf{r}_N).
\]

Its eigenfunctions are the natural orbitals, its eigenvalues the natural occupation numbers. Clearly, \( D \) is Hermitian. Furthermore, translating all the spatial arguments of \( \Psi \) by a lattice vector \( \mathbf{R} \) multiplies the wavefunction by a phase factor. Since these phase factors cancel inside the density matrix, we find \( D(\mathbf{r} + \mathbf{R}, \mathbf{r}' + \mathbf{R}) = D(\mathbf{r}, \mathbf{r}') \). Thus the Bloch theorem applies and, in analogy to the Hamiltonian of a periodic solid in an independent-particle picture [16], the eigenvalues of the density matrix form bands \( n_j(\mathbf{k}) \), the occupation bands, with the corresponding natural orbitals being Bloch functions

\[
\Phi_{j,\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} U_{j,\mathbf{k}}(\mathbf{r}),
\]

where \( j \) is the band index, \( \mathbf{k} \) a vector in the Brillouin zone, and the \( U_{j,\mathbf{k}}(\mathbf{r}) \) are periodic functions with respect to the real space primitive cell. They are the eigenvectors of the \( \mathbf{k} \)-dependent density matrix

\[
D_{\mathbf{k}} = \sum_j n_j(\mathbf{k}) |U_{j,\mathbf{k}}\rangle\langle U_{j,\mathbf{k}}|.
\]

Using the standard prescription [16], we can then construct Wannier functions, which, for obvious reasons, we call natural Wannier functions:

\[
W_j(\mathbf{r} - \mathbf{R}) = \frac{V}{(2\pi)^3} \int_{BZ} d\mathbf{k} e^{i\mathbf{k}\cdot(\mathbf{r} - \mathbf{R})} U_{j,\mathbf{k}}(\mathbf{r});
\]

where \( V \) is the volume of the real-space cell and the integration is over the Brillouin zone (BZ).

The construction of the natural Wannier functions is very similar to that of the conventional Wannier functions for a non-interacting system, the density matrix \( D \) taking the place of the non-interacting Hamiltonian. In the limit of vanishing interaction, the natural Wannier functions do, however, not reduce to the conventional Wannier functions. This is clear, since in the limit of no interaction the density matrix becomes the projector onto the occupied subspace; i.e. all occupied (unoccupied) states are degenerate with eigenvalue one (zero). By degenerate perturbation theory, in the limit of vanishing interaction, the natural Wannier functions will therefore have to diagonalize the
first term in the perturbation expansion in their respective subspace (occupied or unoccupied) [15]. They are therefore generalized Wannier functions in the sense of [4], which, respectively, span the space of occupied and unoccupied states. Clearly, using several bands increases the flexibility to construct more localized Wannier functions, since one can take advantage of the unitary transformations allowed in the space spanned by the Bloch functions of different bands. In the extreme limit, where we allow all bands, it is obviously possible to construct Wannier functions that are perfectly localized delta functions. In this respect it is interesting to note that quantum Monte Carlo calculations of the lowest natural orbitals show that they are very similar to the occupied Kohn-Sham orbitals [17]. Hence the corresponding natural Wannier functions are also expected to be very similar to their Kohn-Sham counterparts. This similarity is presumably no longer valid for the Wannier functions arising from the unoccupied bands. Whereas the virtual Kohn-Sham Wannier functions are much less localized than the occupied ones [18] this seems not to be the case for the natural Wannier functions.

We now want to show under what conditions the natural Wannier functions are exponentially localized. The key input to the corresponding proofs for the ordinary Wannier functions [11,12] is the analyticity of the projection operator onto the band states. The proof for energy-bands was given in [14]. For the natural Wannier functions we have to prove the analyticity on a strip $K = \{ k' + ik'' , |k''| < A \}$ of the projector $P(k)$ onto an isolated set $B$ of occupation bands. Because of the special properties of the density matrix, the proof is straightforward. $D_k$ is hermitian for real $k$; $N$-representability [15] requires for the eigenvalues $n_i \in [0,1]$; moreover, since a unit cell contains only a finite number of electrons and the bands are continuous, $\text{Tr} D_k$ is finite, i.e. $D_k$ is trace class, and therefore, in particular, compact. If we assume that $D_k$ is analytic on $K$, we can apply the analytic Fredholm theorem [19], which guarantees that the resolvent of $D_k$ is meromorphic on $K$ with the residues at the eigenvalues being finite rank operators. Choosing a contour $C$ in $K$, which exclusively encircles all occupation numbers in $B$ (here we use that the bands in $B$ are isolated, i.e. they do not intersect with, or touch any band not in $B$), we obtain the occupation band projector

$$P(k) = \frac{1}{2\pi i} \int_C \frac{d\eta}{\eta - D_k}, \quad (3)$$

which, as desired, is analytic on $K$. Given the analytic band projection operator, the proofs given in [11,12], with the Hamiltonian replaced by the one-body density matrix, guarantee the existence of exponentially localized Wannier functions if $B$ contains only a single band, or, for complex bands $B$, in the tight-binding limit and in perturbation theory (of $D_k$) around a situation, where there do exist exponentially localized Wannier functions.
In the proof given above, we assumed that the $k$-dependent density matrix is analytic in $k$. It is then natural to ask what the analyticity of $D_k$ means. Let us therefore distinguish two cases: Either $D_k$ is analytic in $k$ or it has a non-analyticity at some $k = k_F$. Then, in the absence of degeneracies, in the first case, the occupation band structure $n_j(k)$ will be analytic in $k$ [19], while in the second case it will have a non-analyticity at $k_F$. In the first case there is no Fermi surface, and we thus associate this case with an insulator. As for the second case, assuming that perturbation theory holds, a discontinuity in $n_j(k)$ implies a Fermi liquid, while an algebraic singularity would point to Luttinger liquid behavior. We thus associate this case with a metal. But we note that we are not aware of a proof that a discontinuity in $n_j(k)$ by itself guarantees metallic behavior even when perturbation theory breaks down [20]. It is, however, hard to imagine an insulator with such a discontinuity, in particular, since it would also show up in the momentum distribution $N(p) = \int d\mathbf{r}d\mathbf{r}' \exp(-i\mathbf{p}(\mathbf{r} - \mathbf{r}'))D(\mathbf{r}, \mathbf{r}')$, which is, in contrast to $n_j(k)$, an experimentally accessible quantity. From the Paley-Wiener theorem [19] (see also [14]), it follows that for an insulator the density matrix, being the Fourier transform of $D_k(\mathbf{r}, \mathbf{r}')$, decays exponentially with increasing distance $|\mathbf{r} - \mathbf{r}'|$. In the three-dimensional case, the decay constant will in general be different along different directions. For a metal, on the other hand, where $D_k(\mathbf{r}', \mathbf{r})$ is non-analytic on the Fermi surface, the density matrix decays only algebraically. The decay properties of the density matrix for interacting systems are thus qualitatively the same as for non-interacting systems [14]. We emphasize that this result is valid for zero temperature, while at finite $T$ the decay should become exponential also for a metal [21].

As a specific and important example, we now analyze the localization properties of the density-matrix for a prototypical correlated system, the one-dimensional Hubbard model (with lattice constant $a$ and nearest-neighbor hopping matrix element $t$) [22]

$$H = -t \sum_{\langle i,j \rangle, \sigma} c^\dagger_{j\sigma} c_{i\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}.$$  \hspace{1cm} (4)

For $U = 0$ the system is metallic, while for any finite $U$ it is a Mott insulator [23]. Motivated by the result shown in Fig. 1 we choose as an Ansatz the product of the exact density matrix for $U = 0$ and an exponential factor:

$$D_{0,m} = \langle \Psi | c^\dagger_m c_{0\sigma} | \Psi \rangle = \frac{\sin(\pi m/2)}{\pi m} e^{-\gamma a|m|}.$$ \hspace{1cm} (5)

To estimate the decay constant, we calculate $D_{0,1}$ from the exact ground-state energy of the Hubbard chain [23]. Using the Hellmann-Feynman theorem we
Fig. 1. Exponential decay of the density matrix for the Hubbard chain. The symbols give the numerical values of the density matrix for a chain of 16 sites. The lines connect the values of Ansatz (5), using the decay constants given in (7).

find

$$D_{0,1} = -\frac{1}{4}\epsilon_{\text{kin}}(U)/t = -\frac{1}{4}\left(1 - U \frac{d}{dU}\right) e(U)/t,$$

which, together with the Ansatz (5), yields

$$\gamma_a = -\ln\left(\pi \int_0^\infty dx \frac{J_0^2(x) - J_1^2(x)}{1 + e^{Ux/2t}}\right),$$

where $J_0$ and $J_1$ are Bessel functions. Fig. 2 shows a plot of $\gamma_a$ as a function of the gap $E_g/t = U/t - 4 + 8 \int_0^\infty dx J_1(x)/(x(1 + \exp(Ux/2t)))$. For large $E_g$ we find the asymptotic behavior

$$\gamma_a = -\ln\left(\frac{2\pi \ln(2)}{E_g/t + 4}\right),$$

while for small gap there exists no expansion, as for the Hubbard chain the point $U = 0$ is non-analytic. The behavior of $\gamma_a$ is thus qualitatively different in both the large and small gap limit from the analytical results for non-interacting systems (cf. Fig. 1 in Ref. [2]), although the overall shapes of the curves look similar. We note that the decay of the density matrix (5) has a power-law exponent different from the universal exponent for the non-interacting density-matrix found in [24].

Fourier transforming the Ansatz (5), we obtain

$$n(k) = \frac{1}{2} + \frac{1}{\pi} \arctan\left(\frac{\cos(ka)}{\sinh(\gamma_a)}\right).$$
Fig. 2. Decay constant $\gamma a$ of the density matrix for the Hubbard chain as function of the gap $E_g$. The dashed line shows the large-$U$ approximation (8).

Fig. 3. Occupation band structure for periodic Hubbard chains (determined by exact diagonalization) compared to (9) with $\gamma a$ obtained from (7). All curves still differ significantly from the expression for $n(k)$ in the large-$U$ limit [25], which, for $U = 12$, is plotted for comparison.

By construction, this is the step function for $U \to 0$, while in the limit of large $U$ we have $n(k) = \frac{1}{2} + \frac{\ln(2)}{2U} \cos(ka)$; i.e. our Ansatz (5) is exact, both for $U \to 0$ and $U \to \infty$ [25]. For intermediate $U$, we compare (9) with the momentum distribution obtained from exact diagonalization of finite Hubbard chains. This is shown in Fig. 3: For $U$ large, but still far from the large-$U$ limit, the agreement is perfect, and even for fairly small $U$, where the decay length $1/\gamma a$ is of the order of the chain length, the agreement between (9), which was derived for the infinite chain, and the results for the finite rings is amazingly good.

The half-filled Hubbard chain is a Mott insulator for $U > 0$, only for $U = 0$ it is a metal. As expected, this is reflected in the decay properties of the density matrix. While for $U = 0$ it decays as $1/m$, for any finite $U$ the decay is exponential. It is interesting to compare the decay constant $\gamma a$ with the localization length $\lambda/a = \sqrt{d}/2\pi$, with $d = -\lim_{N \to \infty} N \ln |z_N|^2$ and $z_N =$
\[ \langle \Psi_0 | e^{\frac{2\pi i}{N} \sum \hat{x}_i} | \Psi_0 \rangle, \] as defined in [7]. We find that both criteria describe the metal-insulator transition correctly. As a surprising fact we note that \(1/\lambda\) seems to be linear in \(U\) down to very small values of \(U\), with the constant of proportionality given by a large \(U\) expansion.

Away from half-filling we have tried an Ansatz in the spirit of (5) of the form
\[ \frac{\sin(k_F m)}{\pi m} |m + 1|^{-\alpha} \]
Fourier transforming leads to an \(n(k)\) with a Luttinger-like singularity with exponent \(\alpha\) at \(k_F\). It fails, however, to also produce a singularity at \(3k_F\) [26].

To summarize, we have defined natural Wannier functions for interacting systems from the eigenfunctions of the density matrix, and have shown under what conditions they can be proven to be exponentially localized. The natural Wannier functions provide an optimal, localized basis for describing a correlated system. In terms of computational efficiency, they will allow the use of \(O(n)\) methods in many-body calculations. We also expect them to provide understanding of the bonding in correlated solids. In addition, we have shown that the characterization of metals and insulators by the decay properties of the density matrix does also apply to interacting systems, at least as long as perturbation theory holds. Finally, we have investigated the one-dimensional Hubbard model, proposed an expression for the density matrix of that model, and, for this Mott insulator, derived a relation between the decay constant of the density matrix and the gap.

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Note added in proof: After finishing the present work we became aware of the paper [27], in which also introduces natural Wannier functions. We note, however, that in that paper the analyticity of the Bloch functions for simple bands is take for granted, rather than proven.

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