Exponential localization of Wannier functions in insulators
Christian Brouder, Gianluca Panati, Matteo Calandra, Christophe Mourougane, Nicola Marzari

To cite this version:

HAL Id: hal-00082714
https://hal.science/hal-00082714v2
Submitted on 7 Mar 2007
Exponential localization of Wannier functions in insulators

Christian Brouder,1 Gianluca Panati,2 Matteo Calandra,1 Christophe Mourougane,3 and Nicola Marzari4

1Institut de Minéralogie et de Physique des Milieux Condensés, CNRS UMR 7590, Université Paris 6 et 7, IPGP, 140 rue de Lourmel, 75015 Paris, France.
2Zentrum Mathematik und Physik Department, Technische Universität München, 80290 München, Germany.
3Institut de Mathématiques de Jussieu, 175 rue du Chevaleret, 75013 Paris, France.
4Department of Materials Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge MA 02139-4307, USA.

(Dated: March 7, 2007)

The exponential localization of Wannier functions in two or three dimensions is proven for all insulators that display time-reversal symmetry, settling a long-standing conjecture. Our proof relies on the equivalence between the existence of analytic quasi-Bloch functions and the nullity of the Chern numbers (or of the Hall current) for the system under consideration. The same equivalence implies that Chern insulators cannot display exponentially localized Wannier functions. An explicit condition for the reality of the Wannier functions is identified.

PACS numbers: 71.23.An, 03.65.Vf

Wannier functions play a fundamental role in the description of the electronic properties of solids [1]. They allow for an intuitive interpretation of the bonding properties of solids [2], they are at the center of the modern theory of polarization [3], and they form a very efficient basis for order-N calculations or the construction of model Hamiltonians [4].

For one-dimensional systems, Kohn [5] proved that Wannier functions are exponentially localized. This property has many desirable consequences, such as the existence of moments (\(r^n\)) for all \(n\), the exponential convergence of numerical calculations [6], and the possibility to use Wannier functions for the description of surfaces [7]. In two and three dimensions, the existence of exponentially localized Wannier functions is one of the few unsolved problems of one-particle condensed-matter physics [8]. In the absence of a proof for exponential decay, this property is commonly used as working hypothesis [9], checked with numerical calculations [10], or simply taken for granted [11, 12].

In this paper, we demonstrate that exponentially localized Wannier functions exist for insulators in two and three dimensions [13]. To prove this, we show the equivalence between the existence of analytic quasi-Bloch functions and the nullity of the Chern numbers (or of the Hall current) for the system under consideration. The same equivalence implies that Chern insulators cannot display exponentially localized Wannier functions. An explicit condition for the reality of the Wannier functions is identified.

Wannier functions are defined as

\[ u_{n\mathbf{k}}(\mathbf{r} + \mathbf{R}) = e^{-i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}). \]

where \(u_{n\mathbf{k}}(\mathbf{r})\) is a periodic eigenstate of the Hamiltonian \(H = -\Delta + V(\mathbf{r})\) (in Ry), where the potential \(V\) is real and periodic. According to Bloch’s theorem, the eigenfunctions of \(H\) can be written as \(\psi_{n\mathbf{k}}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r})\), where the Bloch states \(u_{n\mathbf{k}}(\mathbf{r})\) are periodic eigenstates of the Hamiltonian \(H(\mathbf{k}) = -i\nabla + \mathbf{k}^2 + V(\mathbf{r})\) with energy \(\epsilon_n(\mathbf{k})\) and satisfy the boundary conditions

\[ u_{n\mathbf{k}+\mathbf{G}}(\mathbf{r}) = e^{-i\mathbf{G}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}). \]  

In the simplest procedure, the energies \(\epsilon_n(\mathbf{k})\) and the Bloch functions \(u_{n\mathbf{k}}\) are determined as a function of \(\mathbf{k}\) by ordering the eigenvalues by increasing energies \(\epsilon_1(\mathbf{k}) \leq \epsilon_2(\mathbf{k}) \leq \ldots\). Although this procedure is standard in band-structure calculations, it gives Bloch states \(u_{n\mathbf{k}}\) that are not regular in \(\mathbf{k}\) since the phase of \(u_{n\mathbf{k}}\) is random. Moreover, at a crossing point the energy \(\epsilon_n(\mathbf{k})\) can have a kink. In one dimension, Kohn [5] showed that it is possible to define Bloch states that are analytic functions of \(\mathbf{k}\). In two and three dimensions this is generally not possible [13] because band crossings lead to Wannier functions that can decay as \(1/R^{\alpha}\) [2].

Blount [22] noticed that the decay properties of Wannier functions can be improved by considering a set of eigenstates, called a composite band [23], which are...
separated by a gap from all others. More precisely, if ε₁(k),...,ε_M(k) are the eigenstates of the composite band, we assume that there is an a > 0 such that |εₙ(k) - εₘ(k)| > a for any k in Ω, 1 ≤ n ≤ M and m > M. In insulators it is always possible to define a composite band since gaps exist between valence and conduction bands and between valence bands and core states. This is not necessarily the case in metals. An example of a composite band is given in Fig. 1 for silicon. Exponentially localized Wannier functions are obtained if we can find M analytic functions vₙ(k) (called quasi-Bloch states) that span the same vector space as the M Bloch states of the composite band. The problem of the existence of analytic quasi-Bloch states was solved for a single isolated band (i.e. M = 1) in 3D by des Cloizeaux [8] and Nenciu [3]. In this work we determine when analytic quasi-Bloch functions exist for any composite band in two and three dimensions.

The quasi-Bloch functions vₙ(k) can be expressed as

\[ vₙ(k) = \sum m uₙm(k) Uₘn(k), \]

where U(k) is a unitary matrix, defining the Wannier functions

\[ wₙ(r) = \frac{1}{|Ω|} \int_{Ω} dk e^{i k ⋅ r} \sum m uₙm(k) Uₘn(k). \]  

The improved localization of these Wannier functions was observed on many systems [8][23].

To study the analytic properties of the Bloch functions, we consider a complex vector k = k′ + i k″. Then, H(k) is not Hermitian but there still are eigenvalues εₙ(k) and eigenstates uₙk such that H(k)uₙk = εₙ(k)uₙk. The branch points of εₙ(k) determine the points of non-analyticity of the function uₙk [8][22]. Even very simple crystals, such as silicon, exhibit such branch points [22].

If the potential V(r) is square integrable, it can be shown that H(k) is analytic [22]. The reality of V(r) implies that H*(k) = H(k*) and H*(k) = H*(-k*). From a practical point of view, the condition of square integrability of V(r) encompasses potentials with Coulomb singularities and the potentials used in LDA and GGA calculations. The calculation of band structures in the complex plane is available in some standard band-structure packages [27].

Several authors [8][8][23] noted that the obstacles to the existence of quasi-Bloch states are topological. To illustrate this, we introduce a simple mathematical concept, that of a fibre. It is quite common in physics to consider a space (let us call it the base) and to associate a vector space (called the fibre) to each point of the base. For example, in magnetostatics, the base B is the space R³ and to each point r of the base we associate a three-dimensional vector space (the fibre F(r) = R³). The vector potential A(r) is a vector of the space F(r). In the Born-Oppenheimer approximation, the base B is the set of possible positions of the nuclei R₁,...,R_N and the fiber F(R₁,...,R_N) corresponding to a nuclear configuration is the vector space generated by the solutions of the Schrödinger equation with clamped nuclei. For a composite band, the base is the Brillouin zone Ω and the fibre F(k) is the M-dimensional vector space generated by the Bloch states uₙk. The quasi-Bloch states vₙk can now be defined precisely as a basis of F(k) such that each vₙk is analytic in k, periodic in the reciprocal space and satisfies the boundary conditions [8].

To show that topology might forbid the existence of quasi-Bloch states, we consider the simple example where the base B is a circle represented by the angular variable x ∈ [0, 2π] and the fibre is F(x) = R. We denote by v(x) a basis of F(x). Topology intervenes when we determine how the fiber at x = 0 is related to the fiber at x = 2π: we can use periodic boundary conditions v(2π) = v(0) (cylinder) or antiperiodic boundary conditions v(2π) = -v(0) (Möbius strip, illustrated in Fig. 2). In the case of a cylinder, we can choose a basis v(x) = e (a non-zero vector independent of x). This basis is obviously regular.
and periodic in \( x \). In the case of a Möbius strip, let us assume that a regular and antiperiodic basis \( \mathbf{v}(x) \) of \( F(x) \) exists. Let \( y(x) \) be the component of \( \mathbf{v}(x) \) with respect to the periodic basis \( \mathbf{e} \), namely \( \mathbf{v}(x) = y(x)\mathbf{e} \). Clearly \( y(x) \) has to be regular and antiperiodic (\( y(0) = -y(2\pi) \)). By the intermediate value theorem there needs to be a point \( x_0 \) between 0 and \( 2\pi \) such that \( y(x_0) = 0 \), as shown in Fig. 3. Thus at \( x_0 \), \( \mathbf{v}(x_0) = 0 \). But the null vector cannot be a basis for \( F(x_0) \). Therefore, the topology of the Möbius strip implies that no regular basis of \( F(x) \) can exist.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.

The topological obstruction to the existence of quasi-Bloch states in dimensions one, two and three has been recently studied [29]. It was discovered that quasi-Bloch states exist if and only if all the Chern numbers of the Bloch states in dimensions one, two and three exist.

In three dimensions, we have three Chern-like numbers \( \chi_1, \chi_2, \chi_3 \) defined in dimension [18]. The value of \( \chi_1 \) is given by

\[
\chi_1 = \frac{1}{2\pi} \int_{\partial \mathbf{A}} \mathbf{A} \cdot \mathbf{B}
\]

where \( \mathbf{A} \) is a vector potential and \( \mathbf{B} \) is the magnetic field.
ing $A(k)$. Such an approach provides Wannier functions that are localized on the short and long range.

It is convenient to determine under which condition the Wannier functions are real. In eq. (3), we take the complex conjugate and change the variables $k \rightarrow -k$

$$w^*_n(r) = \frac{1}{|\Omega|} \sum_m \int_{\Omega} \text{d}k e^{ik \cdot r} w^*_{m-k}(r)(U_{mn}(-k))^*.$$  

The reality of $V(r)$ implies that we can choose $w^*_{m-k}(r) = w_{mk}(r)$. Thus, the Wannier functions are real if $(U_{mn}(-k))^* = U_{mn}(k)$ for real $k$. This property is satisfied if $U$ satisfies $U(-k^*) = U(k)^*$ for complex $k$.

Wannier functions are often obtained by minimizing a functional $\Omega(U)$ \cite{2}. The time-reversal symmetry $J$ acts as $(JU)_{nn}(k) = (U_{mn}(-k))^*$ for real $k$. If the functional $\Omega$ satisfies the symmetry $\Omega(JU) = \Omega(U)$ and if $\Omega$ has a unique minimum (up to a possible overall phase), then $U = JU$ and the Wannier functions are real. The spread functional $\Omega$ defined by Marzari and Vanderbilt \cite{3} satisfies the symmetry $\Omega(JU) = \Omega(U)$. Thus, when the spread functional has a unique minimum $U$, the corresponding Wannier functions are real, proving for this case the conjecture of Ref. \cite{2}.

In this paper, we assumed that the system is time-reversal symmetric. If this is not the case (e.g. for the Haldane Hamiltonian \cite{33}), the present approach implies that exponentially localized Wannier functions exist in regions of the parameter space where the Chern numbers are zero or, equivalently, when the Hall current is zero. Thus, the vanishing of the Hall current is a measure of the exponential localization of the Wannier functions. This confirms rigorously Thouless’ observation \cite{34}. As a corollary, we deduce that no exponentially localized Wannier functions exist for Chern insulators (i.e. insulators with non-zero Chern numbers \cite{35}) and that time-reversal symmetric systems cannot be Chern insulators (as noticed by Haldane \cite{33} for the case $M = 1$).

In conclusion, in this work we demonstrated that Wannier functions are exponentially localized for insulators that satisfy time-reversal symmetry, and we showed that the vanishing of the Chern numbers is equivalent to the exponential localization of the Wannier functions. As a corollary, Wannier functions in Chern insulators are not exponentially localized. Moreover, we presented a simple criterion to determine when Wannier functions can be chosen as real.

Electron localization is the key of several physical concepts such as electric polarization \cite{32}, piezoelectricity, orbital magnetization \cite{33} and the nature of the insulating state \cite{31,32}. Our condition for the occurrence of localization (the vanishing of Chern numbers) is consequently a fundamental result for all these subjects.

We thank K. K. Uhlenbeck, M. Putinar, R. Zentner, D. S. Freed, F. Mauri and D. Ceresoli for useful comments, and IDRIS (project 061202) for computer time.