

Exponential localization of Wannier functions in insulators

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

¹*Institut de Minéralogie et de Physique des Milieux Condensés, CNRS UMR 7590, Universités Paris 6 et 7, IPGP, 140 rue de Lourmel, 75015 Paris, France.*

²*Zentrum Mathematik and Physik Department, Technische Universität München, 80290 München, Germany.*

³*Institut de Mathématiques de Jussieu, 175 rue du Chevaleret, 75013 Paris, France.*

⁴*Department of Materials Science and Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge MA 02139-4307, USA.*

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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.

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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 $\langle r^n \rangle$ 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, and show how localization is related to the Berry connection for the set of bands under consideration [2] and to the corresponding Chern number(s). Chern numbers have been playing a rapidly increasing role in solid-state physics, from metal-insulator transitions [13], to quantized transport [14], transition metal nanomagnets [15], quantum Hall effect [16], and its spin analogue [17]. Our central result is that Wannier functions with exponential decay can be constructed if and only if all the Chern numbers are zero. This implies that if the system is symmetric for time-reversal, then the Wannier functions are exponentially localized.

We consider a crystal with a Bravais lattice Γ and a unit cell C . The reciprocal lattice is denoted by Γ^* and the reciprocal unit cell by Ω . A function $f(\mathbf{r})$ is called

periodic if $f(\mathbf{r} + \mathbf{R}) = f(\mathbf{r})$ for any vector \mathbf{R} of Γ , while a function $f(\mathbf{k})$ is called *periodic in the reciprocal space* if $f(\mathbf{k} + \mathbf{G}) = f(\mathbf{k})$ for any vector \mathbf{G} of Γ^* . The dynamics of the electrons in the crystal is described by a 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

$$u_{n\mathbf{k}+\mathbf{G}}(\mathbf{r}) = e^{-i\mathbf{G}\cdot\mathbf{r}} u_{n\mathbf{k}}(\mathbf{r}). \quad (1)$$

Wannier functions are defined as

$$w_n(\mathbf{r} - \mathbf{R}) = \frac{1}{|\Omega|} \int_{\Omega} d\mathbf{k} e^{i\mathbf{k}\cdot(\mathbf{r}-\mathbf{R})} u_{n\mathbf{k}}(\mathbf{r}), \quad (2)$$

where $|\Omega|$ is the volume of Ω . The localization properties of the Wannier functions are related to the regularity of $u_{n\mathbf{k}}$ as a function of \mathbf{k} . In a nutshell, the more regular the states, the more localized the Wannier functions [5, 18, 19]. Exponential decay is obtained if and only if the functions are analytic [18, 20].

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 $\epsilon_1(\mathbf{k}) \leq \epsilon_2(\mathbf{k}) \leq \dots$. 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 [18] because band crossings lead to Wannier functions that can decay as $1/R^4$ [21].

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

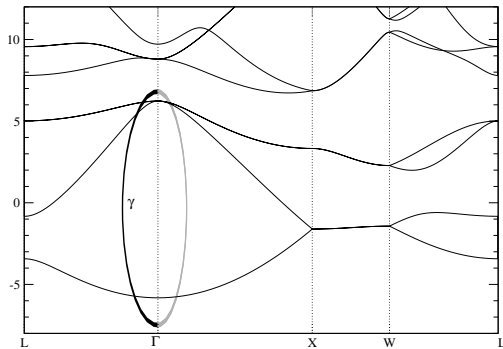


FIG. 1: Band structure of silicon. The contour γ encloses the energies of the composite band.

separated by a gap from all others. More precisely, if $\epsilon_1(\mathbf{k}), \dots, \epsilon_M(\mathbf{k})$ are the eigenstates of the composite band, we assume that there is an $a > 0$ such that $|\epsilon_n(\mathbf{k}) - \epsilon_m(\mathbf{k})| > a$ for any \mathbf{k} in Ω , $1 \leq n \leq M$ 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 $v_{n\mathbf{k}}$ (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 [18] and Nenciu [8]. In this work we determine when analytic quasi-Bloch functions exist for any composite band in two and three dimensions.

The quasi-Bloch functions $v_{n\mathbf{k}}$ can be expressed as $v_{n\mathbf{k}} = \sum_m u_{m\mathbf{k}}(\mathbf{r})U_{mn}(\mathbf{k})$, where $U(\mathbf{k})$ is a unitary matrix, defining the Wannier functions

$$w_n(\mathbf{r}) = \frac{1}{|\Omega|} \int_{\Omega} d\mathbf{k} e^{i\mathbf{k}\cdot\mathbf{r}} \sum_m u_{m\mathbf{k}}(\mathbf{r})U_{mn}(\mathbf{k}). \quad (3)$$

The improved localization of these Wannier functions was observed on many systems [2, 24].

To study the analytic properties of the Bloch functions, we consider a complex vector $\mathbf{k} = \mathbf{k}' + i\mathbf{k}''$. Then, $H(\mathbf{k})$ is not Hermitian but there still are eigenvalues $\epsilon_n(\mathbf{k})$ and eigenstates $u_{n\mathbf{k}}$ such that $H(\mathbf{k})u_{n\mathbf{k}} = \epsilon_n(\mathbf{k})u_{n\mathbf{k}}$. The branch points of $\epsilon_n(\mathbf{k})$ determine the points of non-analyticity of the function $u_{n\mathbf{k}}$ [5, 22]. Even very simple crystals, such as silicon, exhibit such branch points [25].

If the potential $V(\mathbf{r})$ is square integrable, it can be shown that $H(\mathbf{k})$ is analytic [26]. The reality of $V(\mathbf{r})$ implies that $H^\dagger(\mathbf{k}) = H(\mathbf{k}^*)$ and $H^*(\mathbf{k}) = H(-\mathbf{k}^*)$. From a practical point of view, the condition of square integrability of $V(\mathbf{r})$ encompasses potentials with Coulomb singularities and the potentials used in LDA and GGA cal-

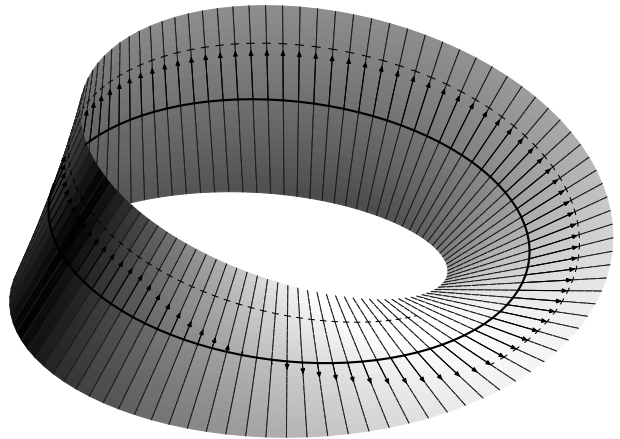


FIG. 2: Möbius strip. The solid circle is the base B and the thin straight lines are the fibres $F(x)$. The dashed line represents the constant but not antiperiodic basis \mathbf{e} . The arrows represent the continuous and antiperiodic basis vectors $\mathbf{v}(x)$. It is clear that $\mathbf{v}(x)$ has to be zero for some x .

culations. The calculation of band structures in the complex plane is available in some standard band-structure packages [27].

Several authors [8, 18, 28] 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 \mathbb{R}^3 and to each point \mathbf{r} of the base we associate a three-dimensional vector space (the fibre $F(\mathbf{r}) = \mathbb{R}^3$). The vector potential $\mathbf{A}(\mathbf{r})$ is a vector of the space $F(\mathbf{r})$. In the Born-Oppenheimer approximation, the base B is the set of possible positions of the nuclei $\mathbf{R}_1, \dots, \mathbf{R}_N$ and the fibre $F(\mathbf{R}_1, \dots, \mathbf{R}_N)$ 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 $F(\mathbf{k})$ is the M -dimensional vector space generated by the Bloch states $u_{n\mathbf{k}}$. The quasi-Bloch states $v_{n\mathbf{k}}$ can now be defined precisely as a basis of $F(\mathbf{k})$ such that each $v_{n\mathbf{k}}$ is analytic in \mathbf{k} , periodic in the reciprocal space and satisfies the boundary conditions (1).

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 $x \in [0, 2\pi]$ and the fibre is $F(x) = \mathbb{R}$. We denote by $\mathbf{v}(x)$ a basis of $F(x)$. Topology intervenes when we determine how the fiber at $x = 0$ is related to the fiber at $x = 2\pi$: we can use periodic boundary conditions $\mathbf{v}(2\pi) = \mathbf{v}(0)$ (cylinder) or antiperiodic boundary conditions $\mathbf{v}(2\pi) = -\mathbf{v}(0)$ (Möbius strip, illustrated in Fig. 2). In the case of a cylinder, we can choose a basis $\mathbf{v}(x) = \mathbf{e}$ (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 2π such that $y(x_0) = 0$, as shown in Fig. 2. Thus at x_0 , $\mathbf{v}(x_0) = \mathbf{0}$. 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 system are zero. Such result is of central importance to our paper, since it shifts the focus of our analysis on the determination of the nullity of Chern numbers in dimensions two and three.

For a given \mathbf{k} , $P(\mathbf{k}) = \sum_{n=1}^M |u_{n\mathbf{k}}\rangle\langle u_{n\mathbf{k}^*}|$ defines a projector [22]. The Riesz formula for the projector is

$$P(\mathbf{k}) = \frac{1}{2\pi i} \int_{\gamma} \frac{dz}{z - H(\mathbf{k})}.$$

where γ is a contour enclosing all the eigenvalues $\epsilon_{n\mathbf{k}}$ of the composite band (see fig.1). $P(\mathbf{k})$ is analytic in \mathbf{k} on a strip $\Omega_C = \{\mathbf{k} = \mathbf{k}' + i\mathbf{k}'', \mathbf{k}' \in \Omega, |k_j''| < A, j = 1, 2, 3\}$ with $A > 0$ even if the states $u_{n\mathbf{k}}$ are not analytic in \mathbf{k} [18]. The value of A is related to the band gap [30]. An example of contour γ for Si is given in Fig. 1.

To calculate the Chern numbers, we introduce the Berry connection corresponding to the basis $u_{n\mathbf{k}}$ of $F(\mathbf{k})$, defined in dimension d as the d -dimensional vector of matrix functions $\mathbf{A}(\mathbf{k}) = (A^1(\mathbf{k}), \dots, A^d(\mathbf{k}))$ with matrix elements

$$\mathbf{A}_{mn}(\mathbf{k}) = \int_C u_{m\mathbf{k}}^*(\mathbf{r}) \nabla_{\mathbf{k}} u_{n\mathbf{k}}(\mathbf{r}) d\mathbf{r}.$$

The trace of the curvature of this connection is [2, 29]

$$\begin{aligned} B^{ij}(\mathbf{k}) &= \text{tr} \left(\frac{\partial A^j}{\partial k_i} - \frac{\partial A^i}{\partial k_j} + [A^i, A^j] \right) \\ &= \text{Tr} \left(P(\mathbf{k}) \left[\frac{\partial P(\mathbf{k})}{\partial k_i}, \frac{\partial P(\mathbf{k})}{\partial k_j} \right] \right), \end{aligned} \quad (4)$$

where tr is the matrix trace and Tr the operator trace.

In two dimensions, this curvature leads to the unique Chern number

$$C_1 = \frac{i}{2\pi} \int_{\Omega} B^{12}(\mathbf{k}) dk_1 dk_2. \quad (5)$$

In three dimensions, we have three Chern-like numbers

$$C_{\ell} = \frac{i}{2\pi} \sum_{i < j} \int_{\Theta_{\ell}} B^{ij}(\mathbf{k}) dk_i \wedge dk_j, \quad (6)$$

where $\ell = 1, 2, 3$. The domain of integration Θ_{ℓ} is a torus defined as the set of points (k_1, k_2, k_3) of Ω such that $k_{\ell} = 0$. The reality of $V(\mathbf{r})$ implies the time-reversal symmetry $P(\mathbf{k}) = (P(-\mathbf{k}))^*$. Therefore, $B^{ij}(-\mathbf{k}) = -B^{ij}(\mathbf{k})$, the Chern numbers are zero and quasi-Bloch functions exist.

With this result in hand, we can repeat the reasoning of des Cloizeaux (section III.B of Ref. 18), to show that $\lim_{|R| \rightarrow \infty} e^{b|R|} w_n(\mathbf{r} - \mathbf{R}) = 0$ for any $b < A$.

Our proof was given for the case of “spinless” electrons. To take spin, spin-orbit and all relativistic corrections into account, we consider a crystal described by the Dirac Hamiltonian [31] $H(\mathbf{k}) = -ic\boldsymbol{\alpha} \cdot (\nabla + i\mathbf{k}) + \beta c^2/2 + V(\mathbf{r})$. The Bloch functions are Dirac spinors $u_{n\mathbf{k}}^{\alpha}(\mathbf{r})$ and the corresponding Berry connection is

$$A_{mn}^{\alpha\beta}(\mathbf{k}) = \int d\mathbf{r} (u_{m\mathbf{k}}^{\alpha}(\mathbf{r}))^* \nabla_{\mathbf{k}} u_{n\mathbf{k}}^{\beta}(\mathbf{r}).$$

The curvature is defined by equation (4), where the trace is over the n and α indices, and the Chern numbers by eqs.(5) and (6). Again, quasi-Bloch functions exist for the Dirac Hamiltonian if and only if all Chern numbers are zero [29] (in particular, if the potential V is square integrable and time-reversal symmetric). In that case, the relativistic Wannier functions are exponentially localized.

Given the existence of exponentially decaying Wannier functions $v_{n\mathbf{k}}$, we need an algorithm to determine the unitary matrix $U(\mathbf{k})$ such that $v_{n\mathbf{k}}(\mathbf{r}) = \sum_m U_{mn}(\mathbf{k}) u_{m\mathbf{k}}(\mathbf{r})$. We sketch now a possible approach. Being analytic, the quasi-Bloch functions satisfy the Cauchy-Riemann equation $\bar{\partial} v_{n\mathbf{k}} = 0$, where $\bar{\partial} = (\bar{\partial}_x, \bar{\partial}_y, \bar{\partial}_z)$ and, if $\mathbf{k} = \mathbf{k}' + i\mathbf{k}''$, $\bar{\partial}_j = (1/2)(\partial_{k_j'} + i\partial_{k_j''})$. If we denote by $\mathbf{u}(\mathbf{k})$ and $\mathbf{v}(\mathbf{k})$ the vectors with components $u_{n\mathbf{k}}$ and $v_{n\mathbf{k}}$, the Cauchy-Riemann equation gives us $\bar{\partial}\mathbf{v} = (\bar{\partial}\mathbf{u})U + \mathbf{u}(\bar{\partial}U) = 0$. Multiplying by the conjugate of \mathbf{u} , we find $\bar{\partial}U = XU$, where $X_{mn}(\mathbf{k}) = -\langle u_{m-\mathbf{k}}^* | \bar{\partial} u_{n\mathbf{k}} \rangle$. Note that, on regions where $u_{n\mathbf{k}}$ is analytic, $X_{mn}(\mathbf{k}) = 0$. The equation $\bar{\partial}U = XU$ should be solved in the space of matrices $U(\mathbf{k})$ periodic in the reciprocal space such that $U(\mathbf{k})^{\dagger} = U(\mathbf{k}^*)^{-1}$. This condition ensures the orthogonality of the quasi-Bloch functions on the real axis. Explicit integral expressions are available to solve the Cauchy-Riemann equation $\bar{\partial}f = g$ [32]. They turn $\bar{\partial}U = XU$ into an integral equation that can be solved numerically.

Note that, if U is a particular solution of $\bar{\partial}U = XU$, the general solution can be written UA , where A is any analytic matrix periodic in the reciprocal space and satisfying $A(\mathbf{k})^{\dagger} = A(\mathbf{k}^*)^{-1}$. For instance, $A(\mathbf{k})$ can be any matrix of the form $A(\mathbf{k}) = \exp(\sum_{\mathbf{R}} a_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}})$, where the sum is over a finite number of sites of Γ and the matrices $a_{\mathbf{R}}$ satisfy $a_{\mathbf{R}}^{\dagger} = -a_{-\mathbf{R}}$. This very large set of solutions corresponds to the fact that exponential decay is a long-range property: Any finite linear combination of exponentially decaying Wannier functions centered on various sites is still exponentially decaying. Therefore, it is still necessary to optimize localization around the centers of the Wannier functions [2] by properly choos-

ing $A(\mathbf{k})$. 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 $\mathbf{k} \rightarrow -\mathbf{k}$

$$w_n^*(\mathbf{r}) = \frac{1}{|\Omega|} \sum_m \int_{\Omega} d\mathbf{k} e^{i\mathbf{k}\cdot\mathbf{r}} u_{m-\mathbf{k}}^*(\mathbf{r}) (U_{mn}(-\mathbf{k}))^*.$$

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

Wannier functions are often obtained by minimizing a functional $\Omega(U)$ [2]. The time-reversal symmetry I acts as $(IU)_{mn}(\mathbf{k}) = (U_{mn}(-\mathbf{k}))^*$ for real \mathbf{k} . If the functional Ω satisfies the symmetry $\Omega(IU) = \Omega(U)$ and if Ω has a unique minimum (up to a possible overall phase), then $U = IU$ and the Wannier functions are real. The spread functional Ω defined by Marzari and Vanderbilt [2] satisfies the symmetry $\Omega(IU) = \Omega(U)$. Thus, when the spread functional has a unique minimum U , the corresponding Wannier functions are real, proving for this case the conjecture of Ref. 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 [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 [34]. As a corollary, we deduce that no exponentially localized Wannier functions exist for Chern insulators (i.e. insulators with non-zero Chern numbers [35]) and that time-reversal symmetric systems cannot be Chern insulators (as noticed by Haldane [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 [36], piezoelectricity, orbital magnetization [37] and the nature of the insulating state [1, 9]. Our condition for the occurrence of localization (the vanishing of Chern numbers) is consequently a fundamental result for all these subjects.

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