

---

# WANT User's Guide

Version 2.0.0

## AN INTEGRATED APPROACH TO AB INITIO ELECTRONIC TRANSPORT FROM MAXIMALLY-LOCALIZED WANNIER FUNCTIONS

October 2005

([www.wannier-transport.org](http://www.wannier-transport.org))

---

This *User's Guide* describes how to run and use the various features of the integrated WANT approach. This guide includes a description of the capabilities of the program, how to use these capabilities, the necessary input files and formats, and how to run the program on both uniprocessor and parallel machines.

# WANT Version 2.0.0

**CREDITS.** The development and maintenance of the WANT code is promoted by the National Research Center on nanoStructures and bioSystems at Surfaces (S3) of the Italian INFN-CNR (<http://www.s3.infn.it>) and the Physics Department North Carolina State University (NCSU) (<http://ermes.physics.ncsu.edu>) under the coordination of Arrigo Calzolari, Andrea Ferretti and Marco Buongiorno Nardelli.

The present release of the WANT package has been realized by Andrea Ferretti (S3), Arrigo Calzolari (S3) and Marco Buongiorno Nardelli (NCSU).

A list of contributors includes Carlo Cavazzoni (CINECA), Benedetta Bonferroni (S3) and Nicola Marzari (MIT).

The routines for the calculation of the maximally-localized Wannier functions were originally written by Nicola Marzari and David Vanderbilt (©1997); Ivo Souza, Nicola Marzari and David Vanderbilt (©2002); Arrigo Calzolari, Nicola Marzari, and Marco Buongiorno Nardelli (©2003).

The routines for the calculation of the quantum conductance were originally written by Marco Buongiorno Nardelli (©1998); Arrigo Calzolari, Nicola Marzari, and Marco Buongiorno Nardelli (©2003).

**GENERAL DESCRIPTION.** WANT is an open-source, GNU General Public License suite of codes that provides an integrated approach for the study of coherent electronic transport in nanostructures. The core methodology combines state-of-the-art Density Functional Theory (DFT), plane-wave, pseudopotentials calculations with a Green's functions method based on the Landauer formalism to describe quantum conductance. The essential connection between the two, and a crucial step in the calculation, is the use of the maximally-localized Wannier function representation to introduce naturally the ground-state electronic structure into the lattice Green's function approach at the basis of the evaluation of the quantum conductance. Moreover, the knowledge of the Wannier functions of the system allows for the direct link between the electronic transport properties of the device with the nature of the chemical bonds, providing insight onto the mechanisms that govern electron flow at the nanoscale.

The WANT package operates, in principles, as a simple post-processing of any standard electronic structure code. In its present version 2.0.0 the user will find a wrapper to run WANT from the results of a self-consistent calculation done using the Pwscf package ([www.pwscf.org](http://www.pwscf.org)).

WANT calculations will provide the user with: - Quantum conductance spectrum for a bulk (infinite, periodic) system and for a lead-conductor-lead geometry - Density of states spectrum in the conductor region - Centers and spreads of the maximally-localized Wannier functions of the system.

**TERMS OF USE.** Although users are not under any obligation in the spirit of the GNU General Public Licence, the developers of WANT would appreciate the acknowledgment of the effort to produce such codes in the form of the following reference:

In the text: "The results of this work have been obtained using the WANT package.[ref]"

In references: "[ref] WANT code by A. Calzolari, A. Ferretti, C. Cavazzoni, N. Marzari and M. Buongiorno Nardelli, ([www.wannier-transport.org](http://www.wannier-transport.org)). See also: A. Calzolari, N. Marzari, I. Souza and M. Buongiorno Nardelli, Phys. Rev. B 69, 035108 (2004)."

**DISCLAIMER.** While the developers of WANT make every effort to deliver a high quality scientific software, we do not guarantee that our codes are free from defects. Our software is provided "as is". Users are solely responsible for determining the appropriateness of using this package and assume all risks associated with the use of it, including but not limited to the risks of program errors, damage to or loss of data, programs or equipment, and unavailability or interruption of operations. Due to the limited human resources involved in the development of this software package, no support will be given to individual users for either installation or execution of the codes. Finally, in the spirit of every open source project, any contribution from external users is welcome, encouraged and, if appropriate, will be included in future releases.

**LICENCE.** All the material included in this distribution is free software; you can redistribute it and/or modify it under the terms of the GNU General Public License as published by the Free Software Foundation; either version 2 of the License, or (at your option) any later version. These programs are distributed in the hope that they will be useful, but WITHOUT ANY WARRANTY; without even the implied warranty of MERCHANTABILITY or FITNESS FOR A PARTICULAR PURPOSE. See the GNU General Public License for more details. You should have received a copy of the GNU General Public License along with this program; if not, write to the Free Software Foundation, Inc., 675 Mass Ave, Cambridge, MA 02139, USA.

# Contents

<b>1</b>	<b>Theoretical Background</b>	<b>5</b>
1.1	Quantum transport . . . . .	5
1.1.1	Electron transmission and Green's functions . . . . .	5
1.1.2	Transmission through a bulk system. . . . .	7
1.1.3	Transmission through a left lead-conductor-right lead (LCR) system. . . . .	8
1.2	Maximally localized Wannier functions . . . . .	9
1.2.1	Wannier functions: definitions and problems . . . . .	9
1.2.2	Maximally localized Wannier functions . . . . .	10
1.2.3	Real space hamiltonians . . . . .	13
<b>2</b>	<b>Installation procedure</b>	<b>15</b>
2.1	Step one: configuring . . . . .	15
2.2	Step two: compiling . . . . .	16
2.3	List of directories . . . . .	17
<b>3</b>	<b>How to run WANT : a step by step description</b>	<b>18</b>
3.1	Preliminary Steps: DFT-PW Calculations . . . . .	18
3.2	Calculation of maximally-localized Wannier functions . . . . .	18
3.3	Calculation of electronic transport . . . . .	19
<b>4</b>	<b>How to prepare an input file</b>	<b>21</b>
4.1	Input for DFT-PW calculations . . . . .	21
4.2	Input for Wannier function calculations . . . . .	22
4.3	Input for electronic transport calculations . . . . .	28
4.4	Input for post-processing calculations . . . . .	31
4.4.1	bands.x . . . . .	31
4.4.2	plot.x . . . . .	32

# 1 Theoretical Background

WANT is an open-source, GNU General Public License suite of codes that provides an integrated approach for the study of coherent electronic transport in low-dimensional, extended nanostructures. The core methodology combines state-of-the-art Density Functional Theory (DFT), plane-wave, pseudopotentials calculations with a Green's functions method based on the Landauer formalism to describe quantum conductance. The essential connection between the two, and a crucial step in the calculation, is the use of the maximally-localized Wannier function representation to introduce naturally the ground-state electronic structure into the lattice Green's function approach at the basis of the evaluation of the quantum conductance. Moreover, the knowledge of the Wannier functions (Wfs) of the system allows the direct link between the electronic transport properties of the device with the nature of the chemical bonds, providing insight onto the mechanisms that govern electron flow at the nanoscale.

WANT scheme was originally described in Ref. [1]:

A. Calzolari, N. Marzari, I. Souza, and M. Buongiorno Nardelli, *Phys. Rev. B* **69**, 035108 (2004).

In the following we review the theoretical background that holds the WANT method.

## 1.1 Quantum transport

Calculations of the quantum conductance are based on a recently developed efficient method for evaluating quantum transport in extended systems [2, 3, 4]. This method is applicable to any Hamiltonian that can be expanded within a localized-orbital basis and can be used as a general theoretical scheme for the computation and analysis of the electrical properties of nanostructures.

### 1.1.1 Electron transmission and Green's functions

Let us consider a system composed of a conductor,  $C$ , connected to two semi-infinite leads,  $R$  and  $L$ , as in Fig. 1. A fundamental result in the theory of electronic transport is that the conductance through a region of interacting electrons (the  $C$  region in Fig. 1) is related to the scattering properties of the region itself via the Landauer formula [5]

$$\mathcal{C} = \frac{2e^2}{h} \mathcal{T}, \quad (1)$$

where  $\mathcal{T}$  is the transmission function and  $\mathcal{C}$  is the conductance. The former represents the probability that an electron injected at one end of the conductor will transmit to the other end. In principle, we can compute the transmission function for a coherent conductor<sup>1</sup> starting from the knowledge of the scattering matrix,  $S$ . The latter is the mathematical quantity that describes the response at one lead due an excitation at another. In principle, the scattering matrix can be uniquely computed from the solution of the Schroedinger equation and would suffice to describe the transport processes we are interested in this work. However, it is a general result of conductance theory that the elements of the S-matrix can be expressed in terms of the Green's function of the

---

<sup>1</sup>A conductor is said to be coherent if it can be characterized by a transmission matrix that relates each of the outgoing wave amplitudes to the incoming wave amplitudes at a given energy

conductor [6, 7, 8] which, in practice, can be sometimes simpler to compute.

Let us consider a physical system represented by an Hamiltonian  $H$ . Its Green's function for an energy  $E$  is defined by the equation

$$(E \pm i\eta - H)G(\vec{r}, \vec{r}') = \delta(\vec{r}, \vec{r}') \quad (2)$$

where  $i\eta > 0$  is an infinitesimal imaginary part added to the energy to incorporate the boundary conditions into the equation. The solution with  $+$  sign is the retarded Green's function  $G^r$ , while the solution with  $-$  sign is called advanced Green's function  $G^a$ . The transmission function can then be expressed in terms of the Green's functions of the conductors and the coupling of the conductor to the leads in a simple manner using the Fisher and Lee formula [7]:

$$\mathcal{T} = \text{Tr}(\Gamma_L G_C^r \Gamma_R G_C^a), \quad (3)$$

where  $G_C^{\{r,a\}}$  are the retarded and advanced Green's functions of the conductor, and  $\Gamma_{\{L,R\}}$  are functions that describe the coupling of the conductor to the leads.

In the following we are going to restrict the discussion to discrete systems that we can describe by ordinary matrix algebra. More precisely, we are going to work with matrices representing a physical system in the basis of localized electronic orbitals centered on the atoms constituting the system. It includes in particular the tight-binding model.

For a discrete media, the Green's function is then the solution of a matrix equation

$$(\epsilon - H)G = I \quad (4)$$

where  $\epsilon = E \pm i\eta$  with  $\eta$  arbitrarily small and  $I$  is the identity matrix. To simplify the notations, we drop the exponent  $\{a, r\}$  referring to advanced and retarded functions when implicitly defined by  $\epsilon$ . For an open system, consisting of a conductor and two semi-infinite leads (see Fig. 1), the above Green's function can be partitioned into sub-matrices that correspond to the individual subsystems

$$\begin{pmatrix} g_L & g_{LC} & g_{LCR} \\ g_{CL} & G_C & g_{CR} \\ g_{LRC} & g_{RC} & g_R \end{pmatrix} = \begin{pmatrix} (\epsilon - h_L) & -h_{LC} & 0 \\ -h_{LC}^* & (\epsilon - H_C) & -h_{CR} \\ 0 & -h_{CR}^* & (\epsilon - h_R) \end{pmatrix}^{-1}, \quad (5)$$

where the matrix  $(\epsilon - H_C)$  represents the finite “isolated” conductor (with no coupling elements to the leads),  $(\epsilon - h_{\{R,L\}})$  represent the semi-infinite leads, and  $h_{CR}$  and  $h_{LC}$  are the coupling matrices between the conductor and the leads, and  $h^*$  denotes the familiar conjugate transpose of  $h$ . As a convention, we use lower case letters for (semi-)infinite matrices and upper case for finite dimension matrices. In Eq.(5) we have made the assumption that there is no direct interaction between the left and right leads. From this equation it is straightforward to obtain an explicit expression for  $G_C$ [6]

$$G_C = (\epsilon - H_C - \Sigma_L - \Sigma_R)^{-1} \quad (6)$$

where the finite dimension matrices

$$\Sigma_L = h_{LC}^*(\epsilon - h_L)^{-1}h_{LC}, \quad \Sigma_R = h_{RC}(\epsilon - h_R)^{-1}h_{RC}^* \quad (7)$$



Figure 1: A conductor described by the Hamiltonian  $H_C$ , connected to two semi-infinite leads  $L$  and  $R$ , through the coupling matrices  $h_{LC}$  and  $h_{CR}$ .

are defined as the self-energy terms due to the semi-infinite leads. The self-energy terms can be viewed as effective Hamiltonians that arise from the coupling of the conductor with the leads. The coupling functions  $\Gamma_{\{L,R\}}$  can then be obtained as[6]

$$\Gamma_{\{L,R\}} = i[\Sigma_{\{L,R\}}^r - \Sigma_{\{L,R\}}^a], \quad (8)$$

where the advanced self-energy  $\Sigma_{\{L,R\}}^a$  is the Hermitian conjugate of the retarded self-energy  $\Sigma_{\{L,R\}}^r$ . The core of the problem lies in the calculation of the self-energies of the semi-infinite leads.

It is well known that any solid (or surface) can be viewed as an infinite (semi-infinite in the case of surfaces) stack of principal layers with nearest-neighbor interactions [9, 10]. This corresponds to transforming the original system into a linear chain of principal layers. For a lead-conductor-lead system, the conductor can be considered as one principal layer sandwiched between two semi-infinite stacks of principal layers. The next three sections are devoted to the computation of the self-energies using the principal layers approach for different geometries.

### 1.1.2 Transmission through a bulk system.

Within the principal layer approach, the matrix elements of Eq.(4) between layer orbitals yield a series of matrix equations for the Green's functions

$$\begin{aligned} (\epsilon - H_{00})G_{00} &= I + H_{01}G_{10} \\ (\epsilon - H_{00})G_{10} &= H_{01}^*G_{00} + H_{01}G_{20} \\ &\vdots \\ (\epsilon - H_{00})G_{n0} &= H_{01}^*G_{n-1,0} + H_{01}G_{n+1,0} \end{aligned} \quad (9)$$

where the finite dimension matrices  $H_{nm}$  and  $G_{nm}$  are formed by the matrix elements of the Hamiltonian and Green's function between the layer orbitals. We assume that in a bulk system  $H_{00} = H_{11} = \dots$  and  $H_{01} = H_{12} = \dots$ . Following [11, 12], this chain can be transformed in order to express the Green's function of an individual layer in terms of the Green's function of the preceding (or following) one. This is done via the introduction of the transfer matrices  $T$  and  $\bar{T}$ , defined such that

$$G_{10} = TG_{00}$$

and

$$G_{00} = \bar{T}G_{10}.$$

Using these definitions, we can write the bulk Green's function as [13]

$$G(E) = (\epsilon - H_{00} - H_{01}T - H_{01}^*\bar{T})^{-1}. \quad (10)$$

The transfer matrix can be easily computed from the Hamiltonian matrix elements via an iterative procedure, as outlined in [11, 12]. In particular  $T$  and  $\bar{T}$  can be written as

$$\begin{aligned} T &= t_0 + \tilde{t}_0 t_1 + \tilde{t}_0 \tilde{t}_1 t_2 + \dots + \tilde{t}_0 \tilde{t}_1 \tilde{t}_2 \dots t_n \\ \bar{T} &= \tilde{t}_0 + t_0 \tilde{t}_1 + t_0 t_1 \tilde{t}_2 + \dots + t_0 t_1 t_2 \dots \tilde{t}_n \end{aligned}$$

where  $t_i$  and  $\tilde{t}_i$  are defined via the recursion formulas

$$\begin{aligned} t_i &= (I - t_{i-1} \tilde{t}_{i-1} - \tilde{t}_{i-1} t_{i-1})^{-1} t_{i-1}^2, \\ \tilde{t}_i &= (I - t_{i-1} \tilde{t}_{i-1} - \tilde{t}_{i-1} t_{i-1})^{-1} \tilde{t}_{i-1}^2 \end{aligned}$$

and

$$\begin{aligned} t_0 &= (\epsilon - H_{00})^{-1} H_{01}^*, \\ \tilde{t}_0 &= (\epsilon - H_{00})^{-1} H_{01}. \end{aligned}$$

The process is repeated until  $t_n, \tilde{t}_n \leq \delta$  with  $\delta$  arbitrarily small. Usually no more than 5 or 6 terms are required to converge the above sum.

If we compare Eq.(10) with Eq. (6), in the hypothesis of leads and conductors being of the same material (bulk conductivity), we can identify one principal layer of the bulk system with the conductor  $C$ , so that  $H_{00} \equiv H_C$ . In particular, by comparing with Eq.(6), we obtain the expression of the self-energies of the conductor-leads system

$$\Sigma_L = H_{01}^* \bar{T}, \quad \Sigma_R = H_{01} T. \quad (11)$$

The coupling functions are then obtained from the sole knowledge of the transfer matrices and the coupling Hamiltonian matrix elements:  $\Gamma_L = -\text{Im}(H_{01}^* \bar{T})$  and  $\Gamma_R = -\text{Im}(H_{01} T)$  [2].

*In the application of the Landauer formula, it is customary to compute the transmission probability from one lead to the other assuming that the leads are connected to a reflectionless contact whose electron energy distribution is known (see for instance Datta p. 59).*

### 1.1.3 Transmission through a left lead-conductor-right lead (LCR) system.

The procedure outlined above can also be applied in the case of electron transmission through one or more interfaces, between different media. For the calculation of conductances in realistic experimental geometry, the method can be expanded to the general configuration of a Left-lead-Conductor-Right-lead (LCR) systems — as displayed in Fig.1. To study this case we make use of the Surface Green's Function Matching (SGFM) theory, pioneered by [14, 13].

We have to compute the Green's function  $G_I$ , where the subscript  $I$  refers to the interface region composed of two principal layers — one in each media — (L, C, R in our case). Using the SGFM method,  $G_I$  is calculated from the bulk Green's function of the isolated systems, and the coupling between the two principal layers at the two sides of the interface. Via the calculation of the transmitted and reflected amplitudes of an elementary excitation that propagates from one medium to another, it can be shown that the interface Green's function obeys the following secular equation[13]:



$$\begin{aligned}
G_{LCR} &= \begin{pmatrix} G_L & G_{LC} & G_{LR} \\ G_{CL} & G_C & G_{CR} \\ G_{RL} & G_{RC} & G_R \end{pmatrix} \\
&= \begin{pmatrix} \epsilon - H_{00}^L - (H_{01}^L)^* \bar{T} & -H_{LC} & 0 \\ -H_{CL} & \epsilon - H_C & -H_{CR} \\ 0 & -H_{RC} & \epsilon - H_{00}^R - H_{01}^R T \end{pmatrix}^{-1}.
\end{aligned} \tag{12}$$

where  $H_{nm}^{\{L,R\}}$  are the block matrices of the Hamiltonian between the layer orbitals in the left and right leads respectively, and  $T_{\{L,R\}}$  and  $\bar{T}_{\{L,R\}}$  are the appropriate transfer matrices. The latter are easily computed from the Hamiltonian matrix elements via the iterative procedure already described in the bulk case (Sec.1.1.2). Correspondingly,  $H_{LC}$  and  $H_{CR}$  are the coupling matrices between the conductor and the leads principal layers in contact with the conductor.

It is straightforward to obtain in the form of Eq.(6),  $G_C = (\epsilon - H_C - \Sigma_L - \Sigma_R)^{-1}$ , where  $\Sigma_L$  and  $\Sigma_R$  are the self-energy terms due to the semi-infinite leads, and identify[2]

$$\begin{aligned}
\Sigma_L &= H_{LC}^* (\epsilon - H_{00}^L - (H_{01}^L)^* \bar{T}_L)^{-1} H_{LC}, \\
\Sigma_R &= H_{CR} (\epsilon - H_{00}^R - H_{01}^R T_R)^{-1} H_{CR}^*.
\end{aligned} \tag{13}$$

The transmission function in the LCR geometry can then be derived from Eq.(3) and (8).

The knowledge of the conductor's Green's function  $G_C$  gives also direct information on the electronic spectrum of the system via the spectral density of electronic states

$$N(E) = -(1/\pi) \text{Im}[\text{Tr}(G_C(E))].$$

We have assumed a truly one-dimensional chain of principal layers, which is physical only for systems like nanotubes or quantum wires that have a definite quasi-one-dimensional character. The extension to a truly three-dimensional case is straightforward using Bloch functions in the directions perpendicular to the conduction. The introduction of the principal layer concept implies that along the direction of the layer expansion the system is described by an infinite set of  $k_\perp$  while  $k_\parallel$  is still a good quantum number for the problem. The above procedure effectively reduces the three-dimensional system to a set of non-interacting linear-chains, one for each  $k_\parallel$  [9, 10]. We can then use the usual  $k$ -point summation techniques to evaluate, for instance, the quantum conductance

$$T(E) = \sum_{k_\parallel} w_{k_\parallel} T_{k_\parallel}(E)$$

where  $w_{k_\parallel}$  are the relative weights of the different  $k_\parallel$  in the irreducible wedge of the surface Brillouin zone [15].

## 1.2 Maximally localized Wannier functions

### 1.2.1 Wannier functions: definitions and problems

Bloch orbitals cannot be used directly to evaluate electronic transport with the method outlined in Sec. 1.1. As we have pointed out, the quantum conductance is computed starting from the

knowledge of the lattice Green's function, whose calculation relies on a localized orbital representation of the electronic states in real space. **Bloch orbitals, that are intrinsically delocalized, have to be transformed into localized functions in order to construct the sparse, short-ranged matrix elements of the Hamiltonian.** The core of our proposed methodology is to use *maximally-localized Wannier functions* (Wfs) for the system considered. These are the most natural choice for a set of localized orbitals that still span the same Hilbert space of the Hamiltonian eigenfunctions, and they allow to bridge plane-wave electronic structure and lattice Green's function calculations in a coherent fashion.

A Wannier function  $w_{n\mathbf{R}}(\mathbf{r})$ , labeled by the Bravais lattice vector  $\mathbf{R}$ , is usually defined via a unitary transformation of the Bloch functions  $\psi_{n\mathbf{k}}(\mathbf{r})$  of the  $n$ th band:

$$w_{n\mathbf{R}}(\mathbf{r}) = \frac{V}{(2\pi)^3} \int_{BZ} \psi_{n\mathbf{k}}(\mathbf{r}) e^{-i\mathbf{k}\cdot\mathbf{R}} d^3k, \quad (14)$$

where  $V$  is the volume of the unit cell and the integration is performed over the entire Brillouin Zone. It is easy to show that the WFs defined as above form an orthonormal basis set, and that any two of them, for a given index  $n$  and different  $\mathbf{R}$  and  $\mathbf{R}'$ , are just translational images of each other. Note that, as the  $\mathcal{N}$  WFs form a (continuous) linear combinations of Bloch functions with different energies, they do not represent stationary states, but still span exactly the same original Hilbert space.

The *ab-initio* eigenstates are well-defined, modulus an arbitrary  $\mathbf{k}$ -dependent phase factor; thus, the definition above does not lead to a unique set of Wannier functions [16, 17], since the electronic structure problem is invariant for the transformation  $\psi_{n\mathbf{k}} \rightarrow e^{i\phi_n(\mathbf{k})} \psi_{n\mathbf{k}}$ . Besides this freedom in the choice of phases  $\phi_n(\mathbf{k})$  for the Bloch functions, there is a more comprehensive gauge freedom stemming from the fact that the many-body wavefunction is actually a Slater determinant: a unitary transformation between orbitals will not change the manifold, and will not change the total energy and the charge density of the system. In all generality, starting with a set of  $\mathcal{N}$  Bloch functions with periodic parts  $u_{n\mathbf{k}}$ , we can construct infinite sets of  $\mathcal{N}$  WFs displaying different spatial characteristics:

$$w_{n\mathbf{R}}(\mathbf{r}) = \frac{V}{(2\pi)^3} \int_{BZ} \left[ \sum_m U_{mn}^{(\mathbf{k})} \psi_{m\mathbf{k}}(\mathbf{r}) \right] e^{-i\mathbf{k}\cdot\mathbf{R}} d^3k. \quad (15)$$

The unitary matrices  $U^{(\mathbf{k})}$  include also the gauge freedom on phase factors afore mentioned. [18]

The present WANT method is based on a localization algorithm that allows to transform a set of BFs - calculated by means of *ab initio* approaches - into a unique set of Maximally localized Wannier functions (MLWFs), as proposed by Marzari and Vanderbilt in 1997 [18]. The formulation of this minimum-spread criterion extends the concept of *localized molecular orbitals*, proposed by Boys [19] for molecules, to the solid-state case. However, its generality allows to deal with both "extended" (periodic and disordered) systems as well as with "isolated" clusters and molecules, in the limit of large supercells.

### 1.2.2 Maximally localized Wannier functions

For our purposes, we need to transform the Bloch eigenstates in WFs with the narrowest spatial distribution. Following the procedure proposed by Marzari and Vanderbilt [18] **we search the par-**

**ticular unitary matrix  $U_{mn}^{(\mathbf{k})}$  that transform the Bloch eigenstates in the WFs with the narrowest spatial distribution.**

A measure of the spatial delocalization of WFs is given by a *Spread Operator* ( $\Omega$ ), defined as the sum of the second moments of the Wannier functions in the reference cell ( $\mathbf{R}=\mathbf{0}$ ):

$$\Omega = \sum_n [\langle r^2 \rangle_n - \langle \mathbf{r} \rangle_n^2], \quad (16)$$

where the sum is over a selected group of bands, and

$$\begin{aligned} \langle \mathbf{r} \rangle_n &= \langle \mathbf{0}n | \mathbf{r} | \mathbf{0}n \rangle \\ \langle r^2 \rangle_n &= \langle \mathbf{0}n | r^2 | \mathbf{0}n \rangle \end{aligned} \quad (17)$$

The value of the spread  $\Omega$  depends on the choice of unitary matrices  $U^{(\mathbf{k})}$ ; thus it is possible to evolve any arbitrary set of  $U^{(\mathbf{k})}$  until the minimum condition

$$\frac{\delta \Omega_{\mathbf{k}}}{\delta U^{(\mathbf{k})}} = 0 \quad (18)$$

is satisfied. At the minimum, we obtain the matrices  $(U^{(\mathbf{k})})^{ML}$  that transform the first-principles  $\psi_{n\mathbf{k}}^{FP}(\mathbf{r})$  into the *maximally-localized WFs*  $w_{n\mathbf{R}}^{ML}(\mathbf{r})$ :

$$\begin{aligned} \psi_{n\mathbf{k}}^{ML}(\mathbf{r}) &= \sum_m (U_{mn}^{(\mathbf{k})})^{ML} \psi_{m\mathbf{k}}^{FP}(\mathbf{r}), \\ w_{n\mathbf{R}}^{ML}(\mathbf{r}) &= \frac{1}{N} \sum_{\mathbf{k}} e^{-i\mathbf{k} \cdot \mathbf{R}} \psi_{n\mathbf{k}}^{ML}(\mathbf{r}). \end{aligned} \quad (19)$$

If we restrict to the case of  $\mathbf{k}$ -point mesh calculations, we can use finite differences in reciprocal space to evaluate the derivatives of Eq. 18. For this purpose we rewrite the expectation values of operators  $\mathbf{r}$  and  $r^2$ , as proposed by Blount [20]:

$$\begin{aligned} \langle \mathbf{0}n | \mathbf{r} | \mathbf{0}n \rangle &= i \frac{1}{N} \sum_{\mathbf{k}} e^{+i\mathbf{k} \cdot \mathbf{R}} \langle u_{\mathbf{k}n} | \nabla_{\mathbf{k}} | u_{\mathbf{k}n} \rangle \\ \langle \mathbf{0}n | r^2 | \mathbf{0}n \rangle &= \frac{1}{N} \sum_{\mathbf{k}} e^{+i\mathbf{k} \cdot \mathbf{R}} \langle u_{\mathbf{k}n} | \nabla_{\mathbf{k}}^2 | u_{\mathbf{k}n} \rangle, \end{aligned} \quad (20)$$

where  $|u_{\mathbf{k}n}\rangle = e^{-i\mathbf{k} \cdot \mathbf{r}} |\psi_{\mathbf{k}n}\rangle$  is the period part of the Bloch function. Making the assumption that the BZ has been discretized into a uniform  $\mathbf{k}$ -point mesh, and letting  $\mathbf{b}$  the vectors that connect a mesh point to its near neighbors, we can define the overlap matrix between Bloch orbitals as

$$M_{mn}^{(\mathbf{k}, \mathbf{b})} = \langle u_{\mathbf{k}m} | u_{\mathbf{k}+\mathbf{b}, n} \rangle. \quad (21)$$

Using the expression of the gradient in terms of finite differences and substituting  $M_{mn}^{(\mathbf{k}, \mathbf{b})}$  in Eq. 20 we obtain the expressions for  $\mathbf{r}$  and  $r^2$ , to be used in the localization procedure:

$$\begin{aligned} \langle \mathbf{r} \rangle_n &= -\frac{1}{N} \sum_{\mathbf{k}, \mathbf{b}} w_b \mathbf{b} \text{Im} \left[ \ln M_{nn}^{(\mathbf{k}, \mathbf{b})} \right] \\ \langle r^2 \rangle_n &= \frac{1}{N} \sum_{\mathbf{k}, \mathbf{b}} w_b \left\{ \left[ 1 - |M_{nn}^{(\mathbf{k}, \mathbf{b})}|^2 \right] + \left[ \ln M_{nn}^{(\mathbf{k}, \mathbf{b})} \right]^2 \right\}. \end{aligned} \quad (22)$$

Here,  $w_b$  are the weight of the vectors  $\mathbf{b}$ , and must satisfy the completeness condition  $\sum_{\mathbf{b}} w_b b_\alpha b_\beta = \delta_{\alpha\beta}$ . Substituting the above expression into Eq. 16, we obtain the expression for the spread operator as a function of the overlap matrix  $M_{mn}^{(\mathbf{k},\mathbf{b})}$ .

In order to calculate the gradient of Eq. 18, we consider the first order change in  $\Omega$  arising from an infinitesimal transformation  $U_{mn}^{(\mathbf{k})} = \delta_{mn} + dW_{mn}^{(\mathbf{k})}$ , where  $dW$  is an infinitesimal antiunitary matrix ( $dW^\dagger = -dW$ ). The gauge transformation rotates the wave functions according to Eq. 15 into  $|u_{\mathbf{k}n}\rangle \rightarrow |u_{\mathbf{k}n}\rangle + \sum_m dW_{mn}^{(\mathbf{k})} |u_{\mathbf{k}m}\rangle$ . Following the elegant description of Ref. [18], we obtain the final expression for the gradient of the spread functional

$$G^{(\mathbf{k})} = \frac{\delta\Omega}{dW^{(\mathbf{k})}} = 4 \sum_{\mathbf{b}} w_b \left\{ \frac{R^{(\mathbf{k},\mathbf{b})} - [R^{(\mathbf{k},\mathbf{b})}]^\dagger}{2} - \frac{T^{(\mathbf{k},\mathbf{b})} + [T^{(\mathbf{k},\mathbf{b})}]^\dagger}{2i} \right\} \quad (23)$$

where

$$R_{mn}^{(\mathbf{k},\mathbf{b})} = M_{mn}^{(\mathbf{k},\mathbf{b})} M_{nn}^{(\mathbf{k},\mathbf{b})*}, \quad T_{mn}^{(\mathbf{k},\mathbf{b})} = \frac{M_{mn}^{(\mathbf{k},\mathbf{b})}}{M_{nn}^{(\mathbf{k},\mathbf{b})}} \left[ \text{Im}(\ln M_{nn}^{(\mathbf{k},\mathbf{b})}) + \mathbf{b} \cdot \langle \mathbf{r} \rangle_n \right]. \quad (24)$$

Note that the entire expression  $G^{(\mathbf{k})}$  is a function of the overlap matrices  $M^{(\mathbf{k},\mathbf{b})}$ . The minimization of the spread functional  $\Omega$  is obtained via a steepest descent scheme, in the reciprocal space, while the transformation to real space is a post-processing step. This procedure, computationally inexpensive, requires the updating only of the unitary matrices, which appear in the formulation above, and not of the wave functions.

The Wannier functions obtained with the above procedure are real, except for an overall phase factor. It is important to notice that whenever a Born-von Karman discretization of the Brillouin Zone is introduced, even the above-mentioned WFs are not truly localized, but will be periodic in real-space, with a *superperiodicity* determined by the BZ discretization. The truly isolated limit is recovered only in the case of continuous BZ integrations. This is easily seen remembering that  $\psi_{n\mathbf{k}}(\mathbf{r}) = u_{n\mathbf{k}}(\mathbf{r}) e^{i\mathbf{k} \cdot \mathbf{r}}$ , and  $u_{n\mathbf{k}}(\mathbf{r})$  has the periodicity of the direct lattice; thus the phase factors  $e^{i\mathbf{k} \cdot \mathbf{r}}$  determine the *superperiodicity* of the  $\psi_{n\mathbf{k}}$  themselves.

In the standard language of electronic-structure calculations, if the  $\psi_{n\mathbf{k}}$  have  $\mathbf{k}$ 's that are restricted to a uniform Monkhorst-Pack mesh, they will all be periodic with a wavelength inversely proportional to the spacing of the mesh; this periodicity is consequently inherited by the WFs. For  $\mathcal{N}$   $\mathbf{k}$ -points along a direction of the BZ, the WFs will repeat along the corresponding direction every  $\mathcal{N}$  cells. A dense mesh of  $\mathbf{k}$ -points guarantees that the adjacent replicas of a WF are sufficiently distant and do not interact. However, even the case of  $\Gamma$ -sampling is encompassed by the above formulation. In this case the neighboring  $\mathbf{k}$ -points for  $\Gamma$  are given by the homologous  $\Gamma$ -points of the neighboring cells. In this case the algebra becomes simpler and an equivalent real-space formulation is preferred [21, 22].

The method described above works properly in the case of *isolated groups* of bands. A Bloch band is called *isolated* if it does not become degenerate with any other band anywhere in the BZ. Conversely, a group of bands is said to form a *composite group* if bands are inter-connected by degeneracy, but are *isolated* from all the other bands [18]. On the other hand to study quantum conductance in extended systems we often need to compute WFs for a subset of energy bands that are entangled or mixed with other bands. Most often we are interested in the states that lie in the

vicinity of the Fermi level of a conductor in a restricted energy range. Since the unitary transformations  $U^{(\mathbf{k})}$  mix energy bands at each  $\mathbf{k}$ -point, any arbitrary choice of states inside a prescribed window will affect the localization properties of WFs unless energy gaps effectively separate the manifold of interest from higher and lower bands.

This problem has been solved by Souza, Marzari, and Vanderbilt, introducing an additional disentanglement procedure [23] that automatically extracts the best possible manifold of a given dimension from the states falling in a predefined energy window. This is the generalization to *entangled* or metallic cases of the maximally-localized WF formulation. The procedure relies on minimizing the subspace dispersion across the Brillouin Zone, and effectively extracts the bands of interest from the overall band structure.

In practice, first we select a desired number of bands in an energy window; then we determine the optimally-connected subspace that can be extracted from that band structure; and finally we proceed with a standard localization procedure inside the selected subspace, using the same kind of spread functional  $\Omega$  and of unitary matrices  $U_{mn}^{(\mathbf{k})}$ . The resulting orbitals have the same good localization properties, and allow to apply our formalism to arbitrary systems, independently of the insulating or metallic nature of the band manifold. It should be stressed that the WFs obtained in the later case are not the WFs of the occupied subspace (that would exhibit poor localization properties), but are those of a well connected, continuous subspace that in general will contain both occupied and unoccupied Bloch functions.

### 1.2.3 Real space hamiltonians

In order to calculate the conductance according to the prescriptions outlined in Sec. 1.1, we need as an input the matrix elements of the Hamiltonian calculated on a localized basis: in our case, it is the minimal basis of the maximally-localized WFs. The advantages of this choice are twofold: firstly, besides being a minimal basis, the WFs span *exactly* the Hilbert space of an insulator and, with arbitrary accuracy, of an entangled metallic system. Secondly, their localization assures the choice of the system with the smallest number of atomic layers. The Hamiltonian matrices ( $H_{mn}^{LR}$ ,  $H_C$ ,  $H_{LC}$ ,  $H_{CR}$ ) can be formally obtained from the *on site* ( $H_{00}$ ) and *coupling* ( $H_{01}$ ) matrices between *principal layers*. In our formalism, and assuming a BZ sampling fine enough to eliminate the interaction with the periodic images, we can simply compute these matrices from the unitary matrix  $U^{(\mathbf{k})}$  obtained in the localization procedure.

By definition of energy eigenvalues ( $\tilde{\epsilon}_{m\mathbf{k}}$ ), the Hamiltonian matrix  $\tilde{H}_{mn}(\mathbf{k}) = \tilde{\epsilon}_{m\mathbf{k}}\delta_{m,n}$ , is diagonal in the basis of the Bloch eigenstates. We can calculate the Hamiltonian matrix in the rotated basis,

$$H^{(rot)}(\mathbf{k}) = (U^{(\mathbf{k})})^\dagger \tilde{H}(\mathbf{k}) U^{(\mathbf{k})}. \quad (25)$$

Next we Fourier transform  $H^{(rot)}(\mathbf{k})$  into a set of  $N_{kp}$  Bravais lattice vectors  $\mathbf{R}$  within a Wigner-Seitz supercell centered around  $\mathbf{R}=0$  :

$$H_{mn}^{(rot)}(\mathbf{R}) = \frac{1}{N_{kp}} \sum_{\mathbf{k}} e^{-i\mathbf{k}\cdot\mathbf{R}} H_{mn}^{(rot)}(\mathbf{k}) = \langle w_{m0} | \hat{H} | w_{n\mathbf{R}} \rangle, \quad (26)$$

where  $N_{kp}$  derives from the folding of the uniform mesh of  $\mathbf{k}$ -points in the BZ. The term with  $\mathbf{R}=0$  provides the *on site* matrix  $H_{00} = \langle w_{m0} | \hat{H} | w_{n0} \rangle$ , and the term  $\mathbf{R}=1$  provides the *coupling* matrix

$H_{01} = \langle w_{m\mathbf{0}} | \hat{H} | w_{n\mathbf{1}} \rangle$ : These are the only ingredients required for the evaluation of the quantum conductance.

## 2 Installation procedure

NOTES: (i) The present version of the code adopts the installation procedure of PWSFC (for more details see also [www.pwscf.org](http://www.pwscf.org)). (ii) This installation procedure is still experimental, and only a limited number of architectures are currently supported. Installation procedure is also reported in the **docs/README.install** file.

Installation is a two-step procedure:

1. “cd” to the top directory of the WANT tree (that should be the one where this file is), and issue this command at the shell prompt:  
./configure
2. Now run:  
make target

where “target” is one (or more) of the following: disentangle, wannier, conductor, bands, blc2wan, plot. Running “make” without arguments prints a list of available targets.

Cross-compilation is not currently supported.

### 2.1 Step one: configuring

“configure” is a GNU-style configuration script, automatically generated by GNU Autoconf. (If you want to play with it, its source file is “conf/configure.ac”; you may also want to edit “conf/make.sys.in”) It generates the following files:

\$TOPDIR/make.sys	compilation settings and flags
\$TOPDIR/*/makedeps.sh	dependencies, per source directory

where \$TOPDIR is the top directory of the WANT source tree.

“dependencies” files are actually generated by the “makedeps.sh” shell script. If you modify the program sources, you might have to rerun it. Note that you must run it from the directory it is in.

To force using a particular compiler, or compilation flags, or libraries, you may set the appropriate environment variables when running the configuration script. For example:

```
./configure CC=gcc CFLAGS=-O3 LIBS="-llapack -lblas -lfftw"
```

Some of those environment variables are:

TOPDIR	: top directory of the WANT tree (defaults to ‘pwd’)
F90, F77, CC	: Fortran 90, Fortran 77, and C compilers
CPP	: source file preprocessor (defaults to “\$CC -E”)
LD	: linker (defaults to \$F90)
CFLAGS, FFLAGS, F90FLAGS, CPPFLAGS, LDFLAGS	: compilation flags
LIBDIRS	: extra directories to search for libraries (see below)

You should always be able to compile the WANT suite of programs without having to edit any of the generated files. If you ever have to, that should be considered a bug in the configuration script and you are encouraged to submit a bug report.

**IMPORTANT:** WANT can take advantage of several optimized numerical libraries:

- ESSL on AIX systems (shipped by IBM)
- MKL together with Intel compilers (shipped by Intel, free for non-commercial use)
- ATLAS (freely downloadable from <http://math-atlas.sourceforge.net/>)
- FFTW (freely downloadable from <http://www.fftw.org/>)

The configuration script attempts to find those libraries, but may fail if they have been installed in non-standard locations. You should look at the LIBS environment variable (either in the output of the configuration script, or in the generated “make.sys”) to check whether all available libraries were found.

If any libraries weren’t found, you can rerun the configuration script and pass it a list of directories to search, by setting the environment variable LIBDIRS; directories in the list must be separated by spaces. For example:

```
./configure LIBDIRS="/opt/intel/mkl/mkl61/lib/32 /usr/local/lib/fftw-2.1.5"
```

If this still fails, you may set the environment variable LIBS manually and retry. For example:

```
./configure LIBS="-L/cineca/prod/intel/lib -lfftw -llapack -lblas"
```

Beware that in this case, you must specify *\*all\** the libraries that you want to link to. The configuration script will blindly accept the specified value, and will *\*not\** search for any extra libraries.

If you want to use the FFTW library, the “fftw.h” include file is also required. If the configuration script wasn’t able to find it, you can specify the correct directory in the INCLUDEFFTW environment variable. For example:

```
./configure INCLUDEFFTW="/cineca/lib/fftw-2.1.3/fftw"
```

## 2.2 Step two: compiling

Here is a list of available compilation targets:

make all	compile	wannier/disentangle.x	(step 1)
		wannier/wannier.x	(step 2)
		wannier/bands.x	(post proc)
		wannier/plot.x	(post proc)
		wannier/blc2wan.x	(post proc)
		transport/conductor.x	(step 3)
make clean	remove	Object files and executables	
make veryclean	remove	Configuration files too	



IMPORTANT: If you change any compilation or precompilation options after a previous (successful or failed) compilation, you must run “make clean” before recompiling, unless you know exactly which routines are affected by the changed options and how to force their recompilation.

## 2.3 List of directories

Within the top directory of the WANT tree there are the following directories:

<b>conf</b>	includes the configuration and compilation files
<b>docs</b>	includes the documentation files and manuals
<b>iotk</b>	includes the files for the iotk input/output interface
<b>transport</b>	includes the source files and modules of for program conductor.x
<b>wannier</b>	includes the source files and modules of for programs disentangle.x, wannier.x, bands.x, plot.x, blc2wan.x
<b>bin</b>	includes links of all the executable file *.x
<b>include</b>	includes the environmental file *.h
<b>libs</b>	includes source file for internal libraries
<b>tests</b>	includes tutorial examples for the use of WANT suite of codes
<b>utility</b>	includes some useful tools for WANT use.

### 3 How to run WANT : a step by step description

NOTE: At present the WANT code is implemented to work as post processing of the self-consistent calculations done using the Pwscf package ([www.pwscf.org](http://www.pwscf.org)); thus we will refer to that code in the following description.

Following the theoretical description of Section 1, the evaluation of transport properties requires a three separate steps:

1. Calculation of DFT-PW electronic structure:
2. Calculation of maximally-localized Wannier functions
3. Calculation of the quantum conductance

IMPORTANT: For a correct results, the following steps **MUST** be done in this order.

#### 3.1 Preliminary Steps: DFT-PW Calculations

- i) Self-consistent calculation with the Pwscf code.  
For the description of the input and for further details see the Pwscf manual at [www.pwscf.org](http://www.pwscf.org).
- ii) Bandstructure calculation.  
Starting from the self-consistent charge calculated in point (1), we calculate the Bloch functions for a **UNIFORM k-point grid in the COMPLETE Brillouin Zone**; Gamma point must be included. Time-reversal reduction of k-points is not allowed. In order to obtain the correct k-point grid, include flag "nosym = .true." in the namelist "&SYSTEM" and the complete list of k-points in the "K\_POINTS" card.
- iii) From Pwsc to Wannier code.  
Using the post processing **pw\_export.x** (distributed in the Pwscf package), the input data, necessary for the following Wannier calculations, will be extracted from PWscf outputs and stored in the new-created directory *prefix.export*.

NOTE: Steps (i-iii) may be run, using the parallel version of the code, paying attention to use the same number of processors. From this point to the end, instead, the code is scalar.

#### 3.2 Calculation of maximally-localized Wannier functions

Following the list of input parameter of Section 4 (also reported in the file docs/README.input) and the examples in directory Tests, create the input file, that will be used for the following 2 steps (a-b).

- a) Starting from the data stored at level (iii), **disentangle.x** selects the working energy window, from which it will extract a selected number ( $N$ ) of WFs. For each k-point, the energy window MUST contain at least a number of band equal to the WFs to be calculated  $N$ . It is possible to use an inner window (inside the working one) to treat frozen-states, for details see Phys. Rev. B 65 035109 (2002). Disentangle.x extracts a manifold of  $N$  bands along the Brillouin Zone, from an entangled group in the selected energy window. Trial wannier centers are not mandatory in this step (depending on itrial flag). Disentangle.x produces a standard output with the main results, and two internal files: *prefix.ovp* and *prefix.space*

- b) Running **wannier.x** (using the same input as before), we perform the localization procedure and the optimal unitary matrix  $U(\mathbf{k})$ , that minimizes the spread operator, is obtained. In this step the definition of the trial centers is necessary, even in the case they have not been used in step (a). Wannier.x produces a standard output with the main results, and a set of files (*prefix.RHAM.XXX*) containing the complex hamiltonian matrices, written by columns. The first two integers in the first line give the dimension of the matrix, the last three integers describe the lattice vector, the WFs are belonging to (e.g. 0 0 0 is the reference cell, 1 0 0 is the first neighbour cell along the first axis direction, etc). Wannier.x produces also two internal binary files: *prefix.ham*, *prefix.wan*. The former file will be (one of ) the input for the transport calculation.

NOTES: Steps (a-b) are the most expensive ones of the overall procedure, both in term of memory and in term of CPU time. Calculations for big systems may take over several hours! On the contrary other steps of the chain are quite inexpensive.

Further physical information may be obtained as external post-processing of the Wfs calculation. They are not necessary for the transport calculation, but they may be very useful for a better understanding of the intrinsic electronic properties of the system. These codes require a separate input, to be generated following Section 4 or the file docs/README.input.

- The code **bands.x** calculates the interpolated bandstructure for the system along selected direction in the Brillouin Zone. The comparison with the original bandstructure calculated using the Pwscf code constitutes a good test to check the accuracy of the results. For details see also Phys. Rev. B 65 035109 (2002). The code produces three files *prefix.dftband.dat*, *prefix.intband.dat*, *prefix.wanband.dat* that may be used for a direct visualization.
- The code **plot.x** calculates Wfs (among the calculated ones) in the unitary cell. Wfs are expressed in a numerical form on the real space FFT mesh, and formatted into a txt file. You can use free visualization software (e.g. gOpenMol) to plot Wfs.
- The code **blc2wan.x** transforms a generic operator  $\hat{A}$  expressed on the Bloch eigenstates (BF) into the Wannier function basis set. Using the unitary matrices  $U(\mathbf{k})$  calculated at step (b), we first calculate the corresponding matrix in the rotated basis,  $A^{(rot)}(\mathbf{k}) = (U(\mathbf{k}))^\dagger \tilde{A}(\mathbf{k}) U(\mathbf{k})$  then we Fourier transform  $A^{(rot)}(\mathbf{k})$  into a set of  $N_{kp}$  Bravais lattice vectors  $\mathbf{R}$  within a Wigner-Seitz supercell centered around  $\mathbf{R}=0$ .

### 3.3 Calculation of electronic transport

Using the hamiltonian matrices calculated in step (b) we can calculate both the "bulk" and the "two terminal" transmittance. Input file may be generated following Section 4 or the file docs/README.input. The result (*cond.dat*) is expressed in unit of the quantum of conductance ( $2e^2/h$ ). Additional output file *dos.dat* contains the density of states ( $N(E)$ ) of the overall open system, deriving from the trace of the conductor Green's function  $G_C$  ( $N(E) = -(1/\pi)Im[TrG_C(E)]$ ).

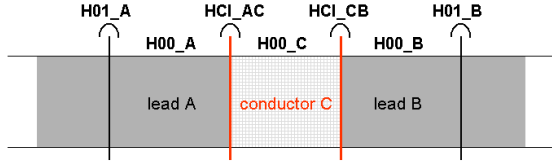
**conductor.x** calculates both the **bulk** and the **two-terminal** transmittance for the systems.

- In the case of **bulk** transmittance, the conductor and the leads are constituted of the same system. Only two hamiltonian matrices are needed: that belonging to the reference cell ( 0 0 0 three last number in the first line, see step (b) ) and that belonging to the first cell in

the direction along which we calculate the transport ( e.g. 1 0 0 in the first line, in order to calculate the transport along x-direction).

- In the case of **two terminal** transmittance we need, in principle, three sets of calculations (two only if the leads are of the same material): bulk calculations for the two infinite leads and a supercell calculation for the conductor and the contacts (see PRB 69, 035108 (2004)). The labels for the hamiltonian matrices used in the input of conductor.x refer to the scheme below.

**Matrix definition** Given a conductor (C) bonded to a right lead (A) and a left lead (B)



$H00\_A$  =  $N \times N$  on site hamiltonian of the leads A (H00 from bulk calculation)  
 $H01\_A$  =  $N \times N$  hopping hamiltonian of the leads A (H01 from bulk calculation)  
 $H00\_B$  =  $P \times P$  on site hamiltonian of the leads B (H00 from bulk calculation)  
 $H01\_B$  =  $P \times P$  hopping hamiltonian of the leads B (H01 from bulk calculation)  
 $H00\_C$  =  $M \times M$  on site hamiltonian of the conductor C (H00 from bulk calculation)  
 $HCLAC$  =  $N \times M$  coupling matrices between lead A and conductor C  
 $HCLCB$  =  $M \times P$  coupling matrices between conductor C and lead B  
 the coupling matrices have to be adapted from H01 of supercell calculation

**NOTES:** (i) The definition of the coupling matrices HCL **MUST BE DONE CAREFULLY case by case**, depending on the particular system definition. (ii) In order to match the hamiltonian matrices at the boundary, it is necessary to check that the diagonal elements of the H00 matrices were aligned. If not a rigid shift must be done.

## 4 How to prepare an input file

According to the methodological scheme of Section 3, it is necessary to use separate input files at the different step of the WANT procedure.

Input files organized as several **NAMelist**, followed by other fields introduced by **CARDs**. Namelist are defined from the flag "&NAMelist" at the beginning to the "/" bar at the end. The order of variables within a namelist is arbitrary. Most variables have default value. If a variable is not explicitly defined in the input file, its default value is assumed.

In the following we report the list and the description of the parameters for each required input file.

### 4.1 Input for DFT-PW calculations

#### Step 1. i-ii: pw.x

WANT is currently interfaced with Pwscf code. For the description of the input for steps 1-2 (Sec. 3) and for further details see the Pwscf manual at [www.pwscf.org](http://www.pwscf.org).

#### Step 1. iii: pw\_export.x

Namelist allowed : &inputpp

Input file layout

&inputpp

...

/

---

List of variables

---

<b>prefix</b>	STRING the first part of the name of all the file written by the code DEFAULT = mandatory
<b>outdir</b>	STRING the scratch directory where the massive data file will be written DEFAULT = "./"
<b>pseudo_dir</b>	STRING directory containing pseudopotential (PP) files DEFAULT = "./"
<b>psfile(i)</b>	STRING files containing i-th PP, where i=1, ntype PP numbering must follow the ordering defined in the input of pw.x
<b>single_file</b>	LOGICAL XXXXXXX aggiungere!!! DEFAULT = ".FALSE."

```

ascii          LOGICAL
                  XXXXXXXX aggiungere!!!
                  DEFAULT = ".FALSE."

```

## 4.2 Input for Wannier function calculations

### Step 2. a-b: disentangle.x wannier.x

Both codes for the WF calculation (disentangle.x and wannier.x) uses the same input file.

Namelist allowed : &CONTROL, &SUBSPACE, &LOCALIZATION

Cards allowed : WANNIER\_CENTERS

Input file layout

```
&CONTROL
```

```
...
```

```
/
```

```
&SUBSPACE
```

```
...
```

```
/
```

```
&LOCALIZATION
```

```
...
```

```
/
```

```
WANNIER_CENTERS ( "crystal" | "angstrom" | "bohr" )
```

```
<type1>      <specific_fmt>
```

```
...
```

```
<typeN>      <specific_fmt>
```

---

```
NAMELIST &CONTROL
```

---

```

prefix          STRING
                  the first part of the name of all the file written by the code
                  DEFAULT = mandatory

```

```

postfix         STRING
                  the tail of the names of the above mentioned files (useful e.g. to distinguish

```

	among different calculations having a common part) DEFAULT = ""
<b>work_dir</b>	STRING the scratch directory where the massive data file will be written) DEFAULT = "./"
<b>title</b>	STRING the title of the calculation) DEFAULT = "Wannier Transport Calculation"
<b>restart_mode</b>	STRING ( "from_scratch"   "restart" ) define whether to restart a previous calculation; at the moment the "restart" choice implies to overwrite the input variables OVERLAPS, PROJECTIONS, START_MODE_DIS and START_MODE_WAN , with the value "from_file" (see below for thier meanings) DEFAULT = "from_scratch"
<b>verbosity</b>	STRING ( "low"   "medium"   "high" ) the level of detail of the code output DEFAULT = "medium"
<b>overlaps</b>	STRING ( "from_scratch"   "from_file" ) determine how to get overlap integrals: "from_scratch": overlaps are calculated from wfcs "from_file": overlaps are read from a previous data file. In this second case dimensions should be consistent DEFAULT = "from_scratch"
<b>projections</b>	STRING ( "from_scratch"   "from_file" ) determine how to get projections integrals: Meaning as before DEFAULT = "from_scratch"
<b>assume_ncpp</b>	LOGICAL if .TRUE. avoids the reading of pseudopotential files assuming that the DFT calculation has been performed within norm-conserving pseudopotentials (no knowledge of them is required in the WANT calc) DEFAULT = ".FALSE."
<b>unitary_thr</b>	REAL threshold for the check of matrix unitary DEFAULT = 1.0d-8

---

NAMELIST &SUBSPACE

---

<b>dimwann</b>	INTEGER
----------------	---------

	the number of wannier functions, i.e. the dimension of the wannier subspace DEFAULT = mandatory
<b>win_min</b>	REAL the lower limit [eV] of the energy window containing the states forming the starting subspace for Wannier functions) DEFAULT = 0.0
<b>win_max</b>	REAL the upper limit [eV] of the energy window described above DEFAULT = 0.0
<b>froz_min</b>	REAL the lower limit [eV] of the energy window containing 'frozen' states which will not enter the calculation of WFs DEFAULT = -2.0000
<b>froz_max</b>	REAL upper limit [eV] of the frozen window described above DEFAULT = -1.0000
<b>alpha_dis</b>	REAL mixing parameter for the disentangle iterative procedure DEFAULT = 0.5
<b>maxiter_dis</b>	INTEGER maximum number of iterations during the disentangle procedure DEFAULT = 1000
<b>nprint_dis</b>	INTEGER every nprint_dis iterations in disentangle minimization write to stdout DEFAULT = 10
<b>nsave_dis</b>	INTEGER every nsave_dis iterations save subspace data to disk DEFAULT = 10
<b>use_blimit</b>	LOGICAL if .TRUE., b vectors are set to zero when calculation overlap augmentations. This essentially means we are doing a sort of thermodynamic limit even if this is not consistent with the actual kpt grid. The .TRUE. value should be considered for debug purposes DEFAULT = ".FALSE."
<b>disentangle_thr</b>	REAL threshold for convergence of the iterative disentangle procedure DEFAULT = 1.0d-8
<b>subspace_init</b>	STRING ( "randomized"   "lower_states"   "upper_states"   "center_projections"   "from_file" ) Determine how the trial subspace is chosen



"randomized" : random starting point is chosen  
 "lower\_states" : the lower DIMWANN bands from DFT calculation are used to define the subspace  
 "upper\_states" : the upper DIMWANN bands from DFT calculation are used to define the subspace  
 "center\_projections" : a subspace is extracted from the DFT bands by means of a projections on the given WANNIER\_TRIAL\_CENTERS (see the section WANNIER\_CENTERS)  
 "from\_file" : subspace initialization is read from an existing data file; this is the choice used during restart  
 DEFAULT = "center\_projections"

**spin\_component** STRING  
 ( "up" | "down" | "none" )  
 defines whether the calculation is spin polarized and if the case which spin component is to be treated  
 DEFAULT = "none"

---

#### NAMELIST & LOCALIZATION

---

**wannier\_thr** REAL  
 threshold for convergence of the iterative wannier minimization  
 DEFAULT = 1.0d-6

**alpha0\_wan** REAL  
 mixing parameter during the first CG part of the wannier minimization  
 DEFAULT = 0.5

**alpha1\_wan** REAL  
 mixing parameter during the second part of the wannier minimization  
 DEFAULT = 0.5

**maxiter0\_wan** INTEGER  
 maximum number of iterations for the first CG part of the wannier minimization  
 DEFAULT = 0.5

**maxiter1\_wan** INTEGER  
 maximum number of iterations for second part of the wannier minimization  
 DEFAULT = 0.5

**nsave\_wan** INTEGER  
 every nsave\_dis iterations save subspace data to disk  
 DEFAULT = 10

**ncg** INTEGER  
 every ncg iterations in the second minimization part, do a CG minimization  
 DEFAULT = 3

**localization\_init** STRING  
 ( "no\_guess" | "randomized" | "center\_projections" | "from\_file" )

Determine how the wannier localization is started  
 "no\_guess" : disentangle states are used as starting point without any further localization guess  
 "randomized" : a random rotation is applied to the states found by the disentangle procedure  
 "center\_projections" : a subspace is extracted from the DFT bands by means of a projections on the given WANNIER\_TRIAL\_CENTERS (see the section WANNIER\_CENTERS)  
 "from\_file" : subspace initialization is read from an existing data file; this is the choice used during restart  
 DEFAULT = "center\_projections"

**ordering\_mode**    STRING  
 ( "none" | "spatial" | "spread" | "complete" ) specifies whether to order the computed Wannier functions and which ordering criterion adopt  
 "none": no ordering is performed  
 "spatial": ordering based on WF center positions  
 "spread": ordering based on WF increasing spreads  
 "complete": SPATIAL + SPREAD for WF with the same centers  
 DEFAULT = "none"

**a\_condmin**        REAL  
 the amplitude of the conditioned minimization functional. If set to zero ordinary minimization is performed  
 DEFAULT = 0.0

**niter\_condmin**    INTEGER  
 the number of steps for which minimization is conditioned.  
 DEFAULT =  $\begin{matrix} \text{maxiter0\_wan} + \text{maxiter1\_wan} & (\text{if } a\_condmin \neq 0.0) \\ 0 & (\text{otherwise}) \end{matrix}$

**dump\_condmin**    REAL  
 the dumping factor for a\_condmin during the conditioned minimization.  
 If the dumping factor is specified, after niter\_condmin iterations a\_condmin is dumped according to  $a\_condmin = a\_condmin * dump\_condmin$  at each iteration  
 DEFAULT = 0.0

---

CARD WANNIER\_CENTERS

---

**WANNIER\_CENTERS** ( "crystal" | "angstrom" | "bohr" )

Aside the tag WANNIER\_CENTERS, units for positions maybe specified:

"crystal" : relative coordinates on the basis of a1,a2,a3 lattice vector (default)  
 "bohr" : cartesian coordinates in bohr  
 "angstrom" : cartesian coordinates in angstrom

Next the card contains DIMWANN lines giving the trial centers for the WFs. Depending on the <TYPE> flag at the beginning of the line, formats are different.

<TYPE> may assume the values: "atomic" , "1gauss", "2gauss"

IF ( TYPE == "atomic" )	→	atomic	iatomic	l m		[weight]
IF ( TYPE == "1gauss" )	→	1gauss	x y z	l m	rloc	[weight]
IF ( TYPE == "2gauss" )	→	2gauss	x y z	xx yy zz	rloc	[weight]

### TYPE == "1gauss"

The trial center is given by a single gaussian set at a given position with a given angular momentum. Standard positions are usually atomic sites or bond midpoints.

**x, y, z** REAL  
define the position of the trial function. Units maybe specified aside the tag WANNIER\_CENTERS: see above for more details.

**l, m** INTEGER  
are the angular momentum quantum numbers for the spherical harmonics giving the angular part of the trial WF. l can be set equal to 0, 1, or 2, (and m values are then as usual) for standard spherical harmonics or l == -1 indicate the sp3 geometry. Here spherical harmonics are the real ones:

l == -1:	m = -4	→	1,1,-1	dir
	m = -3	→	1,-1, 1	dir
	m = -2	→	-1, 1,1	dir
	m = -1	→	-1,-1,-1	dir
	m = 1	→	1,1, 1	dir
	m = 2	→	1,-1,-1	dir
	m = 3	→	-1,1,-1	dir
	m = 4	→	-1,-1, 1	dir
l == 0:	m = 0	→	spherical	
l == 1:	m = -1	→	x	
	m = 0	→	z	
	m = 1	→	y	
l == 2:	m = -2	→	x <sup>2</sup> - y <sup>2</sup>	
	m = -1	→	xz	
	m = 0	→	3z <sup>2</sup> - r <sup>2</sup>	
	m = 1	→	yz	
	m = 2	→	xy	

**rloc** REAL  
specifies the spread of the gaussian used for the radial part of the trial WF. Units are bohr for both "bohr" and "crystal" and angstrom for "angstrom" specifier.

**weight** REAL  
this value is required when conditioned minimization is performed. In case, it should be in the interval [0, 1] and weights the relative importance of each center in the penalty functional. weight = 0 is used to switch off the constrain for a given center

**TYPE == "2gauss"**

The trial function is given as the difference between gaussians with s-symmetry placed at positions selected by the user. This is useful to mimic a antibonding state.

<b>x, y, z</b>	REAL as before for TYPE == "1gauss"
<b>xx, yy, zz</b>	REAL as before for x,y,z for the units, specify the center of a second gaussian used to build up the trial WF. This second case could be useful to describe anti-bonding WF.
<b>rloc</b>	REAL as before for TYPE == "1gauss"
<b>weight</b>	REAL as before for TYPE == "1gauss"

**TYPE == "atomic"**

Atomic (pseudo)-orbitals from pseudopotential files are used as trial functions. They are specified by the atomic index and the required angular momentum quantum numbers.

<b>iatom</b>	INTEGER the index of the chosen atom, the same of Pwscf calculation
<b>l, m</b>	REAL as before for TYPE == "1gauss"
<b>weight</b>	REAL as before for TYPE == "1gauss"

**4.3 Input for electronic transport calculations****Step 3 : conductor.x**

Both bulk and two-terminal calculations use similar input. Labels follows the scheme of Section 3.

Namelists allowed : &INPUT\_CONDUCTOR

Cards allowed (tagged): <HAMILTONIAN\_DATA>, <subcard> (xlm format) Input file layout

```

&INPUT_CONDUCTOR
...
/
<HAMILTONIAN_DATA>
  <subcard>
    &MATRIX_DATA
    ...

```

```

/
< /subcard>
< /HAMILTONIAN_DATA>

```

---

NAMELIST &INPUT.CONDUCTOR

---

<b>dimA</b>	INTEGER number of sites in the lead A DEFAULT = 0
<b>dimB</b>	INTEGER number of sites in the lead B DEFAULT = 0
<b>dimC</b>	INTEGER number of sites in the conductor C DEFAULT = 0
<b>calculation_type</b>	STRING ( "conductor"   "bulk" ) determines which kind of calculation should be performed: "conductor": ordinary transport calculation for a leads conductor lead interface "bulk": transport in a bulk system DEFAULT: "conductor"
<b>looverlap</b>	LOGICAL If .TRUE. reads the overlap matrices from file, otherwise basis orthonormality is assumed (which is by definition the case of Wannier functions) DEFAULT : .FALSE.
<b>ne</b>	INTEGER dimension of the energy grid for transmittance and spectral function calculation DEFAULT = 1000
<b>niterx</b>	INTEGER maximum number of iterations in the calculation of transfer matrices DEFAULT = 200
<b>emin</b>	REAL lower limit [eV] of the energy grid dimensioned by NE DEFAULT = -10.0
<b>emax</b>	REAL upper limit [eV] of the energy grid dimensioned by NE DEFAULT = +10.0
<b>bias</b>	REAL bias voltage [eV] across the conductor region. DEFAULT = 0.0



Allowed value are:

- R = 0** → On site H00 hamiltonian  
 $H00_{mn} = \langle w_{m\mathbf{0}} | \hat{H} | w_{n\mathbf{0}} \rangle$   
i.e. the hamiltonian matrix corresponding to WFs localized  
in the origin cell (**R = 0 0 0**)
- R = 1** → Hopping H01 hamiltonian  
 $H01_{mn} = \langle w_{m\mathbf{0}} | \hat{H} | w_{n\mathbf{1}} \rangle$   
i.e. the hamiltonian matrix among to WFs localized  
in the origin cell and in the first neighbor cell along  
x direction (**R = 1 0 0**)
- R = 2** → Hopping H01 hamiltonian  
 $H01_{mn} = \langle w_{m\mathbf{0}} | \hat{H} | w_{n\mathbf{1}} \rangle$   
i.e. the hamiltonian matrix among to WFs localized  
in the origin cell and in the first neighbor cell along  
y direction (**R = 0 1 0**)
- R = 3** → Hopping H01 hamiltonian  
 $H01_{mn} = \langle w_{m\mathbf{0}} | \hat{H} | w_{n\mathbf{1}} \rangle$   
i.e. the hamiltonian matrix among to WFs localized  
in the origin cell and in the first neighbor cell along  
z direction (**R = 0 0 1**)

## 4.4 Input for post-processing calculations

### 4.4.1 bands.x

Namelist allowed : &INPUT

Input file layout

&INPUT

...

/

kpt\_label\_1

...

kpt\_label\_N

---

NAMELIST &INPUT

---

**prefix**

STRING

the first part of the name of all the file written by the code should be equal to the value given in the main calculations.

DEFAULT = mandatory

**postfix**

STRING

the tail of the names of the above mentioned files (useful e.g. to distinguish

among different calculations having a common part). should be equal to the value given in the main calculations.

DEFAULT = ""

<b>work_dir</b>	STRING the scratch directory where the massive data file will be written DEFAULT = "./"
<b>verbosity</b>	STRING ( "low"   "medium"   "high" ) the level of detail of the code output DEFAULT = "medium"
<b>nkpts_in</b>	INTEGER number of edge kpts defining the directions on which bands will be calculated DEFAULT: mandatory
<b>nkpts_max</b>	INTEGER maximum number of interpolated kpoints DEFAULT = 100
<b>spin_component</b>	STRING ( "up"   "down"   "none" ) define whether the calculation is spin polarized and if the case which spin component is to be treated. DEFAULT = 100

After the INPUT namelist for each of the NSPTS kpts two lines with the following format must be provided:

<b>kpt_label</b>	kx ky kz
<b>kpt_label</b>	CHARACTER(*) it is a string with the name of the kpoint
<b>kx, ky, kz</b>	REAL component of the kpt vector in units of crystal reciprocal lattice vector i.e. $k = kx * b1 + ky * b2 + kz * b3$

#### 4.4.2 plot.x

Namelists allowed : &INPUT

Input file layout

&INPUT

...

/

---

NAMELIST &INPUT

---



<b>prefix</b>	<p>STRING</p> <p>the first part of the name of all the file written by the code should be equal to the value given in the main calculations.</p> <p>DEFAULT = mandatory</p>
<b>postfix</b>	<p>STRING</p> <p>the tail of the names of the above mentioned files (useful e.g. to distinguish among different calculations having a common part). should be equal to the value given in the main calculations.</p> <p>DEFAULT = ""</p>
<b>work_dir</b>	<p>STRING</p> <p>the scratch directory where the massive data file will be written</p> <p>DEFAULT = "./"</p>
<b>wann</b>	<p>STRING</p> <p>specifies the indexes of the Wannier functions to be plotted. It is a string of format e.g "1-3,5,7-9" (analogous to the fmt used to specify pages to very common print tools)</p> <p>DEFAULT = mandatory</p>
<b>data_type</b>	<p>STRING</p> <p>("modulus"   "real"   "imaginary")</p> <p>specifies the type of data plotted:</p> <p>"modulus": plot the real space square modulus of the WFs. "real": plot the real part (in real space) of the WFs. "imaginary": plot the imaginary part (in real space) of the WFs this choice should be intended as a check because WFs are expected to be more or less "real".</p> <p>DEFAULT = "modulus"</p>
<b>output_fmt</b>	<p>STRING</p> <p>( "plt"   "txt"   "cube" )</p> <p>Define the format of the output file. PLT is binary and smaller than CUBE and TXT. While CUBE deals also with non-orthorombic lattices, TXT is suitable to be converted to further format.</p> <p>DEFAULT = "plt"</p>
<b>r1min, r1max</b>	<p>REAL</p> <p>the starting and ending points of the plotting cell along a1 dir, in units of a1 lattice vector (crystal coord).</p> <p>DEFAULT = -0.5, 0.5</p>
<b>r2min, r2max</b>	<p>REAL</p> <p>as before but for a2 direction.</p> <p>DEFAULT = -0.5, 0.5</p>
<b>r3min, r3max</b>	<p>REAL</p> <p>as before but for a3 direction.</p> <p>DEFAULT = -0.5, 0.5</p>

<b>assume_ncpp</b>	<p>LOGICAL</p> <p>if using DFT pseudopotentials not readable in WANT set this value to .TRUE. in order to avoid PP reading.</p> <p>DEFAULT = ".FALSE."</p>
<b>locate_wf</b>	<p>LOGICAL</p> <p>if .TRUE. move the WFs in a unit cell centered around the midpoint of the plotting cell. Useful to plot purposes.</p> <p>DEFAULT = ".TRUE."</p>
<b>spin_component</b>	<p>STRING</p> <p>( "up"   "down"   "none" )</p> <p>define whether the calculation is spin polarized and if the case which spin component is to be treated.</p> <p>DEFAULT = "none"</p>

## References

- [1] A. Calzolari, N. Marzari, I. Souza, and M. Buongiorno Nardelli, Phys. Rev. B **69**, 035108 (2004).
- [2] M. Buongiorno Nardelli, Phys. Rev. B **60**, 7828 (1999).
- [3] M. Buongiorno Nardelli and J. Bernholc, Phys. Rev. B **60**, R16338 (1999).
- [4] M. Buongiorno Nardelli, J.-L. Fattebert, and J. Bernholc, Phys. Rev. B **64**, 245423 (2001).
- [5] R. Landauer, Philos. Mag. **21**, 863 (1970).
- [6] S. Datta, *Electronic transport in mesoscopic systems*, Cambridge University Press, 1995.
- [7] D. S. Fisher and P. A. Lee, Phys. Rev. B **23**, 6851 (1981).
- [8] Y. Meir and N. S. Wingreen, Phys. Rev. Lett. **68**, 2512 (1992).
- [9] D. H. Lee and J. D. Joannopoulos, Phys. Rev. B **23**, 4988 (1981).
- [10] D. H. Lee and J. D. Joannopoulos, Phys. Rev. B **23**, 4997 (1981).
- [11] M. Lopez-Sancho, J. Lopez-Sancho, and J. Rubio, J. Phys. F: Metal Phys. **14**, 1205 (1984).
- [12] M. Lopez-Sancho, J. Lopez-Sancho, and J. Rubio, J. Phys. F: Metal Phys. **15**, 851 (1985).
- [13] F. Garcia-Moliner and V. Velasco, *Theory of Single and Multiple Interfaces*, World Scientific, Singapore, 1992.
- [14] F. Garcia-Moliner and V. Velasco, Phys. Rep. **200**, 83 (1991).
- [15] A. Baldereschi, Phys. Rev. B **7**, 5212 (1973).
- [16] W. Kohn, Phys. Rev. **115**, 809 (1959).
- [17] M. Geller and W. Kohn, Phys. Rev. B **48**, 1485 (1993).
- [18] N. Marzari and D. Vanderbilt, Phys. Rev. B **56**, 12847 (1997).
- [19] S. F. Boys, Rev. Mod. Phys. **32**, 296 (1960).
- [20] E. Blount, Surf. Sci. **13**, 305 (1962).
- [21] P. L. Silvestrelli, N. Marzari, D. Vanderbilt, and M. Parrinello, Solid State Commun. **107**, 7 (1998).
- [22] L. Bernasconi and P. A. Madden, J. Mol. Struct. **544**, 49 (2001).
- [23] I. Souza, N. Marzari, and D. Vanderbilt, Phys. Rev. B **65**, 035109 (2002).