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Interference and k -point sampling in the supercell approach to phase-coherent transport

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We present a systematic study of interference and k -point sampling effects in the supercell approach to phase-coherent electron transport. We use a representative tight-binding model to show that interference between the repeated images is a small effect compared to the error introduced by using only the Γ -point for a supercell containing $(3,3)$ sites in the transverse plane. An insufficient k -point sampling can introduce strong but unphysical features in the transmission function which can be traced to the presence of van Hove singularities in the lead. We present a first-principles calculation of the transmission through a Pt contact which shows that the k -point sampling is also important for realistic systems.

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The growing interest for exploring the electronic conduction properties of nanostructures has led to the development of a variety of numerical methods aimed at describing phase-coherent electron transport in atomic-scale systems from first principles. Many of these methods are based on a supercell approach where the system of interest is defined inside a finite computational cell which is then repeated periodically in the directions perpendicular to the transport direction while the appropriate open boundary conditions needed to describe the coupling to the electrodes are imposed in the parallel direction.¹⁻¹¹ Within the supercell approach, one is obviously limited to consider transport through an infinite array of contacts, however, in practice this limitation is usually ignored. Whether the supercell approach can provide a good description of transport through a single, isolated contact depends on the degree of interference between the repeated images. As the transverse dimensions of the supercell are increased the interference is expected to decrease and the result should approach that of a single contact. For practical first-principles calculations this limit is very hard to realize due to the computational cost associated with an enlargement of the supercell and therefore relatively small supercells containing, e.g., 3×3 atoms in the transverse plane are typically used as a compromise.^{2,3,6-9,11}

A second approximation which is also commonly used, is to evaluate the transmission function only at the Γ -point of the two-dimensional Brillouin zone (BZ) of the plane perpendicular to the transport direction. For example, this approximation has been applied in transport studies of Au (Ref. 9) and Na (Ref. 10) wires and to molecular contacts containing di-thiol benzene.¹¹ Again, when the transverse dimensions of the supercell are increased the Γ -point approximation eventually becomes valid. While the role of k -point sampling for total energy calculations is well understood, its influence on the transmission function has, to our knowledge, so far not been systematically explored. In a simple picture the transmission function is similar to a density of states (DOS) and as such it should be more sensitive to the k -point sampling than the total energy which is an integral of the DOS.

In this paper we investigate the effects of k -point sampling and interference on the transmission function by means

of two examples. The first example is based on a tight-binding (TB) model. The simplicity of this model allows for a complete separation of interference and k -point sampling effects and thus provides an opportunity to study the relative importance of these factors for the transmission. Our results show that even for a small cell containing 3×3 sites in the transverse directions, interference effects are not very significant compared to the error introduced by an insufficient k -point sampling. The second example is a first-principles calculation for an atomic Pt contact demonstrating that k -point sampling is essential also for realistic systems.

We consider a tight-binding model for resonant transport through an array of identical states which we refer to as “molecular states.” The leads are described by semi-infinite cubic lattices with zero on-site energies and hopping parameter t_i between i th nearest neighbors. In all calculations we use the parameters $t_1=1.0$, $t_2=0.4$, $t_3=0.1$ to describe the leads. The molecular states have on-site energy ϵ_a and are coupled to the leads via a single lattice site by the hopping parameter t . The number of lattice sites between the periodically repeated molecular states is N in each direction. A two-dimensional version of the model is illustrated in Fig. 1.

The transmission function is calculated from the general formula¹²

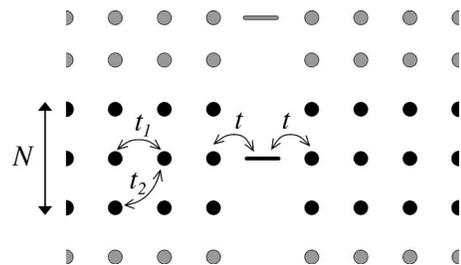


FIG. 1. Two-dimensional illustration of the TB model. A supercell is indicated by the black dots. The molecular states have on-site energy ϵ_a and are coupled symmetrically to semi-infinite cubic lattices via the hopping parameter t . All on-site energies of the lattice are zero and the hopping connecting i th nearest neighbors is denoted t_i .

$$T(\varepsilon) = \text{Tr}[G_C^r(\varepsilon)\Gamma_L(\varepsilon)G_C^a(\varepsilon)\Gamma_R(\varepsilon)], \quad (1)$$

where $G_C^{r/a}$ is the retarded/advanced Green's function of the central region. The coupling strength is given by $\Gamma_{L/R} = i(\Sigma_{L/R} - \Sigma_{L/R}^\dagger)$, where $\Sigma_{L/R}$ is the self-energy due to the left/right lead. In the TB model the central region consists of the (infinite) array of molecular states.

We label all TB orbitals in the model, including the molecular states, by integer coordinates $|n, m, l\rangle$ where n refers to the direction of transport (x axis) and m, l refer to the perpendicular directions (y, z axes). We say that an orbital $|n, m, l\rangle$ belongs to the central supercell if $m, l \in \{1, \dots, N\}$. For each orbital in the central supercell we form the Bloch states

$$|k_y, k_z; n, m, l\rangle = \sum_{M, L \in \mathbb{Z}} e^{i(k_y M + k_z L)} |n, m + MN, l + LN\rangle, \quad (2)$$

where $k_y, k_z \in]-\pi; \pi]$. Due to the periodicity of the system in the directions perpendicular to the transport, all matrices entering Eq. (1) are diagonal with respect to $\mathbf{k} = (k_y, k_z)$ and consequently the total transmission decomposes into a sum of \mathbf{k} -dependent transmissions,

$$T(\mathbf{k}; \varepsilon) = \text{Tr}[G_C^r(\mathbf{k}; \varepsilon)\Gamma_L(\mathbf{k}; \varepsilon)G_C^a(\mathbf{k}; \varepsilon)\Gamma_R(\mathbf{k}; \varepsilon)]. \quad (3)$$

The transmission *per* supercell is then evaluated as

$$T(\varepsilon) = \frac{1}{\Omega_{\text{BZ}}} \int_{\text{1BZ}} T(\mathbf{k}; \varepsilon) d\mathbf{k}, \quad (4)$$

where Ω_{BZ} is the area of the first Brillouin zone. In practice the integral is converted into a finite sum $\int d\mathbf{k}/\Omega_{\text{BZ}} \rightarrow \sum_{\mathbf{k}} W_{\mathbf{k}}$, where $W_{\mathbf{k}}$ represent the weights of the discrete \mathbf{k} points.

In order to distinguish between interference effects due to the repeated supercells and electronic structure effects resulting from an insufficient \mathbf{k} -point sampling we study the TB model in three different limits which we denote by A, B and C. Limit A is the ideal situation where the result for the transmission function has been converged both with respect to the transverse dimensions of the supercell (N) and the denseness of the \mathbf{k} -point sampling. This is the situation where no interference between the repeated molecular states occur and it thus corresponds to transmission through a single state. In limit B the transverse dimensions of the supercell are fixed to $N=3$ and the \mathbf{k} -point sampling is converged to account properly for the electronic structure. Limit C is like B except that the 1BZ is sampled only at the Γ -point.

In Fig. 2 we show the transmission function in each of the three limits for the TB model with parameters $\varepsilon_a=0$ and $t=2$. This choice for the parameter t corresponds to rather strong coupling between the molecular states and the leads, and is relevant for atomic metal contacts and chains or molecular contacts in which the molecule forms a covalent bond with the leads. The fully converged limit A is reached for $N=10$ and with 25 \mathbf{k} -points, where all interference and \mathbf{k} -point sampling effects have disappeared. The similarity between curves A and B shows that interference effects are present but not very strong, even for this small supercell with

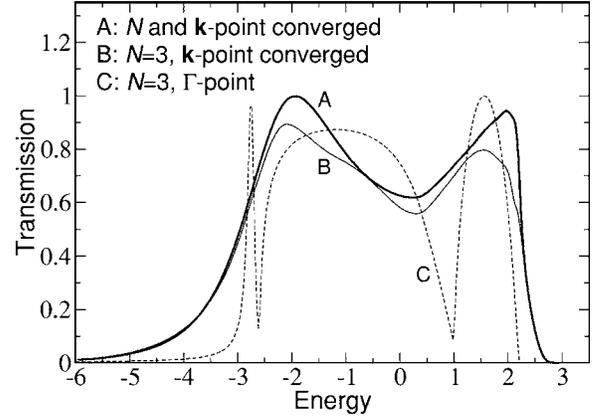


FIG. 2. Transmission through an array of molecular states in the 3d TB model with $\varepsilon_a=0$ and $t=2$. For A the result has been converged both with respect to N and the number of \mathbf{k} points. For B the number of \mathbf{k} -points has been converged, while C represents the case of Γ -point only. The difference between A and B is thus pure interference while B and C differ due to an insufficient \mathbf{k} -point sampling.

$N=3$. On the other hand, a correct description of the electronic structure is crucial as can be seen from the large discrepancy between curves A and C.

In order to understand the origin of the large difference between curves B and C, we have calculated the relevant lead DOS for both cases. We use a unit cell containing 3×3 sites in the transverse plane and calculate the DOS of the TB lattice using, respectively, the Γ -point and a converged set of \mathbf{k} -points. The result is shown in the right panel of Fig. 3. The Γ -point DOS has much more structure than the total DOS, and exhibits pronounced peaks due to van Hove singularities at the 1d band edges, see left panel. The strong van Hove singularity is a characteristic of 1d systems and does generally not occur in extended bulk systems. By sampling the 1BZ in the transverse plane, the density of van Hove singularities is increased and this provides an effective smearing of the DOS. By comparing the Γ -point DOS with

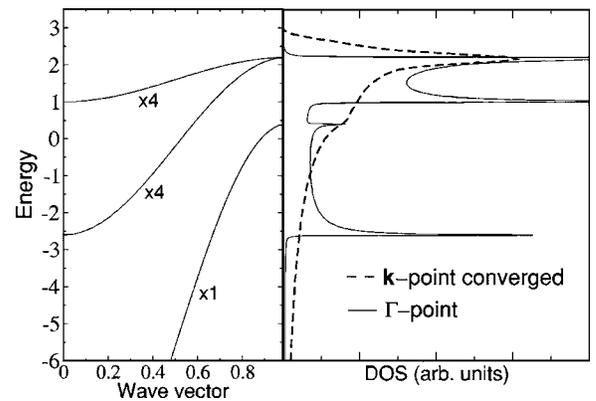


FIG. 3. Left, band diagram for the cubic TB lattice along the line $(k_x, 0, 0)$ corresponding to the transport direction. The unit cell contains 3×3 sites in the transverse plane. The degeneracy of each band is indicated. Right, DOS corresponding to the band diagram (full line) and the total DOS of the TB lattice (dashed line) calculated using a converged set of \mathbf{k} -points.

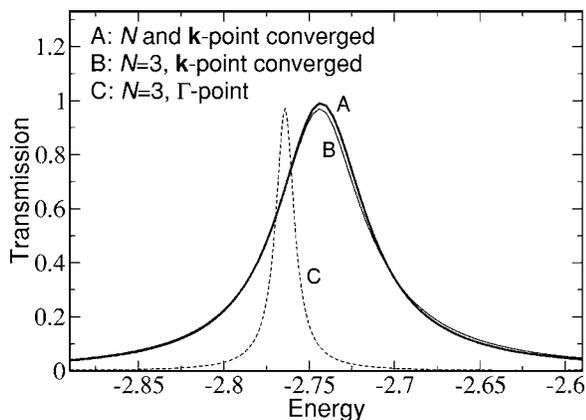


FIG. 4. Like Fig. 2, except that the parameters $\varepsilon_a = -2.7$ and $t = 0.3$ have been used.

curve *C* of Fig. 2 we see that the dramatic dips in the transmission at energies -2.6 and 1.0 coincide with two van Hove singularities. This clearly indicates that the strong variation in the Γ -point transmission is due to the artificial one-dimensional description of the electronic structure of the lead.

As the coupling between the molecular states and the leads is reduced, the transmission function narrows down and eventually turns into a Lorentzian shaped resonance located close to the bare energy, ε_a , with a width determined by the product of t^2 and the DOS of the uncoupled lead at ε_a . This suggests that the width of the resonance should be sensitive to the lead DOS, and in particular that the Γ -point approximation could break down when the resonance lies close to a van Hove singularity of the lead. That this is in fact the case can be seen in Fig. 4 which shows the transmission in the three limits *A*, *B*, and *C* for $\varepsilon_a = -2.7$ and $t = 0.3$. The curves *A* and *B* are almost identical meaning that interference effects are negligible. In contrast the Γ -point resonance is shifted too far down in energy and is about four times too narrow as compared to the \mathbf{k} -point sampled result. We conclude that \mathbf{k} -point sampling can be of importance also for weakly coupled systems. We note, however, that the example shown in Fig. 4 is a worst case scenario due to the close proximity of ε_a with a van Hove singularity.

As a second example we have calculated the transmission function for a Pt contact using first principles techniques to evaluate the Hamiltonian. More details on the computational method can be found in Ref. 13. The supercell contains 3×3 atoms in the directions perpendicular to the transport direction as sketched in the inset of Fig. 5. The transmission has been calculated using 1, 8, and 18 irreducible \mathbf{k} -points, and from the result we conclude that the transmission function has converged to within 5% using 8 \mathbf{k} -points. In fact the same conclusion holds for several other systems that we have studied, indicating that convergence of the transmission function can in general be achieved using 8 irreducible

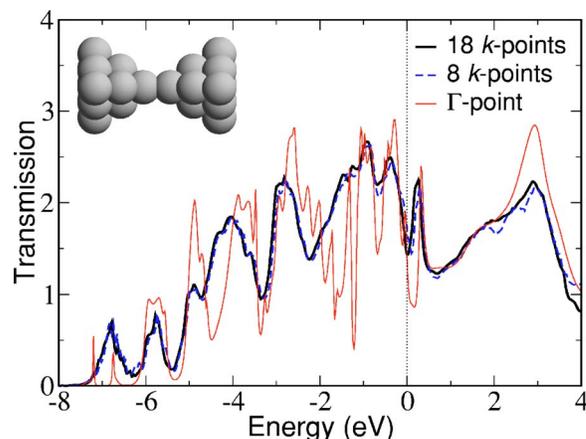


FIG. 5. (Color online) Transmission through the Pt contact shown in the inset. The transmission has been calculated using 1, 8, and 18 \mathbf{k} -points in the transverse BZ.

\mathbf{k} -points for a system containing three atoms in the transverse cell. Returning to Fig. 5 it can be seen that the Γ -point transmission has too much structure and represents a rather poor approximation. This is very similar to what we found for the TB model, except that the number of unphysical dips/peaks in the transmission function is larger for the Pt contact. This is because the effective number of orbitals in a cross section of the supercell, which determines the number of $1d$ bands in the lead and thus the density of van Hove singularities, is larger in the first-principles calculation than in the TB model.

Generally, the Γ -point approximation is expected to be valid when the distance between the van Hove singularities is comparable to the width of the features induced in the transmission function by the van Hove singularities. It is our experience, both from TB studies and first-principles calculations, that the transmission calculated at a single \mathbf{k} -point is improved if the \mathbf{k} -point is chosen away from the Γ -point. This is due to the lower degree of degeneracy among the $1d$ lead bands at a general \mathbf{k} -point, which spreads out the van Hove singularities.

In conclusion, we have investigated the importance of \mathbf{k} -point sampling as well as interference among the repeated images in the supercell approach to coherent electron transport. Using a representative TB model, it was found that the \mathbf{k} -point sampling is the most important factor of the two, and that an insufficient \mathbf{k} -point sampling or, in particular, a Γ -point only sampling, can introduce spurious features in the transmission function which could be wrongly attributed to physical properties of the contact under study. The origin of these spurious features was traced to the presence of van Hove singularities in the lead.

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