Stroboscopic wave packet description of time-dependent currents through ring-shaped nanostructures

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Abstract. We present an implementation of a new method for explicit simulations of time-dependent electric currents through nanojunctions. The method is based on unitary propagation of stroboscopic wave packet states and is designed to treat open systems with fluctuating number of electrons while preserving full quantum coherence throughout the whole infinite system. We demonstrate the performance of the method on a model system consisting of a ring-shaped nanojunction with two semi-infinite tight-binding leads. Time-dependent electron current responses to abrupt bias turn-on or gate potential switching are computed for several ring configurations and ring-leads coupling parameters. The found current-carrying stationary states agree well with the predictions of the Landauer formula. As examples of genuinely time-dependent process we explore the presence of circulating currents in the rings in transient regimes and the effect of a time-dependent gate potential.

1 Introduction

Transition from the state with zero electric current to a state with nonzero current in nanoelectronic devices is a technologically important example of non-equilibrium quantum dynamics of an open many-electron system. It exhibits several significantly different time scales related to relaxation of electrons with important consequences for the prospects of their applications in nanoelectronics. The common problem in theoretical quantum transport description in both stationary and time-dependent situations is proper inclusion of the semi-infinite leads. The number of degrees of freedom in the nanojunction, which are explicitly considered, has to be sufficiently small in order to be numerically tractable. Hence the boundary conditions at the junction must be correctly described and included. An approach widely used for stationary systems are the non-equilibrium Green’s functions (NEGF) with lead self-energies [1,2] which include the physics induced by the leads.

The description of boundary conditions of the finite nanojunction becomes more complicated in time-dependent problems. The non-stationary quantum transport has been addressed for non-interacting electrons [3], within the time-dependent density-functional theory (TDDFT) [4,5] or even the many-body perturbation theory [6]. It has been shown that self-energies – now time-dependent – could in principle be used also in this case as long as the leads are treated at the TDDFT level. In practice, it is a quite complicated approach especially for realistically structured systems so that simplifications of the formalism are necessary [7,8]. Zheng et al. [9] proved the so-called holographic electron density theorem and developed a new method for time-dependent open electronic systems. Further effort in this direction led to computationally more efficient density-functional tight-binding method [8]. Very recent works of Xie et al. [10] on time-dependent quantum transport are based on Liouville-von-Neumann equation for single-electron density matrix. Another recent work by a different group [11] uses generalised master equation approach to mesoscopic time-dependent transport. Non-equilibrium thermodynamical theory of interacting tunnelling transport has been presented by Hyldgaard [12].

Another class of methods to tackle time-dependent transport with open boundary conditions has been in development [13–15]. The scope of these methods is wider than just elastic transport. The methods are known as correlated electron-ion dynamics (CEID). The dynamics is based on Ehrenfest molecular dynamics and its extensions. The electronic degrees of freedom in CEID are divided between the central system and its environment which is facilitated by the formalism of one-particle density matrices. A damping term has been introduced used in the open-boundary equations of motion [13]; this term keeps the environment close to a reference state. In further
development of the method [14], open boundaries have
been introduced in a new way which represents an ex-
plicit realisation of an external battery. We remark that
this method does not conserve coherence between injec-
tion and subsequent scattering of the electrons.

In the present work we address the open-boundaries
time-dependent quantum transport problem using the
stroboscopic wave packet (SWP) basis method, princi-
ple of which have been provided in the works [16,17].
We study time-dependent currents through model-system
nanojunctions formed by small rings inserted to electric
circuit formed by mono-atomic leads. We also present
new developments of the stroboscopic wave packet ap-
proach (SWPA) which are necessary in order to describe
the systems with atomistic structure. While in the origi-
nal works [16,17] structureless electrodes and very sim-
ple tunnelling barriers have been used to demonstrate
the method, here we work with atomistic models of the
electrodes and more complex nanojunctions. The forma-
tion presented here solves the time-dependent Schrödinger
equation (SchE) for independent electrons within the
tight-binding (TB) approximation. SchE is solved numeri-
cally with state vectors expanded in the stroboscopic wave
packet basis representation (SWB) [16,17]. The SWPA has
been designed to be specifically suited for time-dependent
transport through nanojunctions. Its main advantage is to
make possible explicit integration of equations of motions
of nanojunction degrees of freedom with full quantum
coherence preserved throughout the whole infinite sys-
tem. This can be viewed also as an open-system treat-
ment with correct inclusion of boundary conditions. In
the present paper we focus our study on simple model
cases of rings and monoatomic leads with possible exten-
sion to realistic systems. Our main interest are transient
currents developed in response to abruptly applied bias
and time-dependent gate potential. We also refer to sta-
tionary results obtained by other authors using analytical
methods.

Transport properties of atomic-scale sized rings have
been studied by several authors in most cases on the
Hückel/tight-binding level of description and in the sta-
tionary regime. Effect of the asymmetric position of lead
on transmittance has been studied in reference [18] by
means of Green’s functions. Authors of work [19] have
developed a source-sink potential method for convenient
analytical treatment of molecular electronic devices in-
cluding conjugated systems. This approach has been used in
reference [20] to obtain the form of the transmission
with explicitly given dependence on the molecular skele-
ton and its connection to leads. Authors of reference [21]
generalised the so-called waveguide approach [22] (WGA)
to systems described by the TB approximation. Result-
ing methodology – the tight-binding waveguide approach
(TBWGA) – can be interpreted as a route to an exact
solution of the stationary SchE for given class of TB
models. An explicit formula for transmittance has been
derived [21] for rings composed of identical atoms and
identical nearest-neighbour couplings and coupled to two
leads with equal chemical potentials in both of them.

The authors discussed conditions for occurrence of cir-
culating currents in such quantum rings. More recently
Sparks et al. formulated the stationary problem more gen-
erally, considering a multibranch device in a TB approxi-
mation [23]. They provided an explicit formula for system
transmittance valid for a large class of systems includ-
ing those with different chemical potentials in different
leads. The formula has been obtained by an exact solution
of the stationary SchE for the whole system and has also
been related to Green’s function analysis. Explicit results
in reference [23] facilitate the study of quantum interfer-
ence effects in stationary regime for wide class of systems.
We refer to these results especially when the long-time
behaviour of our time-dependent method is discussed. An
inspiring computational work was done by Saha et al. [24].
The authors study a true atomistic model of a quantum-
interference-controlled molecular transistor formed by an
18-annulene attached between zigzag graphene nanorib-
bons. The device was studied by a multi-terminal NEGF-
DFT formalism [25] and the current-switching effect was
confirmed.

On the experimental side, ring-shaped tunnel nano-
junctions can be formed by cyclic organic molecules at-
tached between two electrodes (see for example [26–29]).
Conductors can be formed by metallic (usually Au or
Pt) electrodes or by graphene nanoribbons [30,31]. These
works employ direct attachment of particular molecules in
between metallic electrodes (Pt-C or Au-C bonds) using
mechanically controllable break junctions. Such contacts
are reported to be more conductive and stable than previ-
ously more common junctions employing a bridging thiol
group (thus forming a metal-S-C bond) [32]. Another re-
alisation of nanometer-scale sized rings is fabrication of
structures on proper substrates [33–35]. This can be done
using molecular-beam epitaxy, wet etching and optical or
electron-beam lithography. Finally we note that the con-
cept of a quantum interference driven transistor started
to be more explicitly discussed in literature relatively re-
cently [36]. It still represents an experimental challenge
and a major bunch of experimental results is yet expected
to come.

The paper is organised as follows. In Section 2 we de-
scribe the model of the atomic ring. In Section 3 we ex-
plain the implementation of the stroboscopic wavepacket
method. In Section 4 we provide the formula which we
implement for electron current calculations. Section 5 de-
scribes stationary results as a basis from which we move
into non-stationary regime in Section 6, which contains
our main results.

2 The model of the ring with contacts

We consider a linear chain of atoms, each two being a lat-
tice constant a apart, with one finite ring which presents
an obstacle for the flow of electrons (Fig. 1). All couplings
are considered within the TB approximation in which we
limit our treatment to one orbital per atom. The chain
is periodic apart from the ring region. Hence, the TB
The on-site energies within the ring, if not differently specified, will thus take values \( \epsilon + cU(t)/2 \). The on-site energies in the right lead will remain unchanged, equal to the equilibrium value \( \epsilon \).

Three exceptions from the above prescription of the perturbation \( V(t) = H^1 + \hat{H}^2(t) \) are studied: (i) in Section 6.1.2, where a uniform slope of the potential energy within the ring is used instead of the spatially constant value \( eU(t)/2 \); (ii) in Section 6.3, where a time- and branch-dependent gate potential is used; (iii) finally consideration of variable coupling strength between the ring and the leads in Section 6.4.

### 3 The stroboscopic basis set

The stroboscopic wavepacket basis set consists of wavepackets (Fig. 2) constructed from the eigenstates of the unperturbed (lead’s) Hamiltonian (2). The mathematical expression for the basis set vectors is [16,17]

\[
|n, \alpha, m; t\rangle = \exp \left[ \frac{i}{\hbar} (m\tau_n + t) \hat{H}^0 \right] \frac{1}{\sqrt{2\Delta n}} \int_{E_{n-1}}^{E_n} d\mathcal{E} |\mathcal{E}, \alpha\rangle
\]

where \( \hat{H}^0 \) is the lead’s Hamiltonian and \( |\mathcal{E}, \alpha\rangle \) are its eigenstates normalised so that \( \langle \mathcal{E}, \alpha | \mathcal{E}', \alpha'\rangle = \delta(\mathcal{E} - \mathcal{E}') \delta_{\alpha, \alpha'} \). Each basis function or wavepacket (5) is uniquely characterised by three indexes: the band index \( n \), the time shift index \( m \), the degeneracy index \( \alpha \) and time \( t \).

**The band index \( n \).** The unperturbed Hamiltonian (2) gives the dispersion relation

\[
E(k) = \epsilon + 2t_B \cos(k\pi)
\]

The corresponding energy range \([\epsilon + 2t_B, \epsilon - 2t_B]\) is in the SWPA divided into non-overlapping bands [16]. rth band has its energies \( \mathcal{E} \in [\mathcal{E}_{n-1}, \mathcal{E}_n] \). In our present work we use two equally wide bands\(^1\) \((N_0 = 2)\) spanning the whole TB energy range. The band index \( n \) then takes values 1 and 2.

**The time-shift index \( m \).** Within each band, different, mutually orthogonal basis functions are obtained by time shifts \( m\tau_n \), where the time step

\[
\tau_n = \frac{2\pi \hbar}{\Delta \mathcal{E}_n}
\]

\(^1\) Stroboscopic wavepacket method can in general work with arbitrary bulk (or even non-periodic) Hamiltonian. The bands need not be two and they need not be equally wide. Our implementation is quite general with respect to the division of Hamiltonian energy spectrum into this kind of bands. In all calculations in this work we however use two equally wide bands because such setup is most practical.
The basis set \( (5) \) provides a linear system of differential equations for each of the involved electrons. Due to the time-evolution of the basis functions given by the operator \( \hat{H}^0 \), the equations for amplitudes \( A_o(t) \) include matrix elements of the perturbation only:

\[
\frac{i\hbar}{\partial t} A_o(t) = \sum_{\sigma' = 1} \langle o; t | \hat{V}(t) | o'; t \rangle A_{o'}(t), \tag{11}
\]

where we have introduced a composite index \( o \equiv (n, \alpha, m) \).

The Schrödinger equation (see equation (11) below), very much in the spirit of quantum theory. At each given time \( t \), the wavefunction \( \Psi(t) \) is propagated by the Schrödinger equation, i.e. the quantities \( |\langle t | n, \alpha, m; t \rangle \rangle \) as functions of the lattice site \( l \). Band index is chosen to be \( n = 1 \), propagation direction \( \alpha = 1 \) and the \( m \) indices run through the range \(-8, -7, \ldots, +8\). The very low value of \( m_{\text{max}} = 8 \) is chosen for convenient visualisation and is actual only for this scheme.

The degeneracy index \( \alpha \). Apart from the energy, each eigenstate of the lead’s Hamiltonian has further quantum numbers. In the present system, the only one is the direction of propagation of the Bloch eigenstates: those not strictly localised, by using the finite cutoff of the left lead; see equation (4). For such matrix elements we have

\[
\langle o; t | \hat{V}(t) | o'; t \rangle \approx eU(t)\delta_{o,o'} \tag{12}
\]

in the left lead, for packets far from the central region meaning that probability amplitudes of the wavepackets evolve freely (independently of other amplitudes) far in the left lead. Situation is even simpler far in the right lead where \( \hat{V}(t) \) is zero and corresponding amplitudes do not evolve at all. The constant or freely evolving amplitudes need not be explicitly included into simulation and this omission in principle does not have any impact on accuracy of the simulation. In practice, since the SWPs are not strictly localised, by using the finite cutoff \( m_{\text{max}} \) we introduce certain error into simulations.

The simple form of equations of motion (11) has been possible due to the employment of the moving (i.e. unitarily propagating) basis set. The simplicity is in that the matrix elements are computed only from the interaction term \( \hat{V}(t) = \hat{H}^1 + \hat{H}^2(t) \) of the total Hamiltonian (1).

Wavepackets corresponding to vectors (5) unitarily propagate in time which implies that each particular packet will travel away from the central region after some time. The result would be that (for finite \( m_{\text{max}} \)) at large times the region would not be covered by basis set wave packets at all and no electrons would be present in the central region at long times. In detail, the unitary propagation of the wavepackets is such that during a time interval \( \tau_n \) (defined below (5)) each packet moves exactly to the position of the neighbouring packet (either on its left or on its right, depending on the propagation direction). If we had a very large basis set (\( m_{\text{max}} \to \infty \)) and were looking at a movie of the wavepackets only at the “stroboscopic” times \( m'\tau_n \), we could not see any motion of the packets. For any finite \( m_{\text{max}} \) we could observe similar static picture only for a limited time and in a limited spatial region because the wavepackets constantly propagate from one’s position to the other’s.

To prevent the gradual disappearance of the basis functions from the central region in simulations, we periodically insert new basis wave packets into the system at every period \( \tau_n \). The newly inserted packets are localised far away from the centre, but moving toward it. To avoid the increase of the total number of basis vectors, we also

\[
|\Psi(t)\rangle = \sum_{n=1}^{N_b} \sum_{\alpha = \pm 1} \sum_{m = -m_{\text{max}}}^{m_{\text{max}}} A_{n,\alpha,m}(t) |n, \alpha, m; t\rangle = \sum_{o = 1}^{2N_b(2m_{\text{max}} + 1)} A_o(t) |o; t\rangle, \tag{10}
\]

Fig. 2. Schematic view on stroboscopic wave packets \( |n, \alpha, m; t\rangle \) at time \( t = 0 \). Actual picture shows absolute values of their projections to atomic orbitals \( |l\rangle \), i.e. the quantities \( |\langle t | n, \alpha, m; t \rangle \rangle \) as functions of the lattice site \( l \). Band index is chosen to be \( n = 1 \), propagation direction \( \alpha = 1 \) and the \( m \) indices run through the range \(-8, -7, \ldots, +8\). The very low value of \( m_{\text{max}} = 8 \) is chosen for convenient visualisation and is actual only for this scheme.

is set by the energy width of the band \( \Delta E_{\alpha} = E_n - E_{n-1} \). \( m \) attains all integer values, but in numerical calculations it is restricted to \( m = -m_{\text{max}}, \ldots, +m_{\text{max}} \), as it is discussed at the end of this section.

The time \( t \). The time \( t \) in the notation indicates that the basis state is unitarily propagated [17] by the lead’s Hamiltonian (2). The use of this “moving” basis set significantly simplifies the form of the Schrödinger equation in the SWB representations (see Eq. (11) below), very much in the spirit of using the interaction representation in formal perturbation theory in quantum theory. At each given time \( t \) the SWB vectors form an orthonormal system since

\[
\langle n, \alpha, m; t | n', \alpha', m'; t \rangle = \delta_{n,n'}\delta_{\alpha,\alpha'}\delta_{m,m'}. \tag{8}
\]

The set of basis states (5) is complete if \( m_{\text{max}} \) is infinite.

Given the form of Hamiltonian (1), each electron in the system evolves independently as described by the Schrödinger equation

\[
\frac{i\hbar}{\partial t} |\Psi(t)\rangle = \hat{H}(t)|\Psi(t)\rangle. \tag{9}
\]

State vectors \( |\Psi(t)\rangle \) for each electron are expanded in the basis set (5):

\[
|\Psi(t)\rangle = \sum_{n=1}^{N_b} \sum_{\alpha = \pm 1} \sum_{m = -m_{\text{max}}}^{m_{\text{max}}} A_{n,\alpha,m}(t) |n, \alpha, m; t\rangle = \sum_{o = 1}^{2N_b(2m_{\text{max}} + 1)} A_o(t) |o; t\rangle, \tag{10}
\]
periodically remove basis wave packets (those which have left the central region). In other words, we remove the leading wave packet from the train of the propagating packets and attach a new wave packet at the tail of the train (see also Fig. 2). This removal/insertion procedure is accomplished for each band (indexed by \( n \)) independently; different bands could in principle have different widths and consequently different parameters \( \tau_n \). For given band \( n \) there are two such procedures: one for \( \alpha = -1 \) (packets propagating from right to left), the other for \( \alpha = +1 \) (packets propagating from left to right). In this way we keep the basis set vectors in the region of interest (the “central region”) and also keep the number of the vectors constant.

It is necessary to have \( m_{\text{max}} \) sufficiently large so that the basis vectors \([n, \alpha, m; t]\) with \( m \approx m_{\text{max}} \) are decoupled from other basis vectors (see more reasoning below) and hence the removal of the basis vector \([n, \alpha, m_{\text{max}}; t]\) does not affect the quantum dynamics of the electron, amplitude of which has been removed.

In addition to the basis vector removal/insertion procedure, we also track individual electrons in the simulation. Each newly inserted basis state, if belonging to normally occupied band (\( n = 1 \) in this work), is set to be occupied at the instance of the insertion. Also, an electron, which through time evolution gets far from the central region, is removed from the simulation according to a proper criterion. Therefore the simulation explicitly describes an open system in which the total number of electrons generally fluctuates\(^2\).

The initial conditions applied to the probability amplitudes are such that

\[
A_{n,\alpha,m}(0) = \begin{cases} 
1, & \text{for } n = 1 \\
0, & \text{for } n = 2
\end{cases} \quad \text{for all } \alpha, m, \tag{13}
\]

e.g. only the lower band is initially occupied.

Matrix elements of \( \hat{V}(t) \) are evaluated using the overlaps \( \langle l|\alpha; t \rangle \). In Appendix we derive that the overlaps can be expressed by the formula

\[
\langle l|n, \alpha, m; t \rangle = \sqrt{\frac{|t_n|}{\pi \Delta \xi_n}} \exp \left[ -\frac{i}{\hbar} \left( m \tau_n + t \right) \right] \times \int_{K_n^{-1}} \sqrt{\sin K} \exp \left[ i \left( \alpha K l - \frac{2|t_n|}{\hbar} (m \tau_n + t) \cos K \right) \right] dK. \tag{14}
\]

Quantities \( K_n \equiv k_n a \) are dimensionless wavenumbers corresponding to energies \( \xi_n \) through the TB dispersion relation (6). The cutoff \( l_{\text{max}} \) on atomic sites indexed by \( l \) has to be chosen sufficiently large in order to cover all SWPs (5) included into simulation. For majority of calculations we use \( m_{\text{max}} = 352 \) with rather conservatively chosen \( l_{\text{max}} = 3016 \). In one case we use \( m_{\text{max}} = 1200 \) and corresponding \( l_{\text{max}} = 7576 \). Integrals in (14) have to be evaluated numerically.

The SWPA as described above and used in this work does not include any electron-electron (e-e) interactions except for keeping them always in strictly orthogonal states. We expect that e-e interactions with effects like Coulomb blockade among others become more important for weakly coupled rings (although they can never be neglected in any accurate quantitative description)\(^3\). Further developments of the SWPA will include also realistic e-e interactions at least on a mean-field level of description like in time-dependent Hartree-Fock theory or in TDDFT. Presently we could only include a simple dynamical mean-field interaction according to a chosen model. Having done this we have not found any significant interesting impacts of the interaction. Hence we decided not to include any such results in the present work as it would present an unnecessary complication.

The system of the equations (11) is solved using the modified-midpoint method \([38]\). Its accuracy and stability is fully sufficient for given problem. As a complementary treatment applicable for stationary currents, we compute exact stationary currents for given TB Hamiltonian (1) in cases when it is time-independent; see Section 5 for more details.

### 4 Electron current formula

In this section we briefly provide an expression for local electron current. For this purpose we consider one-dimensional infinite TB chain, specified in Section 2. For such a system the local current operator at bond between sites \( l_0 \) and \( l_0 + 1 \) is given by the formula (see Ref. [1], p. 162 therein):

\[
\hat{I}_{l_0,l_0+1} = \frac{1}{i\hbar} \left( H_{l_0+1,l_0} a_{s,l_0+1} a_{s,l_0}^\dagger - H_{l_0,l_0+1} a_{s,l_0}^\dagger a_{s,l_0+1} \right) \tag{15}
\]

with \( s = \pm \frac{1}{2} \) being the spin index. Taking the expectation value in a single-electron state \( |\Psi(t)\rangle \) we obtain

\[
I^{(1)}_{l_0,l_0+1}(t) = \frac{1}{i\hbar} H_{l_0+1,l_0}(t) \langle \Psi(t) | l_0+1 \rangle \langle l_0 | \Psi(t) \rangle + \text{c.c.} \tag{16}
\]

Superscript (1) marks that it is a current caused by single electron; we must sum up over all electrons in the system to obtain the total current. If we express the state vector in the stroboscopic basis orbitals then we obtain

\[
I^{(1)}_{l_0}(t) = \frac{2}{i\hbar} H_{l_0+1,l_0}(t) \left[ \sum_{|\alpha|} A_{\alpha}^*(t) \langle \alpha; l_0+1 | \right] 
\times \left[ \sum_{|\alpha'|=1} \infty \right] A_{\alpha'}(t) \langle l_0 | \alpha' \rangle + \text{c.c.} \tag{17}
\]

\(^3\) Studies of e-e interactions in connection with quantum interference effects have been published; see for example [37].
The factor of 2 has been added to take into account two electrons differing only by their spins. As for the sign convention used for the current expressed by equations (16), (17), it indicates flow of particles (electrons), not the charge. The same purely local result for $I_{t_0}^{(1)}(t)$ as shown above could be derived also for currents in particular ring arms. Hence we use formula (17) to compute time-dependent current (contribution from one electron) through any chain (lead or arm of the ring) of the complete system. The total current is obtained by summing up contributions (17) generated by all individual explicitly included electrons in the system:

$$I_{t_0}(t) = \sum_{\text{ele}=1}^{N_{\text{ele}}} I_{t_0}^{(\text{ele})}(t). \quad (18)$$

5 Stationary currents through ring nanojunctions

Currents through TB rings have been studied in literature mostly in stationary regimes. It is convenient to compare quasi-stationary currents from our method to exact stationary results for the TB model. For this purpose, the whole system - ring with leads - is assumed to be composed of identical atoms described by the simple TB Hamiltonian with all couplings equal to $t_B$. The only perturbation to periodic chain Hamiltonian $\hat{H}^0$ are then terms (3) which represent a perturbation to the topology of the linear chain. It was shown in reference [21] that in the stationary regime there are both conducting and insulating configurations depending on where the leads are attached to the ring. Specifically, all odd-numbered rings are always conductive irrespective of the choice of attachment site. On the other hand, even-numbered rings can exhibit both behaviours. These findings are confirmed and extended by the exact TB results of reference [23].

We have obtained stationary results from exact eigenstates of the TB Hamiltonian $\hat{H}^0 + \hat{H}^1 + \hat{H}^2$ defined in Section 2. This approach is, for the Hamiltonian used in our work, equivalent to the method described in reference [23]. As specified in Section 2, the applied bias $U$ is modelled by lifting on-site energies in the left (source) lead at the value $\epsilon_L = \epsilon + eU$. The on-site energies in the right lead (drain) are kept at $\epsilon_R = \epsilon$. On-site energies of all ring atoms are kept at the intermediate value $\epsilon_a = \epsilon + eU/2 = (\epsilon_L + \epsilon_R)/2$. TB couplings are unchanged, i.e. they all are equal to $t_B$. The eigenstates for such system are expressed in the TB basis,

$$|\psi\rangle = \sum_{l=-\infty}^{\infty} \psi_l |l\rangle. \quad (19)$$

They depend on the eigenenergy $E$ and the direction of propagation $\alpha$. For brevity we do not show these dependences explicitly. We compactly represent the amplitudes by composite formula

$$\psi_l = \begin{cases} \mathcal{P} f(K_S l) + \mathcal{Q} f(-K_S l) \\ \mathcal{A} f(K_L l) + \mathcal{B} f(-K_L l) \\ \mathcal{F} f(K_S l) + \mathcal{G} f(-K_S l) \end{cases} \quad (20)$$

with

$$f(K) = \frac{\rho(E)}{\sqrt{2\pi}} e^{iKl}. \quad (21)$$

The specific form of (20) is an abbreviated non-standard notation mimicking the spatial positions of particular system chains and has to be understood in the sense that $\psi_l = \mathcal{A} f(K_L l) + \mathcal{B} f(-K_L l)$ for $l \in$ left lead, $\psi_l = \mathcal{F} f(K_S l) + \mathcal{G} f(-K_S l)$ for $l \in$ lower ring branch, etc. We introduced the dimensionless $k$-number

$$K \equiv k a \quad (22)$$

with $a$ being the lattice constant. $\rho(E)$ is the local density of states (DOS) of an ideal infinite chain for a particular branch of the system and for the TB model is expressed as

$$\rho(E) = \frac{1}{\sqrt{4t_B^2 - (E-\epsilon)^2}}. \quad (23)$$

$\epsilon$, identified with $\epsilon_L$, $\epsilon_R$ or $\epsilon_a$, is the value of the on-site energies in given branch of the system. In the model under study it also represents Fermi energies of particular bulk systems. Indices $L$, $S$ and $R$ stand for the left lead, small system (the ring) and the right lead, respectively. When convenient we use also notations $\rho_L$, $\rho_R$ and $\rho_S$ to distinguish DOSs in the left lead, right lead and in the small system (ring). Parameters $A$ and $B$ describe wavefunction of the left lead, parameter $C$ in the right lead, $\mathcal{F}$ and $\mathcal{G}$ in the lower ring arm and finally $\mathcal{P}$ and $\mathcal{Q}$ in the upper ring arm.

An electron being in particular eigenstate $|\psi\rangle$ is then interpreted as partially reflected and partially transmitted, with transmittance computed as

$$T(E) = \left| \frac{C}{A} \right|^2. \quad (24)$$

This simple formula is applicable also in cases when the Fermi levels in the two leads are different. In such a case we need additional factors involving the respective group velocities. These factors have been explicitly inserted into functions (21) hence are not explicitly present in the formula (24). The results from exact eigenstates for the ring of $N = 16$ sites and $n = 7$ are shown in Figure 3.

Having transmittances available, we compute the stationary electron currents in the leads using the Landauer formula

$$I = \frac{2e}{h} \int_{E_F}^{E_F + U} T(E) dE. \quad (25)$$

For example, we have $\mathcal{F} = \epsilon_a + 2t_B \cos K_L$ in the left lead and $\mathcal{E} = \epsilon_a + 2t_B \cos K_S$ in the ring.
The plots with higher \( \epsilon \) values are vertically shifted for convenience and the shifts are visually enhanced by horizontal dashed lines. Parameters \( \epsilon \) are on-site energies in the left lead. Right leads have always set \( \epsilon = 0 \). The small system (ring) has \( \epsilon = (\epsilon_L + \epsilon_R)/2 \). The transmittances are computed and shown only for energies at which extended propagating eigenstates exist. \( t_B < 0 \) is the tight-binding hopping parameter, magnitude of which is used as the unit of energy throughout this work.

We remind that the transmittance \( T(E) \) depends parametrically also on the chemical potentials in the leads (which are equal to \( \epsilon_L \) and \( \epsilon_R \)) and on the on-site energies of the ring atoms which are all equal to \( \epsilon \). In the eigenstate approach we always use \( \epsilon_r = 0 \) and \( \epsilon = eU \) (applied bias) and the on-site energies of the ring are set to \( \epsilon = (\epsilon_L + \epsilon_R)/2 \). The integral in equation (25) is done numerically.

In Figure 4 we show the comparison of the exact stationary currents and the currents obtained by the time-dependent wavepacket approach. Results from the SWPA (discrete symbols) represent long-time quasi-stationary values from time-dependent calculations. We see overall agreement between the two approaches, especially for low biases. The results from eigenstates are exact for given Hamiltonian. The SWPA [16,17] is in principle exact too, but its accuracy is lowered by the finite cutoff to the number of basis functions which is \( 2N_{tb}(2m_{\text{max}} + 1) \). The impact of the finite cutoff is typically not important at low biases but increases with the bias and the obtained accuracy is then lower at high bias conditions. Convergence of the current with the cutoff becomes very slow at large \( m_{\text{max}} \). Several examples computed with different basis set sizes are shown in Figure 4. The steady-state current in the leads is always underestimated\(^5\) when finite \( m_{\text{max}} \) is used. To understand the finite basis set impact, we first remark that the dynamics of an individual electron becomes (quite obviously) more accurately described when using larger \( m_{\text{max}} \), hence providing more accurate current (17) generated by single electron. Second, the increased \( m_{\text{max}} \) results in a larger number of electrons \( N_{\text{ele}} \) included into the total current formula (18). As a consequence, the precise dependence of the current on \( m_{\text{max}} \) is a complicated function. The much slower convergence at higher values of \( m_{\text{max}} \) can qualitatively be understood as a result of larger spread of the SWPs at high \( m \) (shown in Fig. 2). In the limit of \( m \to \infty \) the packets become fully delocalised. Individual high-\( m \) packets contribute very weakly to the total local current at given point in space. The less converged results in absolute terms are pronounced especially at larger biases. The convergence is worse also in relative terms: at \( U = 0.2 |t_B|/e \) we reach 95.5% of the exact limit (Fig. 4). At \( U \approx 0.5 \) we get 91.6% and at \( U = 1.8 \) only 89.3%, all at \( m_{\text{max}} = 352 \). The worse convergence at larger biases can be qualitatively understood on the basis of the equations of motion (11) and bias-related matrix elements (12). Using a finite \( m_{\text{max}} \) in (11) we drop some portion of the exact equations of motion, in particular some of the terms (12) which describe the interaction with bias-induced lifts of on-site energies in the left lead. Using higher bias increases significance of those matrix elements and hence makes convergence more difficult. The finite basis set size would not impact the accuracy of our approach if the stroboscopic basis states with large \( |m| \) indices were well localised in space. In reality, even wavepackets with very large \( |m| \) indices exhibit long

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\(^5\) Inspection of the currents through branches of unsymmetrical rings shows that the situation is more complicated inside such rings. See Figure 8 as an example.
tails towards the central point of the lattice where the nanojunction is placed. Ideally, having basis wavepackets (those with large $|m|$ indices) without tails in the nanojunction region would result in perfectly converged results (such an ideal situation is impossible because of the dispersion which spreads the wavepackets). Our further effort in development of the SWPA is directed to overcome the convergence issues despite the presence of the dispersion.

6 Simulations

6.1 Bias voltage switch on ring-shaped nanojunction

Time-dependent voltage bias is modelled by given time-dependent (but spatially uniform) lift of the TB on-site energies in the left lead only. The atoms in the right lead have their on-site energies all set to zero, which is also the value of Fermi energy in equilibrium. The atoms within the ring have their on-site energies set to the half of the bias value applied at given time although we test also different model in Section 6.1.2. In our simulations we first "equilibrate" given system by letting it evolve at zero bias (and zero temperature) for time up to $t_{sw} = 500 \hbar/|t_B|$. The "equilibration" evolves the non-interacting many-electron initial state of the system, described by equation (13), into (again non-interacting) stationary many-electron state of the Hamiltonian with the localised perturbation (the ring system and eventual variations of some of the on-site energies or interatomic couplings). We have verified that the equilibration period is sufficient to obtain converged transient electric currents in the sense that these currents are practically independent of particular choice of $t_{sw}$ if $t_{sw}$ is at least $500 \hbar/|t_B|$. The transient electron currents are evaluated according to formula (17), with applied summation over all contributing electrons in the system. We study how these transient currents depend on several ring parameters like the ring size, placement of the terminals and the magnitude of applied bias. Typical behaviour of the transient electron currents is shown in Figure 5. It is noticeable that both the temporal and spatial evolution must be considered. The intersect of the two ridges corresponds to the time when the bias was turned on and to the spatial location of the ring. The slope of the ridge corresponds to the Fermi velocity, $v_F = \frac{1}{\hbar} \partial_N E(k = \pi/a) = -2at_B/\hbar$, so that the time interval between the instance of switching-on the bias and the arrival of the density or current perturbation to a site $l$ is given by the expression $t_{l} = l \pi / v_F$. The ridge on the left side corresponds to the reflected electron impulse propagating back into the left lead. Similarly, the other ridge represents the transmitted wavefront in the right lead. The function values are mostly positive meaning that the currents in both leads are positive (i.e. electrons flow from the source lead to the drain lead). Results for several specific cases are discussed in the following subsections. Presented ring sizes $N$ vary from 16 to 18. We have calculated also several other sizes, starting from $N = 5$, not shown here. Plotted local currents will typically correspond to a lattice site positioned far from the ring, in most cases site $l = 120$, which is displaced about 100 lattice constants from the considered rings.

6.1.1 Dependence on the location of drain vertex

The dependence of the time-dependent current on the position of the right (drain) lead is shown in Figure 6. We show how and if the transient currents (induced by the abrupt bias switch at time $t_{sw} = 500 \hbar/|t_B|$) depend on chosen drain vertex site $n$. In Figure 6 we plot transient local currents obtained from our simulations for three subsequent rings sizes and several drain-lead attachments. These rings represent various different kinds of electronic transport properties which can be found in rings. The local currents shown have been calculated at lattice site 120.
The finite basis set adds rapid oscillations on the computed local currents\(^6\). We smooth out the oscillations by averaging them over the time interval of about \(\tau\). This averaging does not modify essential features of the plots including the characteristic times and rates. Example of the raw unaveraged data are shown in Figure 7. The “no slope” black curves in that figure are obtained in the same system as plots \(n = 7\) and \(n = 8\) in Figure 6, apart from the time averaging.

The onset of the electronic responses appears at times around 550 \(h/|t_B|\), which is delayed about 50 time units after \(t_{sw} = 500 h/|t_B|\). Basis set size again uses \(m_{max} = 352\). The plots have been smoothed by taking running averages over an interval of about \(\pi h/|t_B|\). The dashed lines represent analytically computed stationary currents as obtained by the exact-eigenstate method described in Section 5.

Fig. 6. Transient currents computed in the drain lead at site \(l = 120\) for several different drain lead attachments (vertices specified by the numbers \(n = 5, 6, 7, 8\) shown at individual plots) of the rings composed of \(N = 16, 17, 18\) atoms. In all cases the applied bias \(U = 0.5 |t_B|/e\) has been switched on abruptly at time \(t_{sw} = 500 h/|t_B|\). Basis set size again uses \(m_{max} = 352\). The plots have been smoothed by taking running averages over an interval of about \(\pi h/|t_B|\). The dashed lines represent analytically computed stationary currents as obtained by the exact-eigenstate method described in Section 5.

\(^6\) The period of these artificial oscillations is always \(\tau\), the quantity defined by equation (7), here equal to \(\pi h/|t_B|\) (same for both the bands). The oscillations are larger at sites closer to the ring. This is explained by the localised nature of the stroboscopic basis states (the wavepackets) and the finite number of them. The train of the wavepackets (see Fig. 2) travels in real space in such a way that during a time interval \([t, t + \tau]\) each packet places itself exactly to the position which was occupied by its neighbour at time \(t\). Since we use the finite cutoff on the basis set, the quality of the description at given point of space fluctuates in time with the period of \(\tau\). Instead of using a huge value of \(m_{max}\) (which would be prohibitively expensive) we smooth out the oscillations by averaging them over the time interval of about \(\tau\). Typical timescales of the processes studied in the present work are in most cases much larger than \(\tau\) and the relevant effects are not affected by the oscillations or by their artificial smoothening.

configurations after the bias is switched on. This feature is shown in the lower panel of Figure 5 and will be discussed also below.

\(N = 16\) rings. The left panel of Figure 6 shows results for rings of size \(N = 16\), but differing by the position of the drain vertex sites which run through the sequence \(n = 5, \ldots, 8\). The odd-numbered cases \((n = 5\) and \(n = 7\)) correspond to conducting configurations. The well conducting state of the ring is given by constructive interference (CI) of the electron amplitudes in the two ring branches. CI are most significant in the case of symmetrically attached ring \(N = 16, n = 9\), the configuration with equally long branches (not shown in the figure). As we can see from the figure, and in agreement with former theoretical analysis \([21,23]\), significant CI is possible for several vertex configurations. The two local currents \((n = 5\) and \(n = 7\)) reach similar long-time limits and the time needed to build up the current is the same for each drain vertex site: \(\Delta t \approx 10 h/|t_B|\). The dashed lines in Figure 6 represent stationary currents as obtained from the exact-eigenstate approach. At intermediate times, around 600 time units, the dynamically computed currents are in good agreement with the stationary approach. The agreement would become slightly worse at larger times when finite basis set errors take effect; see also Figure 4 for comparisons of stationary currents also at other biases. On the other hand there are the even-numbered cases which show almost zero currents after a short transient effect. The peaks of the transient currents (the plots with \(n = 6\) and \(n = 8\)) are very similar each other. The blocking status is again due to quantum interference, now the destructive one. Results for other values of \(n\), not shown in plots, also confirm that the transient characteristics are practically the same for all drain vertex attachments within the particular group (conductive or insulating).

\(N = 17\) rings. Central panel of Figure 6 shows results for rings with size \(N = 17\). In this case there is no such a distinct separation into conductive and insulating configurations, the \(N = 17\) rings are all conductive. Neither constructive nor destructive interferences are now perfect. However, similarly as in the insulating configurations and contrary to the conducting configurations of even-sized rings, we now observe peaks in transient currents. In addition there is a substantially longer relaxation, now lasting for about 100 time units. The peak current depends on the drain vertex site in an oscillatory manner (more DC-conductive rings in this class exhibit lower peak currents). \(N = 17\) rings have shown to be more difficult from computational point of view. The effect of the incomplete basis set would show up at long-time stationary currents which would be, by estimate, 10–25% lower compared to exact results from eigenstates.

\(N = 18\) rings. In this case (right panel of Fig. 6) we again obtain two distinctly different behaviours. The difference from the \(N = 16\) ring is that now the odd-numbered drain vertices correspond to insulating configurations. The long-time limit of the time-dependent treatment would relate to exact stationary results similarly as for \(N = 17\) rings. In addition, the ring with 18 atoms
vertex being either at site \( n = 7 \) (conducting configuration) or at site \( n = 8 \) (insulating configuration). As we can see the differences between the spatially uniform and the spatially varying model are essentially negligible for both conducting and insulating configurations. This finding may not be universally valid. However, to keep the models in the present work simple, we stick at the spatially uniform profile of on-site energies within the studied rings, with obvious exceptions of an applied gate potential (see Sect. 6.5).

### 6.2 Circulating currents

It has been discussed in several studies (e.g. [21]) that circulating currents (CCs) may arise in certain ring configurations. These analyses were done for stationary currents. In this subsection we study circulating currents in transient regime upon the abrupt switch of the bias. By definition, a CC at given instant of time in a ring structure (like that in Fig. 1) exists when the currents in the two ring branches have opposite directions in the sense that, for example, the electrons in the shorter branch flow from the left terminal to the right one while the electrons in the longer branch flow from the right terminal to the left one. The currents in the ring branches are calculated using formulae (18) and (17). These formulae represent strictly local currents: \( I_0(t) \) is in fact the current between sites \( l_0 \) and \( l_0+1 \), i.e. a current through the bond. Our calculation of the current in given ring branch in addition involves averaging over the interatomic bonds of given branch.

Inspection of the currents calculated from exact transmittances of reference [23] shows that stationary CCs are possible for many ring sizes. Actual occurrence of the CCs depends also on chosen drain site and on applied bias. Of the studied rings we found most pronounced CC in \( N = 18 \) ring with its drain vertex at site \( n = 3 \) and \( n = 5 \). The results for the \( n = 5 \) structure are shown in Figure 8.

Upper panel in Figure 8 shows the exact stationary electron currents, calculated with the approach described in [23] (lines), alongside with the quasi-stationary values from our simulations computed with the basis-set parameter \( m_{\text{max}} = 1200 \) (isolated symbols)\(^7\). Red (dark grey solid line) and green (light grey dashed line) plots display currents in the two ring branches and are relevant in verifying if CC is present. The sign convention used on the figure is such that currents in both branches have positive signs for the flow of the particles from left to right, i.e. state without CCs. The negative value of one of the currents indicates the presence of the CC in the ring. From the upper panel of Figure 8 we see that such a negative electron current flows in stationary regime in the longer ring branch for a wide range of voltages (0 to 0.78 \( |t_B|/e \)). Most pronounced CC is found at a bias of \( U = 0.694 |t_B|/e \).

Lower panel of Figure 8 extends the results at the bias 0.694 \( |t_B|/e \) into the non-stationary regime. The non-stationarity is due to the abrupt bias switch at time \( t = 0 \).

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\(^7\) Here we could equivalently use our exact-eigenstate method described in Section 5. We instead used formulas of reference [23] because of their convenience.
Finally, the dip in the right-lead current (black plot on the lower panel of Fig. 8) at times ranging from about 630 to 730 \(h/|t_B|\) is caused by the numerical cutoff on the stroboscopic basis set size. This unphysical dip tends to be more pronounced in certain ring configuration, for certain quantities and for high biases. Ring configuration \((N, n) = (18, 5)\) is significantly affected by the finite basis set error at certain time interval. This is also the reason why we used parameter \(m_{\text{max}} = 1200\) to obtain results shown in Figure 8 while for most of other figures we use \(m_{\text{max}} = 352\). The increased cutoff improves accuracy of time-dependences especially at intermediate times.

6.3 Time-dependent currents controlled by gate field

Quantum interference effects between electrons in the two ring arms can be controlled by an applied electric field. The field value can be time-dependent in general. In our approach it is modelled by a spatially uniform lift of on-site energies in one of the branches of the ring (not including the two terminal atoms); the affected atoms have indices \(n + 1, n + 2, \ldots, N\) in the indexing scheme of Figure 1.

6.3.1 Abrupt gate turn-on

In our first studied application of the gate potential, we control the electron transport using an instantaneously switched gate field which is applied to the two \(N = 16\) rings differing by their drain terminal indices \(n\). The field is turned on at time \(t_g = 800 h/|t_B|\), after the system has been evolving under the constant bias of \(U = 0.5 |t_B|/e\) so that it is essentially in a stationary-current regime before the time when the gate field is lifted. The simulations give results which are depicted in Figure 9. The local current is evaluated at the lattice site 120. We observe that the gate field induces transient effects lasting for about 10–40 time units. The transient effects depend rather strongly on particular bias values. The application of the higher gate field is interesting; it would allow the ring to work as a field effect transistor.

6.3.2 Harmonic gate potential

Another option to control the current flow is to employ a harmonically oscillating gate potential. For this
purpose we decided to use larger rings with sizes $N$ ranging from 70 to 90. Such rings provide longer characteristic times of temporal changes. We study the following three $(N, n)$ configurations: $(70, 61)$, $(80, 71)$, and $(90, 81)$. All the three rings thus have one longer and one shorter branch. The gate field is again applied to atoms with indices in the range $n+1, \ldots, N$, i.e. the atoms of the shorter branch now. The constant bias is again $U = 0.5 |t_B| / e$. The magnitude of the sinusoidal gate potential is always $V_{g0} = 0.25 |t_B| / e$. The potential is harmonic with various angular frequencies as shown in Figure 10. The potential starts to act at time $t_g = 800 \hbar / |t_B|$. The inset of Figure 10 shows typical time dependencies of the current in leads as obtained for the case $(N, n) = (80, 71)$. The three plots of the inset have been recorded for three different gate angular frequencies $\omega_g$: 0.08 (dark blue, solid line), 0.12 (brown, solid line) and 0.16 of $|t_B| / \hbar$ (orange, dashed line). The corresponding periods $T_g = 2 \pi / \omega_g$ are approximately 78.5, 52.4, and 39.3 of $\hbar / |t_B|$. The current response is generally quasi-periodic but anharmonic (the initial transient effect after the gate is turned on is not discussed here). The temporal dependence of the current is given by an interplay of the harmonic gate potential and internal ring effects given also by its size. At low gate frequencies $\omega_g$ (for example 0.08 $|t_B| / \hbar$ or less, see the blue plot, dark solid line, in the inset), the current oscillations typically (not always) exhibit periodic quasi-harmonic pattern at the twice of the gate frequency. The doubled frequency arises from the symmetric dependence of the system transmission on the gate potential $V_g$ and $-V_g$ have the same effect on stationary currents as could be seen from the analytic formulae of Ref. [23] or calculated by formalism of Sect. 5. Rapid driving is not followed by the current in this sense as can be seen from the inset. For example, the gate field with $\omega_g = 0.16 |t_B| / \hbar$ (the orange plot with dashed line on the figure) or at higher gate frequencies results in current oscillations at the same frequency. Intermediate driving frequencies (e.g. 0.12 $|t_B| / \hbar$, the brown plot, solid line, on the figure) yield periodic but anharmonic time dependences.

It is interesting to compute time averages from the oscillating lead currents. The three plots in the main graph of Figure 10 show that the average current for given ring structure depends on the gate frequency (while the bias and the gate amplitude are kept unchanged). The maxima and minima of the average currents vary with varying ring structure: larger rings have the extrema shifted towards lower frequencies. Inspection of the plots show that these variations fulfill the law $\delta \omega / \omega = -6N / N$ which is expected from elementary considerations about resonant frequencies. In this way we have an evidence that the oscillatory pattern of the average current plotted in Figure 10 arises from the internal ring resonances. Given the nanometer-scale size of such ring structures, the characteristic resonant frequencies lie in the optical domain and we do not investigate higher gate frequencies that those shown in the graph.
Electric current dynamics could be investigated also for a fixed ring size \( N \) and varying drain terminal \( n \). However, our inspection has shown that the dynamics is more interesting (i.e. the average currents exhibit more pronounced oscillations as functions of \( \omega_B \)) when the source and drain terminal are relatively close to each other.

### 6.4 Reduced coupling to the leads

Through previous sections it was assumed that all nearest neighbour couplings were identical along the whole composed system, i.e. including the small system (the ring). The couplings between the leads and the ring were then quantified by the hopping parameter \( t_B \). As in previously shown results, bias \( U = 0.5 |t_B|/e \) is abruptly turned on at time \( t_{sw} = 500 \, h/|t_B| \). Number of ring atoms is \( N = 16 \) in both panels and drain is attached to sites \( n = 7 \) (left panel) and \( n = 8 \) (right panel). The dotted lines in the left panel (conducting configuration) show stationary currents computed from the exact-eigenstate approach.

![Fig. 11. Local electron currents evaluated at site 120 for various lead-system coupling strengths \( t_{BS} \). Legends indicate values of \( t_{BS} \) relative to the magnitude of the lead’s hopping parameter \( t_B \). As in previously shown results, bias \( U = 0.5 |t_B|/e \) is abruptly turned on at time \( t_{sw} = 500 \, h/|t_B| \). Number of ring atoms is \( N = 16 \) in both panels and drain is attached to sites \( n = 7 \) (left panel) and \( n = 8 \) (right panel). The dotted lines in the left panel (conducting configuration) show stationary currents computed from the exact-eigenstate approach.](image)

7 Conclusions

Recently proposed stroboscopic wave packets \[16,17\] have been developed to be applicable to systems employing explicit atomistic level of modelling. Time-dependent transport of electrons in an open system with localised perturbation was studied. The localised perturbation had a ring structure described in the tight-binding approximation. Such a system is of interest due to quantum interference effects that affect its transport properties. It might serve as a field driven quantum interference current switching device \[36\] – a nanoscale-sized transistor. We have demonstrated the potential of our newly developed method based on the unitarily propagating stroboscopic wave packets to describe open systems with fluctuating number of explicitly included electrons while the whole system has an infinite number of electrons which can be neglected in a good approximation. In our method full quantum coherence is preserved throughout the whole infinite system. The method can be used for systems with one-dimensional semi-infinite leads. It is capable of spatially resolved description of transient effects like those caused by an abrupt bias switch of a gate field application. However, its most useful application would be found in systems which are exposed to long-lasting varying external fields or biases and for which it is important to preserve quantum coherence in the description. For short-time simulations one could use a less expensive model with cyclic boundary conditions. However, such an approach would become prohibitively expensive for large simulation times as it would require to consider many explicit atoms to describe the leads. In contrast, our method does not have any such limit on simulation time. The weakness of the present implementation of the stroboscopic basis set description is slow convergence of relevant results with increasing number of basis functions. The finite basis set demonstrates itself in a transient unphysical drop of lead current in some of the studied systems. Depending on the studied configuration, long-time stationary currents may also sometime be significantly underestimated. Other consequence of the finite stroboscopic basis set are small artificial rapid oscillations in computed quantities which can however be smoothed.
out and usually does not prevent us from capturing relevant physical time-dependent effects. In our ongoing work we will consider a generalisation of the basis set in order to reduce the finite-basis set errors and to reduce the number of basis states needed.

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Appendix: Derivation of the \( \langle l| n, \alpha, m; t \rangle \) overlaps

Here we derive formula (14) for the overlaps between the TB atomic orbitals \(|l⟩\) and the SWPs \(|n, \alpha, m; t⟩\). In the SWPA [16,17], the eigenstates \(|\mathcal{E}, \alpha⟩\) of the basis-generating Hamiltonian \(\hat{H}^0\) (given by Eq. (2) in this work) use the energy “normalisation” condition which is

\[
\langle \mathcal{E}, \alpha | \mathcal{E}', \alpha' \rangle = \delta(\mathcal{E} - \mathcal{E}') \delta_{\alpha, \alpha'}; \quad (A.1)
\]

see also Section 3 and equation (5) therein. This condition leads to the form

\[
|\mathcal{E}, \alpha⟩ = \sqrt{\frac{1}{2\pi}} |k, \alpha⟩ \quad (A.2)
\]

where \(|k, \alpha⟩\) is the usual Bloch wave in one dimension with the \(k\)-number of magnitude \(k\) and the propagation direction labelled by \(\alpha = \pm 1\). Normalisation of the Bloch waves is assumed to be

\[
\langle k, \alpha | k', \alpha' \rangle = \delta(k - k') \delta_{\alpha, \alpha'} \quad (A.3)
\]

(the \(k\)-numbers will be restricted to the interval \(k = [0, \pi/a]\) where \(a\) is the lattice constant). In case of the TB model with the set of the orthonormal atomic orbitals \(|l⟩\) (one orbital per atom) we obtain formula

\[
|\mathcal{E}, \alpha⟩ = \sqrt{\frac{1}{2\pi}} |l⟩ \sin K \sqrt{\frac{1}{2\pi} \sum_{-\infty}^{\infty} e^{i\alpha\kappa l}} |l⟩ \quad (A.4)
\]

with the summation running over all lattice sites. \(K \equiv ka\) is the dimensionless wavenumber. The eigenstates \(|\mathcal{E}, \alpha⟩\) are used to construct the SWPs according to equation (5). Now we use that formula and write down the overlaps in the form

\[
\langle l| n, \alpha, m; t⟩ = \frac{1}{\sqrt{2\pi}} \int_{\mathcal{E}_{-1}}^{\mathcal{E}_n} \exp \left[-\frac{i}{\hbar} (m\tau_n + t)\mathcal{E}\right] \langle l| \mathcal{E}, \alpha⟩ d\mathcal{E} \quad (A.5)
\]

where we have utilised the equation

\[
\hat{H}^0|\mathcal{E}, \alpha⟩ = \mathcal{E}|\mathcal{E}, \alpha⟩. \quad (A.6)
\]

Using the expression (A.4) for the eigenstates and substituting it into equation (A.5) leads to an integral expression with integration variable \(|\mathcal{E}, \alpha⟩\). With the aid of the TB dispersion relation (6), the integral can be transformed to the integration variable \(K\). The final form of the expression for the overlap \(|l| n, \alpha, m; t⟩\) is then provided by formula (14). We evaluate these integrals numerically.

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