Stroboscopic Wave-Packet Description of Nonequilibrium Many-Electron Problems

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We introduce the construction of an orthogonal wave-packet basis set, using the concept of stroboscopic time propagation, tailored to the efficient description of nonequilibrium extended electronic systems. Thanks to three desirable properties of this basis, significant insight is provided into nonequilibrium processes (both time-dependent and steady-state), and reliable physical estimates of various many-electron quantities such as density, current, and spin polarization can be obtained. Use of the wave-packet basis provides new results for time-dependent switching-on of the bias in quantum transport, and for current-induced spin accumulation at the edge of a 2D doped semiconductor caused by edge-induced spin-orbit interaction.

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Wave packets (WP) are a very useful concept when analyzing quantum-mechanical scattering processes, since they combine local and wavelike aspects on an equal footing. Some of their more recent applications range from studies of the intrinsic spin Hall effect in semiconductors [1,2], spin-flip dynamics [3], thermal averaging, and its influence on interference patterns [4] or transport of an electron through Luttinger liquid [5]. However, the use of traditional WPs in degenerate fermionic systems raises difficulties since the exclusion principle restricts the available eigenstates that are superposed within a single WP. Several orthogonal wave-packet [6–8] and wavelet [9] approaches were put forward in the past to accommodate the exclusion principle; however in contrast to our WPs these do not directly relate to typical many-electron states such as the electronic ground state or moderate perturbations from it at zero temperature. If we forego the time-dependent feature of WPs, the latter problem is conveniently resolved with the introduction of Wannier functions [10,11]: by occupying a finite number of them, we locally recover the exact eigenstates of a system of noninteracting electrons.

In this Letter we combine the advantages of Wannier functions for extended systems with the time-dependent description of WP propagation. This is achieved by generalizing the orthogonal WPs introduced by Martin and Landauer [12] for ideal 1D leads. Our wave-packet basis set (WPB) has the following three properties: (1) each basis function (WP) is localized in space, (2) occupying a subset of the WPB we recover the exact noninteracting many-electron ground state of a reference Hamiltonian, (3) the WPB is generated by time propagation through successive time steps τ of *an initial set* of WPs, according to a reference Hamiltonian.

From the above properties it follows that we can view the whole basis set as *stroboscopic pictures* of a continuous time evolution of a suitably chosen family of *initial* WPs (Fig. 1). Since all WPs are orthonormal, each copy can be occupied by precisely one electron and in time τ each electron will move into its neighboring WP. Similarly, if a single electron is in a superposition of several WPs, in time τ it will be in the *same* superposition but of the WPs obtained from the former by a single shift of the basis functions. This picture is valid as long as the reference Hamiltonian is time independent in the region where the concerned WPs are localized. We will refer to this region as the *bulk* and to the rest—typically a much smaller region—as the *scatterer*. Similarly, the bulk (scattering) WPs are those WPs that are generated with the bulk (bulk + scatterer) Hamiltonian.

To obtain the time-dependent dynamics in the scatterer one needs to perform a full time-dependent simulation of the bulk WPs entering the scatterer. After a certain time, the scattering WPs will return into the bulk where those WPs can once again be expanded into the bulk WPB and propagated as moves of duration τ between the bulk WPs, i.e., analytically. Hence, the WPB offers a very simple interpretation of the processes as well as a convenient framework for numerical time-dependent simulations.

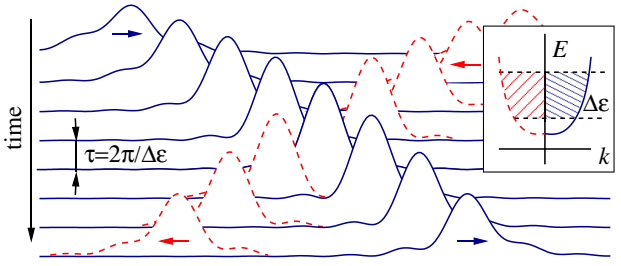

FIG. 1 (color online). Squared amplitudes of orthogonal stroboscopic wave packets obtained by time propagation of the initial WP (in the center) by a constant time-step τ for free electrons in 1D. The right- (blue, or solid) and left-going (red, or dashed) WPs belonging from the same energy band (inset) are shown. These, together with the WPs coming from the bands covering the rest of the spectrum, form a complete orthogonal basis set.

The consistency of the conditions (1) and (3) demands that the reference Hamiltonian possess translational symmetry in the direction of propagation. Its eigenstates in the Bloch form will be sufficient to create a basis such that each WP from the initial set will be spatially localized and their time-propagated WPs will slowly disperse with increasing time. This property can be satisfied only if the reference Hamiltonian is just that of the bulk. We may also construct the WPB for the combined system where the reference Hamiltonian is that of bulk + scatterer, but the scattering will typically result in a strongly delocalized WP with transmitted and reflected components. However, the scattering WPs can be easily expanded into the bulk WPB, a fact of which we will make use later.

Definition of the WPB and its formal properties.—To define the basis set let us take an extended system specified by the reference Hamiltonian $\hat{H}$ with a continuous spectrum of eigenenergies $\epsilon \in (\epsilon_0, \infty)$, $\hat{H}|\epsilon, \alpha\rangle = \epsilon|\epsilon, \alpha\rangle$. To each eigenenergy we will generally have a set of degenerate single-particle eigenstates $|\epsilon, \alpha\rangle$, $\alpha = 1, 2, \ldots, N_\epsilon$, forming all together a complete orthogonal whose normalization we choose such that $\langle\epsilon', \alpha'|\epsilon, \alpha\rangle = \delta(\epsilon - \epsilon')\delta_{\alpha,\alpha'}$.

From the above set we can generate an orthogonal and complete wave-packet basis set (WPB) by first choosing the *initial set* of wave-packets [13]

$$|n, 0, \alpha\rangle = \frac{1}{\sqrt{\Delta\epsilon_n}} \int_{\epsilon_n^\alpha}^{\epsilon_{n+1}^\alpha} d\epsilon' U_{\alpha,\alpha'}(\epsilon')|\epsilon', \alpha'\rangle, \quad n = 0, 1, 2, \ldots \tag{1}$$

for an arbitrarily chosen division of the spectrum into *energy bands* $\{(\epsilon_n^\alpha, \epsilon_{n+1}^\alpha)\}_{n=0}^\infty$, $\alpha = 1, 2, \ldots, N_\epsilon$ with bandwidths $\Delta\epsilon_n^\alpha = \epsilon_{n+1}^\alpha - \epsilon_n^\alpha$. The division into energy bands must cover the full spectrum of $\hat{H}$ but otherwise can be chosen so as to suit the physical situation as discussed later. The unitary, energy-dependent matrix $U_{\alpha\alpha'}(\epsilon)$ represents the second freedom of choice in the construction of the WPB. In this Letter we will use $U_{\alpha\alpha'}(\epsilon) = \delta_{\alpha\alpha'}$ which is satisfactory for our present purposes, but in general it can be used either to adapt the bulk WPB to the scattering processes involved or to improve the localization of the WPs, in analogy with Wannier functions [11]. Any two WPs from different energy band are orthogonal by definition, since they are linear combinations of eigenstates from disjunct energy bands.

The construction of the WPB is completed by forward and backward time propagation of the initial set

$$|n, m, \alpha\rangle = e^{-i\hat{H}m\tau_n}|n, 0, \alpha\rangle, \qquad m = \pm 1, \pm 2, \ldots \tag{2}$$

by regular, band-dependent time steps $\tau_n^\alpha = 2\pi/\Delta\epsilon_n^\alpha$. It is easy to verify that this choice of time step guarantees orthonormality of successive wave packets within each band

$$\langle n, m, \alpha|n, m', \alpha\rangle = \delta_{m,m'}. \tag{3}$$

Because of the orthogonality of the WPs we can uniquely expand any eigenstate of the reference Hamiltonian into the WPB with expansion coefficients $\langle\epsilon, \alpha|n, m, \alpha\rangle = (\Delta\epsilon_n)^{-1/2}\exp\{-i\epsilon m\tau_n\}$, with $\epsilon \in (\epsilon_n, \epsilon_n + \Delta\epsilon_n)$. Conversely, combining this with Eqs. (1) and (2) one obtains that $\sum_m |n, m, \alpha\rangle\langle n, m, \alpha|\epsilon, \alpha\rangle = |\epsilon, \alpha\rangle$, from which follows that the WPB is also *complete* since the original set of eigenstates is a complete one.

It has been already pointed out that the division into energy bands can be exploited to optimize the basis set to the particular physical problem. A typical choice is $\epsilon_n^\alpha = E_F$ for a certain n and all α, where E_F is the Fermi energy of the system. In this way the ground state is described by occupying all of the WPs in the bands below E_F. This means that we need to consider only a few WPs or electrons even though we are describing the *local* ground-state properties of the infinite many-electron system exactly (see [14]). Similarly, the nonequilibrium state is obtained by imposing different effective Fermi energies for WPs with different values of α.

Time-dependent quantum transport.—Understanding the quantum transport of charge through nanojunctions made of individual atoms or molecules will be essential for progress in nanoelectronics. Because of the short spatial scale and short times involved it is clear that transient phenomena play an important role in understanding the functionality of nanodevices. At the same time, it has been recognized that the correct treatment of interactions demands a time-dependent formulation of the density- or current-density functional theory [15–19]. Here we show that the WPB provides physical insight as well as quantitative results for transient times, oscillations, or steady-state current.

Let us first consider a 1D electron gas in which at time $t = 0$ a finite potential difference is applied (Fig. 2). Anticipating the application of the bias ΔV, we choose to split the occupied part of the spectrum of a Hamiltonian for free electrons into occupied bands 0 to ΔV, ΔV to $E_F - \Delta V$, and $E_F - \Delta V$ to E_F (we will refer to the last band as $n = a$, i.e., the *active* band), and two unoccupied bands E_F to $E_F + \Delta V$ and $E_F + \Delta V$ to ∞. The energy-normalized eigenstates are the plane waves $|\epsilon, \alpha\rangle = e^{i\alpha kx}/\sqrt{2\pi k}$, and $\alpha = \pm$ for right- and left-going states, respectively. The resulting WPs $|n, m, \pm\rangle$, obtained according to Eqs. (1) and (2), are examples of the bulk WPs mentioned above and are identical to the WPs employed by Martin and Landauer in their analysis of quantum noise [12]. Because of the hopping of electrons between the WPs in time τ, the current at the position of the mth WP carried by electrons in the *active* band is in general given as $I(t) = N_m(t)/\tau$, where $N_m(t)$ is the occupation of the mth WP.

Switching on the bias ΔV at $x = 0$ and $t = 0$ will energetically align WPs from the highest occupied, band localized in $x < 0$, with the WPs from the lowest unoccupied band localized in $x > 0$ (Fig. 2). A transient phenomenon for time $t \sim 2\pi/E_F \leq \tau = 2\pi/\Delta V$, which needs to be analyzed by performing a time-dependent simulation, will be related to dynamics of those occupied WPs that had for $t < 0$ nonzero amplitude for both $x < 0$ and $x > 0$.

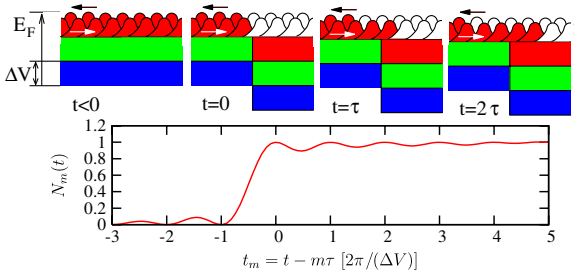

FIG. 2 (color online). Abrupt switching on of the bias in a 1D wire. In response to the bias ΔV, the right-going WPs for $x > 0$ (white, the previously unoccupied band) start to fill the WPs from the left and the occupied left-going WPs for $x < 0$ become empty. The finite extent of each WP causes oscillations, with period τ, of the resulting occupancy $N_m(t)$ and hence the current measured at the fixed position of the mth bulk WP, $x_m = v_F m\tau$ (below).

After that the time-dependent many-electron dynamics for $x > 0$ will result in a train of right-going scattering orthogonal WPs within the *active* band

$$|a, l, +; t) = \int_{E_F-\Delta V}^{E_F} \frac{d\epsilon}{\sqrt{2\pi k \Delta V}} t(k) e^{i(kx-(1/2)k^2(t+l\tau))}, \quad (4)$$

occupied for $l = 0, -1, -2, \ldots$, where $t(k)$ is the transmission amplitude for the applied step potential. The occupation $N_m(t)$ of the mth bulk WP due to this train is $N_m(t) = 2\sum_{l=0}^{-\infty} |(a, l, +; t|a, m, +\rangle|^2$ which, after substituting the above expressions, gives finally

$$I_m(t) = \frac{2}{\tau} \iint_{E_F-\Delta V}^{E_F} d\epsilon d\epsilon' t^*(\epsilon') t(\epsilon) F_{t-m\tau}(\epsilon' - \epsilon), \quad (5)$$

where $F_t(\omega) = (\Delta V)^{-2} \sum_{l=0}^{+\infty} \exp\{-i\omega(l\tau - t)\}$. In fact, this result is equally valid for abrupt switching in 1D wire with an arbitrary scattering potential and it represents a generalization of the well-known Landauer formula for nonlinear time-dependent response to abrupt switching on of a voltage. In the long-time limit we have $F_t(\omega) \to \delta(\omega)/(\Delta V)$ and we recover the nonlinear Landauer formula $I = \int_{\Delta V} |t(\epsilon)|^2 d\epsilon/\pi$.

For the specific case of free electrons, we can put $t(k) = 1$ and perform the integration with the result $N_m(t) = 4\sin^2(\Delta V t/2)/(\Delta V)^2 \sum_{l=0}^{-\infty} \{(l-m)\tau + t\}^{-2}$. In Fig. 2 we show this result, calculated by taking the first 10 terms of this series, i.e., accounting for 10 WPs, for which we get a well-converged answer. The relaxation to the steady-state current is characterized by oscillations with period τ, in agreement with previous numerical calculations [17] based on nonequilibrium Greens functions within a wide band model.

Our treatment here also indicates that the WPB representation can be used to perform numerical *ab initio* time-dependent simulations within the TDDFT framework, i.e., accounting for a time-dependent self-consistent field in the scattering region. The time evolution of the bulk WPs as they enter the scattering region needs to be done numerically, but as soon as the scattered WP leaves this region, by expanding it into a few bulk WPs one can perform its time evolution algebraically in a closed form. The density, current density, or any other many-electron property is obtained by summing contributions from all stroboscopic images of the propagated WPs. While the WPs will typically extend over several atomic distances, relatively few of them will be needed to compute local properties close to the scattering reagion, i.e., for a jellium model of a sodium monoatomic wire with one atom missing is well converged to the exact density of an infinite system with the gap using about 20 occupied WPs [14]. Detailed implementation of the self-consistent mean-field (TDDFT) methodology will be reported elsewhere [21].

Edge-induced spin Hall effect.—It has been recently shown that the interplay between nonzero Rashba-Bytchkov spin-orbit (SO) coupling, the scattering off the edge, and nonzero electric current along this edge leads to a universal spin polarization localized close to the edge of the 2D gas in GaAs quantum wells [22,23]. In parallel, several other authors [24–26] considered the spin-orbit (SO) coupling due to nonzero gradient in potential in-plane, $V_{\rm SO} = -\alpha_E[\hat{\boldsymbol{\sigma}} \times \nabla V(\mathbf{r})] \cdot \hat{\mathbf{p}}$, where α_E is the strength of the SO coupling, $\hat{\boldsymbol{\sigma}}$ is the spin operator, $V(\boldsymbol{r})$ is the confining potential at the edge, and $\hat{\mathbf{p}}$ the momentum operator [27]. The edge-SO scattering, analogous to the mechanism behind impurity scattering in the bulk of the 2D gas, seems to lead to effects similar to the Rashba-Bytchkov mechanism.

Both of these effects can be understood and analyzed within the WPB description, but here we concentrate on the edge-SO scattering. We consider a 2D electron gas confined in the $xy(x > 0)$ half-plane, with its edge being described by a model potential $V(\boldsymbol{r}) = W\theta(-x)$ where θ is the step function. This model is appropriate for typical doping densities $n \sim 10^{12}$ cm^{-2} where the Fermi wavelength $\lambda_F \sim 20$ nm is much larger than atomic spacing, principally determining the abruptness of the edge. The current is imposed in the y direction. Fourier transforming $y \to k_y$, the SO term takes the form $V_{\rm SO} = \alpha_E \hat{\sigma}_z W \delta(x) k_y$, i.e., electrons with up and down spins in the z direction experience different scattering potential at the edge. For each k_y we construct a WP, localized in the x direction and constructed from the eigenstates of a bulk 2D electron gas. If we time-propagate an initial WPs with an average k_x pointing towards the edge and identical for both up and down-spin states (left-going WP), the reflected WPs for up and down spins will have two different phase shifts $\phi_{\uparrow/\downarrow}$, and hence a *mutual spatial shift* l_S with respect to one another. For the model described here the shift, calculated from scattering-states' phase shift is

$$l_S = \left\langle \frac{d}{dk_x}(\phi_\uparrow - \phi_\downarrow) \right\rangle = -4\alpha_E - 8(2W - \langle e \rangle)\alpha_E^3 + \mathcal{O}(\alpha_E^4), \quad (6)$$

where the averaging is over the energy band of the consid-

ered WP and $e = (k_x^2 + k_y^2)/2$. We know that WPs separated by the time-step τ are orthogonal and we may place one electron in each WP. The nonequilibrium situation can be set in the standard fashion: occupying the WPs with $k_y > 0$ up to $E_F + \Delta V$ and those WPs with $k_y < 0$ only up to E_F. Deep inside the 2D bulk this WPs' shift will not contribute to any spin polarization because a series of occupied WPs within each band gives homogeneous density. However, since the up- and down-spin WPs are shifted, this shift must be directly related to the spin accumulation close to the edge so that to first order in α_E

$$n_\uparrow - n_\downarrow \sim \int_{\text{occ}} \frac{dk_y}{2\pi} l_S n(k_y) \sim -\frac{2\alpha_E}{\pi^2}\sqrt{2E_F}\Delta V, \quad (7)$$

where $n(k_y) = \sqrt{2E_F - k_y^2}/\pi$ is the number of initial WPs with momentum k_y. The dependence on the magnitude of the confinement, W comes only in the 3rd order, which follows from Eq. (6) and (7)

$$\frac{d}{dW}(n_\uparrow - n_\downarrow) = -\frac{8\alpha_E^3}{\pi^2}\sqrt{2E_F}\Delta V, \quad (8)$$

and hence the actual magnitude of the confinement potential is rather unimportant. Both of the results, Eqs. (7) and (8), agree very well with more involved and exact Green's function based treatments which will be reported elsewhere [28], and demonstrate the usefulness of the WPB concept not only for qualitative but also for reliable quantitative estimates.

It is interesting to compare the edge-SO scattering with the Rashba-Bytchov mechanism. The latter gives [23] $n_\uparrow - n_\downarrow = -\alpha_R^2(2E_F)^{-3/2}\Delta V/(12\pi^2)$, where α_R is the strength of the Rashba coupling; in the 2D GaAs systems it attains values [29] $\alpha_R \sim 1.8 \times 10^{-10}\,\text{eV cm} = 1.55 \times 10^{-2}\,\text{a.u.}^*$. On the other hand, the estimates for α_E in GaAs quantum wells give [30] $\alpha_E \sim 5.3\,\text{Å}^2 = 5.53 \times 10^{-4}\,\text{a.u.}^*$. The smallness of both α_E and α_R justifies the lowest order expansions used above. Finally, taking for the Fermi energy, $E_F = 36$ meV $= 3.01$ a.u.* corresponding to densities $n \sim 10^{12}$ cm^{-2} we find that the Rasba mechanism is 3 orders of magnitude smaller than the edge spin-orbit scattering.

In conclusion, our stroboscopic wave-packet basis permits both physical understanding and quantitative predictions to be obtained for a variety of nonequilibrium processes in which an extended system of electrons is subject to time evolution while being coupled to bulk reservoirs. The stroboscopic construction permits the time evolution of the system to be described straightforwardly, while the energy localization of the wave packets within precise energy bands ensures that the Pauli principle is properly respected in coupling to the reservoirs.

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