A wavepacket approach to periodically driven scattering

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Abstract

For autonomous systems it is well known how to extract tunneling probabilities from wavepacket calculations. Here we present a corresponding approach for periodically time-dependent Hamiltonians, valid at all frequencies, field strengths, and transition orders. After mapping the periodically driven system onto a time-independent one with an additional degree of freedom, use is made of the correlation function formulation of scattering [J. Chem. Phys. 98, 3884 (1993)]. The formalism is then applied to study the transmission properties of a resonant tunneling double barrier structure under the influence of a sinusoidal laser field, revealing an unexpected antiresonance in the zero photon transition for large field strengths.

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Recently, the quest for experimental controllability of transport processes on the nanoscale has triggered a number of theoretical studies on the influence of periodic external fields on molecular scale conductors [1, 2] as well as on semiconductor resonant tunneling heterostructures [3, 4, 5] and quantum dots [6]. Different theoretical techniques, ranging from Green’s function approaches [4, 5, 6], to the solution of master equations [1] and wavepacket propagation methods [3] have been employed to calculate either transmission probabilities or directly the currents for the considered setups.

In the seminal work of Büttiker and Landauer [7] an analytical approach to calculate the transmission probability of a sinusoidally driven potential barrier was made. The perturbative analysis allowed the calculation of central and side band transmission probabilities at the incoming energy and at the incoming energy plus or minus the photon energy. An analogous perturbative way of extracting those “driven” transmission probabilities was shortly afterwards used in numerical [8] and analytical [9] studies of resonant tunneling. The non-perturbative calculation of tunneling probabilities under absorption or emission of photons is a necessity if the influence of strong fields on the tunneling process is to be investigated, however. These probabilities play a central role in extending the Landauer formalism to conductance calculations in the presence of driving [10]. A viable generalization of standard approaches to make the determination of tunneling probabilities for arbitrary field strengths possible is therefore needed.

A very intuitive way to calculate the scattering $S(E)$ matrix as a function of energy, from which the tunneling probability $T(E)$ can be extracted, has been given in the framework of reactive scattering theory for autonomous systems [11]. This approach relies on the solution of the time-dependent Schrödinger equation and the subsequent Fourier transform of cross-correlation functions. According to the reactive scattering situation one distinguishes between internal and translational degrees of freedom, where the translational one usually represents a tunneling degree of freedom. In the following, this approach will now be modified to include, instead of the internal coordinate a new coordinate representing time in the case of a periodically time-dependent system. The absorption or emission of a quantum of radiation can then be accounted for by calculating the appropriate cross-correlation. We will elucidate the formalism by applying it to a resonant tunneling Hamiltonian modeling e. g. a weak link metal-metal contact.

In order to investigate the phenomenon of tunneling under the influence of an external
periodic field we follow an S-matrix approach. Therefore, the Hamiltonian of the driven system with a periodic driving term of the form \( f(x) \sin(\omega t) \) has to be made explicitly independent of time. This can e. g. be achieved by defining a new pair of canonical variables, \( \Theta = \omega t \) and \( p_\Theta \), with the frequency \( \omega \) of the external field [12]. The autonomous Hamiltonian for a driven particle of mass \( m \) then depends also on the new momentum and coordinate

\[
H(p_x, x, p_\Theta, \Theta) = \frac{p_x^2}{2m} + V(x) + f(x) \sin(\Theta) + \omega p_\Theta.
\]  

(1)

This Hamiltonian can easily be quantized by the usual replacements \( p_x, \Theta = -i\hbar \partial_x, \Theta \). In the following, we assume that this has been done by keeping in mind the operator nature of the Hamiltonian (in a different notation also known as the \((t, t')\) Hamiltonian [13]).

If the coupling term and the potential \( V(x) \) are localized, i. e. \( V(x), f(x) \to 0 \) for \( |x| \to \infty \), then the Hamiltonian at large values of \( |x| \) can be written as a sum of a kinetic energy term plus a term independent of \( x \)

\[
H \to H_0(p_x, x, p_\Theta, \Theta) = \frac{p_x^2}{2m} + h(p_\Theta),
\]  

(2)

where the eigenfunctions of \( h(p_\Theta) = \omega p_\Theta \) are exponentials, in their normalized form given by

\[
\chi_n(\Theta) = \frac{1}{\sqrt{2\pi}} e^{in\Theta},
\]  

(3)

and living on a finite support \( 0 \leq \Theta < 2\pi \). In the following we will refer to \( n \) as the number of quanta in the external field, due to the energy eigenvalue \( E_n = n\hbar\omega \) corresponding to the eigenfunction above. The eigenfunctions of the translational part of the Hamiltonian are energy normalized plane wave states with wavevector \( k \). For the asymptotic Hamiltonian \( H_0 \) this yields the direct product eigenstate and energy

\[
\psi_{n,E} = \chi_n(\Theta) \sqrt{\frac{m}{\hbar k}} \frac{1}{\sqrt{2\pi}} e^{ikx}, \quad E = n\hbar\omega + \hbar^2 k^2/(2m).
\]  

(4)

The eigenstates of the full Hamiltonian \( H \) are created by applying the Møller operator, defined by \( \Omega_{\pm} = \lim_{t \to \pm \infty} \exp\{iHt/\hbar\} \exp\{-iH_0t/\hbar\} \), to the asymptotic eigenstate

\[
\psi_{n,E}^\pm = \Omega_{\pm} \psi_{n,E}
\]  

(5)

and serve to define the on-shell S-matrix by

\[
S_{R,n';L,n}(E)\delta(E - E') = \langle \psi_{R,n';E'}^\pm | \psi_{L,n,E}^\pm \rangle
\]  

(6)
with a slight generalization of notation by including the “channel” indices L,R, denoting to which side of the barrier the eigenstates correspond. As noted in [14], eigenstates can be represented by Fourier transform of propagated wavepackets. This idea applied to Eq. (6) has been used later-on to extract the scattering matrix using wavepackets [11]. This reasoning can now be taken over to the present situation with the only difference that the eigenstates corresponding to the internal motion are states of the field variable \( \Theta \) defined in Eq. (3). The wavefunction to be propagated is a Gaussian wavepacket centered in the asymptotic regime of the translational degree of freedom times an eigenfunction in the field variable

\[
\phi_{L,n}(x, \Theta) = g_L(x)\chi_n(\Theta) = \left(\frac{2\alpha}{\pi}\right)^{1/4} \exp\{-\alpha(x-x_\alpha)^2 + ip_\alpha(x-x_\alpha)\}\chi_n(\Theta).
\]

For the S-matrix this leads to the expression

\[
S_{R,n';L,n}(E) = (2\pi\bar{\hbar})^{-1}\eta^*_{R,n'}(E)\eta_{L,n}(E)\int c_{R,n';L,n}(t)e^{iEt/\bar{\hbar}}dt,
\]

where a cross correlation function of the form

\[
c_{R,n';L,n}(t) = \langle \phi_{R,n'}|\exp\{-iHt/\hbar\}|\phi_{L,n}\rangle,
\]

is used, with a final state wavepacket defined analogously to Eq. (7) but to the right of the barrier. Furthermore, the arbitrariness of the initial and final wavefunctions is removed from the S-matrix expression by the normalization factor [11]

\[
\eta_{R,n}(E) = \sqrt{\frac{m}{2\pi\hbar k_n}}\int e^{-ik_\alpha x}g_R(x)dx,
\]

with \( k_n = \sqrt{2m(E - E_n)/\hbar} \) and a corresponding formula for \( \eta_{L,n'} \). We stress that one does not need to use Møller states as initial and final wavefunctions as long as they are located far in the asymptotic regions of the \( x \)-coordinate, such that the Møller operator becomes the unit operator. Furthermore, we note in passing, that an approach to extract scattering information for periodically driven quantum systems, similar in spirit, but based on Floquet theory has e. g. been pursued in [15], [16].

In the following we calculate transmission probabilities of a periodically driven system by numerically solving a 2 degree of freedom time-dependent Schrödinger equation
with Hamiltonian (1). The $x$ degree of freedom shall correspond to a tunneling electron coupled to the external field, represented by the $\Theta$ degree of freedom. Without driving, the electron is supposed to move in the symmetric resonant tunneling potential

$$V(x) = \left[ V_0(1 + \exp(-\beta x_c)) \right] / \left[ 1 + \exp\{\beta(|x| - x_c)\} \right] - V_{\text{res}} \exp\{-\gamma x^2\}$$

introduced by Bringer et al. [17] and applied in [17, 18] to study inelastic tunneling in the presence of coupling to a harmonic oscillator mode. We are using the parameter values $V_0 = 10$ eV, $\beta = 4$ a.u., $\gamma = 1$ a.u., $x_c = 4$ a.u. and $V_{\text{res}} = 14$ eV taken from [17] in the following. The undriven potential is then of the double barrier type, supporting a resonant tunneling “level” with a width of about 0.4 eV at an energy of $E \approx 4.9$ eV, well below the barrier top.

The periodic field is applied by using the function $f(x) = x V_c \Theta(x_c - |x|)$ localized in $x$ around the double barrier potential. This model is motivated by a lead-molecule-lead setup irradiated by a laser field, the field-matter interaction being treated in dipole approximation and cut off due to the metallic nature of the leads. A dipole driving without cutoff could in principle be dealt with by applying a Kramers-Henneberger (KH) transformation to an oscillatory reference frame [15], [16]. This leads, however, to a complicated dependence on position and time of the potential $V(x,t)$ and has to be dealt with by a transformation to the momentum gauge [16]. The field parameters are chosen to make the expected effects clearly visible. For the frequency we thus choose $\hbar \omega = 1$ eV, while the strength of the coupling to the external field $V_c$ will be varied in order to study its influence on the transmission probabilities. We will use parameters for which a perturbative approach becomes more and more questionable.

Numerical results will be presented for the dynamics of wavefunction (7) with Gaussian width parameter $\alpha = 1$ atomic units (a.u.), an initial center of position $x_\alpha = 10$ a.u. and an initial center of momentum $p_\alpha = 0.7$ a.u., having a sufficiently large overlap with the transmission resonances. We start out by assuming that the $\Theta$ degree of freedom is initially in the $n = 0$ state. Although, the number of quanta in the field has to be very large in order to justify the classical treatment of the driving term in Eq. (11), the total quantum wavefunction can be multiplied with a factor $\exp\{i(m-n)(\Theta - \omega t)\}$ and is still a solution of the time-dependent Schrödinger equation under the Hamiltonian (11). The exponential factor changes the number of quanta from $n$ to $m$, justifying our “shift of the origin” to $n = 0$. The final states, which the time-evolved wavepacket is to be overlapped with, are again direct products as in Eq. (7), but located on the right side of the barrier (with $x_\beta = -x_\alpha$ and
$p_\beta = p_\alpha$), with eigenstates in the $\Theta$ degree of freedom labelled by $n'$. We stress that there is no restriction on the transition (or “side band”) order $n'$ imposed by our formalism. In the following, we focus on the cases $n' = 0, 1, 2$, however. The grid employed in the numerics has 8192 points in the $x$ direction for the electron and 32 points in the $\Theta$ direction. In order to avoid unphysical reflections from the grid boundaries we have employed an absorbing negative imaginary potential smoothly turned on at large values of $|x|$.

In Fig. 1, the correlation functions entering the Fourier transforms in Eq. (8) are depicted for increasing coupling strengths of $V_c = 0.5, 1, 2$ eV as a function of time in atomic units. In panel (a) for the 0-0 transition, it can be seen that the coupling to the field leads to a change in the long time tail of the oscillation of the correlation function $c_{R,0;L,0}(t)$ setting in at about 50 a.u.. For the cases $n' = 1, 2$, i.e. excitation of one, respectively two photons of the field during the scattering process, the oscillations in the time signals last longer than in the $n' = 0$ case and die out at times of around 800 a.u. (not shown). The amplitude of the oscillations is substantially smaller than in panel (a). The overall shape of the signal in Fig. 1(a) is not changed very much by the coupling. This does not imply that the resulting transmission will be unaffected, however. In a semiclassical study of undriven 1d-tunneling it has e.g. been shown that, due to the effect of the normalization, minute differences between the semiclassical and the full quantum signal can lead to pronounced differences in the corresponding tunneling probabilities [19]. A similar effect of real, physical origin can be observed in the following due to the coupling to the light field.

To this end, we now calculate the tunneling probabilities from the correlation functions of Fig. 1 according to

$$T_{n'0}(E) = |S_{R,n';L,n=0}(E)|^2.$$  \(11\)

as a function of (total) energy by using Eq. (8) for the S-matrix. Due to the coupling between the field and the electron, the final photon number $n'$ may increase (decrease), accompanied by an equivalent energy loss (gain) of the electronic system, which has thus “emitted” (“absorbed”) a corresponding number of photons. The numerical results are depicted in Fig. 2. For the 0-0 transition, and for $V_c = 0.5$ eV, an isolated resonance with barely below unit transmission (solid line in panel(a)) at around 4.9 eV is observed, close to the unperturbed resonance energy. For increasing coupling strength, the peak in the transmission curve acquires a shoulder at about $\hbar \omega$ below the original peak and a corresponding dip to the right (panels (b) and (c)). The transmission probabilities with $n' = 1, 2$ increase with increasing
field strength, exhibiting a doubly peaked structure, with the dominant peak shifted from the unperturbed resonance position by $\hbar\omega$ to higher energies and a shoulder blue-shifted by around $2\hbar\omega$ which is most clearly visible for $V_c = 1$ eV. The peak shifted by $\hbar\omega$ describes the scattering of an electron coming in with an energy higher than the resonance but by loosing one quantum of energy closely matching the resonance energy leading to an increased transmission probability. For $V_c = 2$ eV, however, the resonances are already considerably broadened having almost lost the separated peak structure. Furthermore, the maximum of the 0-0 transition is shifted considerably to the left and the antiresonance character of the dip at around 6 eV is clearly emerging. This behaviour can no longer be described and understood perturbatively and is in sharp contrast to the case of inelastic coupling to a harmonic oscillator [18], where there are no dips but only peaks at integer multiples of the oscillator frequency to the right of the unperturbed resonance.

Finally, to check consistency, we have studied the case of a single field quantum being present initially by using $n = 1$. As a function of relative translational energy $E_t = E - n\hbar\omega$ in the incoming channel, the corresponding transmissions (not shown) then match exactly the ones shown in Fig. 2 i. e. $T_{11}(E_t) \rightarrow T_{00}(E)$ and $T_{21}(E_t) \rightarrow T_{10}(E)$. As has been noted before, the field eigenstate quantum number does not influence the scattering process. To compare the results directly, one has to consider the relative translational energy as the independent variable, however. Furthermore, in the present case, there is also an equivalence between e. g. $T_{01}(E)$ and $T_{10}(E)$ (both as a function of absolute energy). The special form of the potential together with the driving term make the Hamiltonian obey generalized parity symmetry (see e. g. [20] for a recent discussion of this symmetry in a similar context). Thus it can be shown by using Eqs. [15-14] that the two transmissions have to be equal. We have checked that this is indeed the case also in our numerical results ($T_{01}$ not shown).

We have shown that nonperturbative, exact numerical calculations of periodically driven tunneling probabilities, to any transition order, can be performed by employing an autonomous Hamiltonian in extended phase space. Within the wavepacket formalism of scattering pioneered in [11], the probabilities for tunneling under absorption or emission of field quanta can thus be extracted from suitably chosen cross-correlation functions. The presented approach can be applied to all driven scattering problems, as long as the field coupling $f(x)$ is localized in space. A way to deal with pure dipole driving in a scattering situation has been devised in [16] by using a KH transformation which could also be employed in the
present formalism.

For resonant tunneling, in sharp contrast to the case of inelastic coupling to a harmonic oscillator, the field case leads to an antiresonance in the 0-0 transition at high frequencies. Furthermore, in addition to the resonant tunneling problem, also systems like driven quantum dots [6] or molecular wire ratchets [1], and nonresonant barrier transmission problems, e.g. through semiconductor heterostructures, could be studied using the new methodology.

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FIG. 1: Real parts of correlation functions $c_{R,L,n;L,n'}$ for $V_c = 0.5$ eV (full line), $V_c = 1$ eV (dashed line), $V_c = 2$ eV (dotted line) as a function of time in a.u.: (a) $n = n' = 0$ (b) $n = 0, n' = 1$ (c) $n = 0, n' = 2$
FIG. 2: Transmission probabilities $T_{n'n}(E)$ for $V_c = 0.5$ eV (a), $V_c = 1$ eV (b), $V_c = 2$ eV (c) as a function of energy (translational equals absolute in the present case) of the electronic degree of freedom in eV: $n = n' = 0$ (full line), $n = 0, n' = 1$ (dashed line) $n = 0, n' = 2$ (dotted line).