

Reciprocity relations for time-independent transition probabilities of time-dependent Hamiltonians

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Reciprocity relations are derived between time-independent transition probabilities for absorption and emission of photons within a system governed by a time-dependent Hamiltonian. The proof combines the use of absorbing boundary conditions to express the probabilities and the (t, t') formalism. The case of a particle incident on an asymmetric double-barrier potential and interacting with an oscillatory field is given for illustration. [S1050-2947(98)04711-8]

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I. INTRODUCTION

Time-independent transition probabilities can be defined for a system subjected to a time-dependent perturbation if asymptotically, before and after the motion in the interaction region between the collision partners, the particles are decoupled from the perturbation. A very important case is that of the dipolar coupling with an oscillatory electric field: if the potential governing the collision is localized in space, by going to the acceleration frame (or Kramers-Henneberger representation [1,2]) this decoupling is guaranteed. The asymptotic forms of the wave functions describe free motion. General expressions for the transition probabilities have been derived [3,4] by applying to this situation Floquet theory in an extended (t, t') space. The expressions for the probabilities are very similar to those of time-independent Hamiltonians. However, the transition operator is built with a Green function containing the new dynamical variable t' and the scalar product requires integration over t' .

It has been shown recently by Vorobeichik and Moiseyev [5] that these transition probabilities can be cast in a different form, using previous work for the description of reactive collisions [6–8]. For the reactive case, the state-to-state transition probabilities are given the form [6–8]

$$P_{f \leftarrow i} = \left| \frac{i}{\hbar} \langle \Phi_f \epsilon_f G(E) \epsilon_i \Phi_i \rangle \right|^2. \quad (1)$$

Φ_f and Φ_i are the free asymptotic wave functions after and before the collision. ϵ_f and ϵ_i are absorbing complex potentials localized in the product and reactant regions. The Green function $G(E)$ is $(E - H)^{-1}$.

For the time-dependent case, the corresponding expression is [5]

$$P_{f \leftarrow i} = \left| \frac{i}{\hbar} \langle \langle \Phi_f \epsilon_f \tilde{G}(E) \epsilon_i \tilde{\Phi}_i \rangle \rangle \right|^2. \quad (2)$$

The Green function is now

$$\tilde{G}(E) = [E - \hat{H}(q, t')]^{-1} \quad (3)$$

defined with the Floquet Hamiltonian

$$\hat{H}(q, t') = -i\hbar \frac{\partial}{\partial t'} + H(q, t') - i\epsilon(q). \quad (4)$$

q represents the usual dynamical variables and t' an additional dynamical variable, whereas $\epsilon(q) = \epsilon_i(q) + \epsilon_f(q)$. Note that in both Eqs. (1) and (2) $\epsilon_i(q)$ and $\epsilon_f(q)$ can be replaced by $\epsilon(q)$. This fact simplifies the proof given in Sec. II. The symbol $\langle \langle | \rangle \rangle$ recalls that integration has to be performed on both q and t' .

The initial free wave function for one-dimensional motion of a particle of mass μ is

$$\tilde{\Phi}_i(q, t') = \sqrt{\frac{\mu}{\hbar k_i}} \exp[ik_i x] \exp[in\omega t] \quad (5)$$

while the final function is

$$\tilde{\Phi}_f(q, t') = \sqrt{\frac{\mu}{\hbar k_f}} \exp[ik_f x]. \quad (6)$$

In Eq. (5) n is positive in case of photon absorption and negative in case of emission. Initial and final energies are related by $E_f = E_i + n\hbar\omega$.

This note is making use of this formalism to prove that the absorption and emission probabilities are related in a special way. We consider the one-dimensional motion of a particle incident on an asymmetric potential $V(x)$. The potential is assumed to go to the same limit as $x \rightarrow \pm\infty$. The proof is given in Sec. II. An example with an asymmetric piecewise constant potential is studied in Sec. III. This case is amenable to an exact treatment [10,11].

II. RECIPROCITY RELATIONS BETWEEN ABSORPTION AND EMISSION PROBABILITIES

We consider an asymmetric one-dimensional potential [i.e., with $V(-x) \neq V(x)$], with x increasing as one goes from left to right. The transition probabilities for absorption or emission of n photons for a particle meeting this potential and interacting with an oscillatory field are *a priori* different for motion from $x = -\infty$ to $x = +\infty$ or the reverse motion. $T_n(E)$ and $R_n(E)$ denote the transmission and reflection probabilities for left to right motion, with the energy E being

changed by n quanta. For the reverse motion they are denoted $\tilde{T}_n(E)$ and $\tilde{R}_n(E)$.

The formulas that will be proven are

$$T_n(E) = \tilde{T}_{-n}(E + n\hbar\omega), \quad \tilde{T}_n(E) = T_{-n}(E + n\hbar\omega) \quad (7)$$

and

$$R_n(E) = R_{-n}(E + n\hbar\omega), \quad \tilde{R}_n(E) = \tilde{R}_{-n}(E + n\hbar\omega) \quad (8)$$

We emphasize that Eq. (7) relates quantities associated to the two ways to approach the interaction region, while Eq. (8) relates quantities describing the same collision at two different energies.

The complete set of eigenenergies E_α of the Floquet Hamiltonian

$$\hat{H}(x, t') = -i\hbar \frac{\partial}{\partial t'} + H(x, t') - i\epsilon(x) \quad (9)$$

and of its left and right eigenfunctions $\phi_{\alpha, m}^L(x, t')$ and $\phi_{\alpha, m}^R(x, t')$ make it possible to express the Green function as

$$\begin{aligned} [E - \hat{H}(x, t')]^{-1} &= \sum_{\alpha, m} |\phi_{\alpha, m}^L(x, t')\rangle [E - (E_\alpha + n\hbar\omega)]^{-1} \\ &\times \langle \phi_{\alpha, m}^R(x, t')| \end{aligned} \quad (10)$$

When real basis sets are used to expand the eigenfunctions and even when the Hamiltonian is complex scaled (this option is not used in our calculations, but the proof of the reciprocity relations holds in this case as well), the left and right eigenfunctions are related in a simple way [9]:

$$\phi_{\alpha, m}^L(x, -t') = \phi_{\alpha, m}^R(x, t'), \quad \phi_{\alpha, m}^R(x, -t') = \phi_{\alpha, m}^L(x, t') \quad (11)$$

Consider first the relation $T_n(E) = \tilde{T}_{-n}(E + n\hbar\omega)$. The probability $T_n(E)$ is the squared modulus of a transition amplitude, which can be written:

$$\begin{aligned} t_n(E) &= -\frac{i}{\hbar^2} \frac{\mu}{\sqrt{k_i k_f \alpha, m}} \sum [E_i + n\hbar\omega - (E_\alpha + m\hbar\omega)]^{-1} \\ &\times \left[\int \int dx' dt' e^{-ik_f x'} \epsilon(x') \phi_{\alpha, m}^L(x', t') \right] \\ &\times \left[\int \int dx'' dt'' e^{ik_i x''} \epsilon(x'') e^{in\omega t''} \phi_{\alpha, m}^R(x'', t'') \right]. \end{aligned} \quad (12)$$

The transmission probability $\tilde{T}_{-n}(E + n\hbar\omega)$ is the squared modulus of the amplitude:

$$\begin{aligned} \tilde{t}_{-n}(E + n\hbar\omega) &= -\frac{i}{\hbar^2} \frac{\mu}{\sqrt{k'_i k'_f \alpha, m}} \sum [E_i - (E_\alpha + m\hbar\omega)]^{-1} \\ &\times \left[\int \int dx' dt' e^{ik'_f x'} \epsilon(x') \phi_{\alpha, m}^L(x', t') \right] \end{aligned}$$

$$\times \left[\int \int dx'' dt'' e^{-ik'_i x''} \epsilon(x'') e^{-in\omega t''} \phi_{\alpha, m}^R(x'', t'') \right]. \quad (13)$$

Because of the permutation of initial and final energies, we have $k_i = k'_f$ and $k_f = k'_i$. Instead of the counting index m we introduce the index $\tilde{m} = n + m$. The transition amplitude is now

$$\begin{aligned} \tilde{t}_{-n}(E + n\hbar\omega) &= -\frac{i}{\hbar^2} \frac{\mu}{\sqrt{k_i k_f \alpha, \tilde{m}}} \sum [E_i + n\hbar\omega - (E_\alpha + \tilde{m}\hbar\omega)]^{-1} \\ &\times \left[\int \int dx' dt' e^{ik_f x'} \epsilon(x') \phi_{\alpha, \tilde{m}-n}^L(x', t') \right] \\ &\times \left[\int \int dx'' dt'' e^{-ik_i x''} \epsilon(x'') e^{-in\omega t''} \phi_{\alpha, \tilde{m}-n}^R(x'', t'') \right]. \end{aligned} \quad (14)$$

Changing t' into $-t'$ in the first integral changes ϕ^L into ϕ^R [cf. Eq. (11)]. Changing t'' into $-t''$ in the second integral has the opposite effect. Two further properties of the Floquet eigenfunctions.

$$\begin{aligned} \phi_{\alpha, \tilde{m}-n}^R(x', t') &= e^{in\omega t'} \phi_{\alpha, m}^R(x', t'), \\ e^{in\omega t''} \phi_{\alpha, \tilde{m}-n}^L(x'', t'') &= \phi_{\alpha, m}^L(x'', t''), \end{aligned} \quad (15)$$

transform the first integral of Eq. (14) into the second integral of Eq. (12) and the second into the first. The relation $\tilde{T}_n(E) = T_{-n}(E + n\hbar\omega)$ can be proven in a similar way. However, changing the energy E into $E - n\hbar\omega$ into the first of the two relations (7) and replacing n by $-n$ produce the second relation.

We turn now to the first of the two relations in Eq. (8). While in the transmission amplitudes the initial and final wave vectors are aligned, in a reflection amplitude the two wave vectors are pointing in opposite directions. This explains that in the latter case we are comparing two amplitudes for the same collision. The transition amplitude for reflection with absorption of n photons in the collision from left to right at energy E is

$$\begin{aligned} r_n(E) &= -\frac{i}{\hbar^2} \frac{\mu}{\sqrt{k_i k_f \alpha, m}} \sum [E_i + n\hbar\omega - (E_\alpha + m\hbar\omega)]^{-1} \\ &\times \left[\int \int dx' dt' e^{ik_f x'} \epsilon(x') \phi_{\alpha, m}^L(x', t') \right] \\ &\times \left[\int \int dx'' dt'' e^{ik_i x''} \epsilon(x'') e^{in\omega t''} \phi_{\alpha, m}^R(x'', t'') \right] \end{aligned} \quad (16)$$

while the transition amplitude for reflection with emission of n photons in the same collision but at energy $E + n\hbar\omega$ is

$$\begin{aligned}
r_{-n}(E+n\hbar\omega) = & -\frac{i}{\hbar^2} \frac{\mu}{\sqrt{k'_i k'_f}} \sum_{\alpha,m} [E_i - (E_\alpha + m\hbar\omega)]^{-1} \\
& \times \left[\iint dx' dt' e^{ik'_f x'} \epsilon(x') \phi_{\alpha,m}^L(x', t') \right] \\
& \times \left[\iint dx'' dt'' e^{ik'_i x''} \epsilon(x'') e^{-in\omega t''} \phi_{\alpha,m}^R(x'', t'') \right].
\end{aligned} \tag{17}$$

Changing the counting index as in the previous derivation and introducing the same properties of the Floquet eigenfunctions transform identically this transition amplitude into that for absorption of n photons at energy E .

The reciprocity relations in the particular case $n=0$ (no net photon absorption or emission, although there may be virtual processes during the collision) produce no information in the case of reflection: Eq. (8) becomes a trivial identity. On the other hand, Eq. (7) tells us that the transmission probabilities $T_0(E)$ and $\tilde{T}_0(E)$ are equal. This is trivial only if the potential is symmetric.

III. NUMERICAL ILLUSTRATION WITH AN ASYMMETRIC PIECEWISE CONSTANT POTENTIAL

Sacks and Szöke [10] have given an exact treatment of electron scattering by a piecewise constant one-dimensional potential in the presence of a radiation field of arbitrary intensity. It was shown recently [11] that this approach can be reformulated with the use of transfer matrices expressing continuity of wave functions and their derivatives at each discontinuity of the potential. This method has shown its efficiency for the calculation of transmission through the heterostructures met in semiconductor research [12,13]. The difference is that now the matrices are, in principle, of infinite dimensionality, since a wave has to be defined for arbitrary numbers of emitted or absorbed photons. As any one-dimensional potential can be viewed as the limit of a piecewise constant potential with steps of vanishing widths, this opens a route to the treatment of scattering by an arbitrary potential. For an illustration of the rules given in Eqs. (7) and (8) we choose the asymmetric double-barrier potential shown in Fig. 1. Two barriers of width 1 a.u. are separated by a well of width 2 a.u. and depth 1 a.u. The first barrier met by an electron coming from the left has a height of 1.5 a.u. while the second barrier has a height of 2 a.u. The calculations are made in the velocity gauge [10]. Exchange of energy between the particle and the field occurs only at the discontinuities of the potential. We have therefore free motion in the asymptotic regions, as in the Kramers-Henneberger representation for a localized potential. However, Gordon-Volkov waves are used instead of plane waves.

Figure 2 gives in panel (a) the transmissivity as a function of energy of the incoming electron in the absence of the radiation field. This transmissivity is the same when the electron approaches the double barrier from the left or from the right. It shows a resonance associated to a level of the intermediate well. The maximum value of the transmissivity is not unity because the potential arrangement is not symmetric. With an electric field of maximum amplitude 0.1 a.u. and

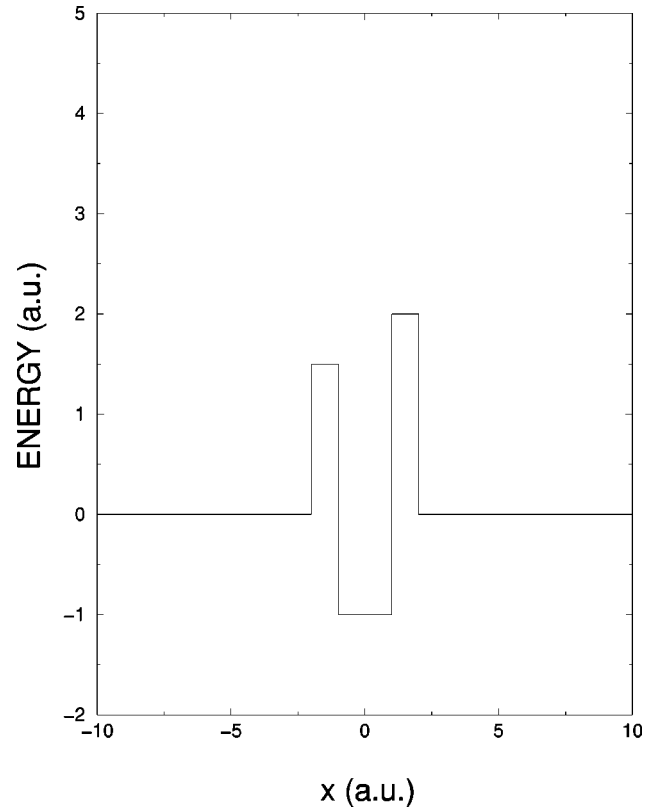


FIG. 1. An asymmetric double-barrier potential used to illustrate the reciprocity relations between transition probabilities for a particle interacting with an oscillatory field. Energies and distances are in a.u.

$\hbar\omega$ equal to 0.4 a.u., the number of amplitudes of incoming and outgoing amplitudes kept at every discontinuity is 21. This is required by the rather high value chosen for the electric field amplitude. The total transmissivity is the thick line of panel (b), while the thin line of this panel shows T_0 . The resonance is shifted, and a multiplet structure appears, with peak-to-peak separation equal to ω [14,11]. These transmissivities are not dependent upon the initial asymptotic position of the particle. Panels (c) and (d) depict reflectivities. In panel (c) we have the total reflectivity for positive initial momentum (thick line), while the thin line gives R_0 . In panel (d) the thick line is also the total reflectivity, but for initial negative momentum. It is identical to the total reflectivity of panel (c). However \tilde{R}_0 , given by the thin line, is different from R_0 . This is expected on the basis of the reciprocity relations for reflectivities [Eq. (8)].

In Fig. 3 we have the illustration of the reciprocity relations. Panel (a) shows $T_1(E)$ and $\tilde{T}_{-1}(E)$ while panel (b) gives $\tilde{T}_1(E)$ and $T_{-1}(E)$. Panel (c) gives $R_1(E)$ and $R_{-1}(E)$, while panel (d) represents $\tilde{R}_1(E)$ and $\tilde{R}_{-1}(E)$. The graphs are exactly as expected from relations (7) and (8). The transmissivities and reflectivities coincide, except for a shift equal to the field quantum. The doublets in the transmissivities are due to the fact that, if E_R is the resonance energy of the well, transmissivity is favoured at energies $E_R - \omega$ and E_R in a one-photon absorption process and at energies E_R and $E_R + \omega$ in a one-photon emission process.

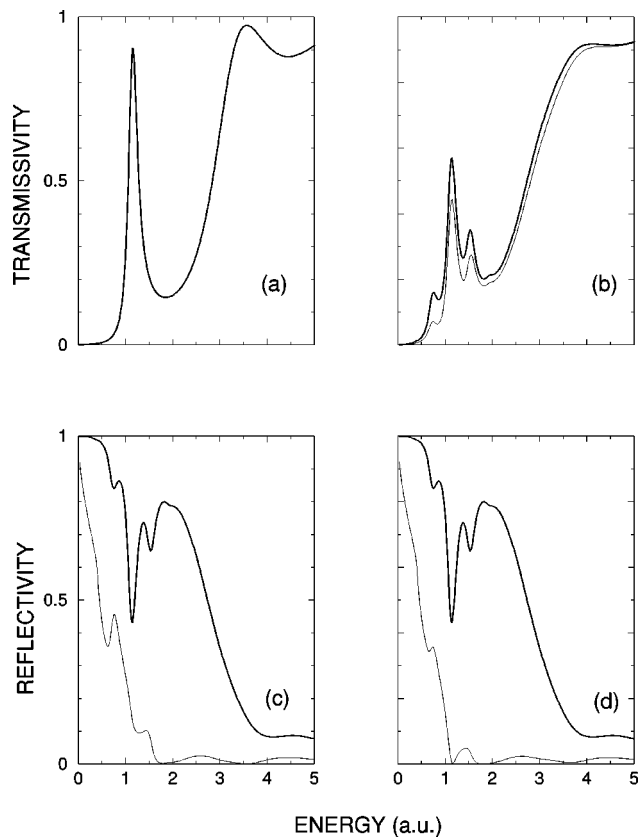


FIG. 2. Total and partial transmissivities and reflectivities for an electron incident in the double-barrier potential of Fig. 1. Panel (a): the field-free transmissivity showing a resonance structure due to a level of the intermediate well. Panel (b): total (thick curve) and zero-photon (thin curve) transmissivities when the electron is subjected to an oscillatory electric field of amplitude 0.1 a.u. and angular frequency 0.4 a.u. Panels (c) and (d): total (thick curve) and zero-photon (thin curve) reflectivities when the initial momentum is positive (c) or negative (d). While the total reflectivities are equal, the zero-photon reflectivities are different.

IV. CONCLUSION

The analysis of transition amplitudes in the Kramers-Henneberger frame, together with the use of the additional dynamical t' variable, is very efficient in establishing rela-

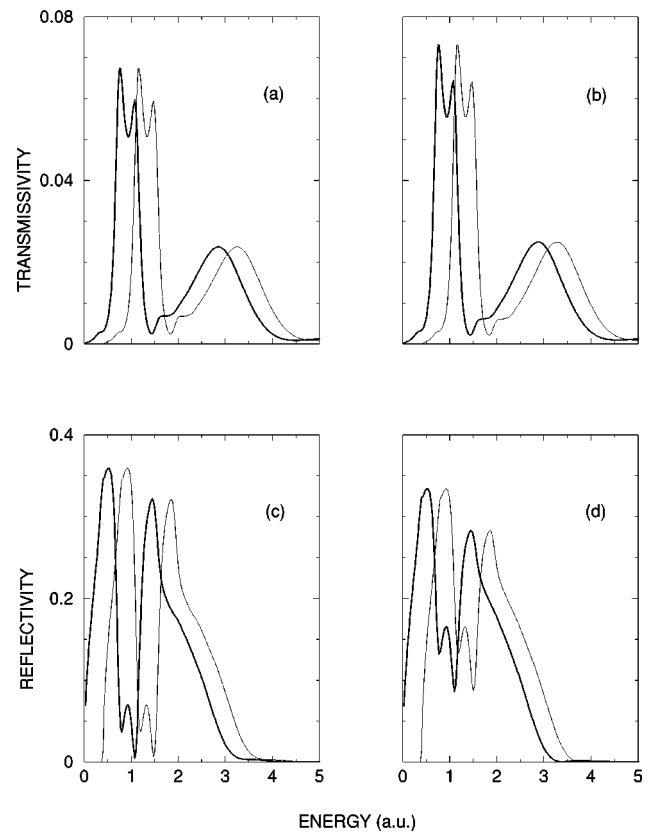


FIG. 3. Illustration of the reciprocity relations between absorption (thick curves) and emission (thin curves) transition probabilities. Panel (a): $T_1(E)$ and $\tilde{T}_{-1}(E)$. Panel (b): $\tilde{T}_1(E)$ and $T_{-1}(E)$. Panel (c): $R_1(E)$ and $R_{-1}(E)$. Panel (d): $\tilde{R}_1(E)$ and $\tilde{R}_{-1}(E)$.

tions among these amplitudes. It is likely that similar relations could be proven when the quantum system interacting with the field is described, in addition to the reaction coordinate, by some other variables.

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