Coherent-Photon-Assisted Cotunneling in a Coulomb Blockade Device

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Abstract

We study cotunneling in a double junction Coulomb blockade device under the influence of time dependent potentials. It is shown that the ac-bias leads to photon assisted cotunneling which in some cases may dominate the transport. We derive a general non-perturbative expression for the tunneling current in the presence of oscillating potentials and give a perturbative expression for the photon assisted cotunneling current.
I. INTRODUCTION

Single electron tunneling (SET) devices have attracted much attention during the recent years. They can be used to manipulate electrons one by one, and it is the hope that they can be applied as e.g. memory cells or as fundamental current standards. However, there are still a number of unanswered questions regarding the limitations of single electron devices. One such limitation is the so-called cotunneling process where an electron charge tunnels through the devices thus giving rise to a leakage current. This process has been studied extensively both theoretically and experimentally. Experiments have confirmed the theory of Averin and Nazarov, who derived the cotunneling current for a double junction system at small bias and low temperatures. It has been argued within the present understanding of these devices that extremely high accuracy could be achieved leading to metrological applications. However theory and experiments differ by several orders of magnitude. At present it not clear what the source of this discrepancy is. The intrinsic noise properties of the materials due to charge fluctuations is one possible source, but the quantum mechanics of the dynamically driven system is also an issue which should be considered in order to understand the fundamental limits.

Therefore an important aspect of single electron devices is their dynamical property and their response to an applied high frequency signal. So far theoretical works on single electron circuits have used an adiabatic approximation where it is assumed that the static cotunneling rate can be used at every time step. At first sight this approximation seems justified when the applied frequency is much lower than $\Delta / h$, where $\Delta$ is the electrostatic energy required to add or remove an electron. As we shall show, this is however not so obvious because activated cotunneling is not exponentially suppressed but increases as a power law of applied energy. The power law is a consequence of the fact that it is phase-space that limits the cotunneling process.

In this paper we investigate the importance of the photon assisted cotunneling induced by time dependent bias and gate voltages in the case of a double junction system in the Coulomb blockade regime. Note that this is different from cases previously discussed in the literature where the assisting photons are supposed to be supplied from an incoherent source, namely the electromagnetic environment connected to a heat bath. The coherent-photon-assisted cotunneling is shown to yield an important and in fact for certain parameters the dominant contribution to the current. We develop a non-perturbative formalism that combines cotunneling and sequential tunneling and thus generalizes previous works which dealt with the static case. However, our approach is restricted to the weak coupling limit and does not include renormalization effects. In the low frequency limit, we separate the photon assisted cotunneling in two contributions, one corresponding to an adiabatic approximation and one corresponding to higher order processes. In particular, we derive a perturbative result for the case of zero dc bias, which we refer to as a cotunneling electron pump.

The paper is organized as follows. In Section we introduce the double junction model. The tunnel current is derived in Section using scattering theory and several limits are considered: weak tunneling, sequential tunneling, the adiabatic limit and the case of small voltage and low temperature where a perturbation expansion is applicable. In Section numerical results are given and, finally, conclusions are in Section Technical details of
the time dependent scattering theory are in an appendix.

II. THE DOUBLE JUNCTION MODEL

We use the standard tunneling model for transport through a double barrier system with Coulomb interactions represented by the electrostatic energies. The model Hamiltonian reads

$$H = H_0 + H_T + H'(t),$$

where $H_0 = H_L + H_R + H_D + H_{\text{int}}$ and $H_L$, $H_R$, and $H_D$ represent the free electron parts in the left lead, in the right lead, and in the dot, respectively. The charging energy is included in $H_0$ and it is given by

$$H_{\text{int}} = E_C(N_D - n_g)^2 = E(N_D),$$

where $E_C$ is the charging energy, $n_g$ is set by the gate voltages and $N_D$ is the number of electrons in the dot. In Eq. (1) $H_T$ is the tunneling Hamiltonian with tunneling matrix element $T_{L(R)}$ connecting the dot to the left(right) lead. The time dependent parts are contained in $H'$ and we consider the case where the potentials of the leads and the dot (by the gate voltage) are modulated in time

$$H'(t) = \sum_{\alpha=L,R,D} \epsilon_\alpha(t) N_\alpha,$$

where $N_\alpha$ is the number operator in section $\alpha$.

By a standard unitary transformation (using $\hbar = 1$): $U = \exp \left\{ i \int_{-\infty}^{t} dt' H(t') \right\}$, we may transform the Hamiltonian into $H \rightarrow H_0 + H_T(t)$, whereby the time dependence is now present in the tunneling Hamiltonian

$$H_T(t) = \sum_{k,p;\beta=L,R} \left( T_\beta u_\beta(t) d_k^\dagger c_{\beta,p} + \text{H.c.} \right).$$

Here $c$ and $d$ are operators in the leads and in the dot, respectively, and

$$u_\beta(t) = \exp \left( i \int_{-\infty}^{t} dt' \left[ \epsilon_\beta(t') - \epsilon_D(t') \right] \right).$$

Without loss of generality we simplify the following discussion by transferring any non-zero dc part from the time dependent potential to the static bias or gate voltages. Consequently, if $\epsilon_\alpha(t)$ is periodic so is $u(t)$ and with the same period.

III. SCATTERING THEORY CALCULATION OF THE TUNNELING CURRENT

In order to calculate the cotunneling rates we generalize the standard time independent scattering theory and obtain an expression for the average transition rate $\bar{\Gamma}_{if}$ between an initial and a final state (both eigenstates of the unperturbed Hamiltonian) under the influence
of a time dependent perturbation (here $H_T(t)$). The $T$-matrix expansion then forms the basis for a non-perturbative expression for the current in the limit of small tunneling matrix elements. This approach thus generalizes previous scattering theory formulations\cite{4,5,7,18} to finite frequency and finite temperature. We furthermore utilize a Breit-Wigner-type approximation which does not include renormalization effects in the strong tunneling regime.\cite{19}

The result for the transition rate is derived in Appendix A. After time averaging it reads

$$\bar{\Gamma}_{fi} = 2\pi \sum_n \delta(E_i - E_f - \omega_n) \left| \langle f | T(n) | i \rangle \right|^2,$$

with $\omega_n = 2\pi n/T_0$ and $T_0$ being the period of the potentials. Here $n$ corresponds to the number of photons absorbed. The scattering operator $T$ is

$$T(n) = H_T(n) + \sum_m H_T(n - m)G_0(m)H_T(m)$$
$$+ \sum_{mm'} H_T(n - m - m')G_0(m + m')$$
$$\times H_T(m)G_0(m')H_T(m') + \cdots,$$

where the Fourier transforms are defined as $F(n) = \int dt e^{i\omega_n t} F(t)/T_0$. and the unperturbed Green’s function operator is given by

$$G_0(m) = \frac{1}{E_i + \omega_m - H_0 + i\eta}.$$ (8)

The scattering operator $T$ in Eq. (3) can be written as

$$T(n) = \sum_{mm'} H_T(n - m)G(m, m')H_T(m'),$$ (9)

where we have defined the Green’s function operator by the Dyson-like equation

$$G(m, m') = G_0(m)\delta_{m, m'}$$
$$+ \sum_{m''} G_0(m)\Sigma(m, m'')G(m'', m'),$$ (10)

where

$$\Sigma(m, m') = \sum_{m''} H_T(m - m'')G_0(m'')H_T(m'' - m').$$ (11)

Note that we have restricted the summation to even powers of $H_T$, since we shall calculate the transition rate of electron transfer through two barries.

A. Weak tunneling approximation

When the tunneling is not too strong, the off-diagonal element of the "self-energy" in Eq. (11) can be neglected, allowing for a solution of the Green’s function. With the notation $A_{\alpha\alpha'} = \langle \alpha | A | \alpha' \rangle$, the Green’s function becomes
\[ G_{\alpha\alpha'}(m, m') = \frac{\delta_{m,m'} \delta_{\alpha,\alpha'}}{[G_{0,\alpha}(m)]^{-1} + \Sigma_\alpha(m)}, \]  

(12)

where

\[ \Sigma_\alpha(m) = \sum_{\alpha',m''} \langle \alpha| H_T(m - m'')|\alpha'\rangle G_{0,\alpha'}(m'') \times \langle \alpha'| H_T(m'' - m')|\alpha\rangle. \]  

(13)

Inserting this into Eq. (8), we obtain for the \( T \)-matrix

\[ T_{if}(n) = \sum_{\alpha,m} \langle i| H_T(n - m)|\alpha\rangle G_{\alpha}(m) \langle \alpha| H_T(m)|f\rangle. \]  

(14)

This formula is equivalent to a Breit-Wigner formula (see e.g. 20), but here it is generalized to the case of a time dependent perturbation.

**B. The tunnel current**

Next we calculate the currents through the double junction system, that is the transition rate \( \gamma^+ \) for transferring an electron from left to right and the rate \( \gamma^- \) for electrons moving in the opposite direction. The total current is \( I = e(\gamma^+ - \gamma^-) \). In the positive direction the final state corresponding to an inelastic cotunneling event is given by

\[ |f^+\rangle = d_k^\dagger c_{L,p}^\dagger c_{R,p'}^\dagger |i\rangle, \]  

(15)

where \( |f^-\rangle \) is obtained by interchanging left and right. The intermediate state denoted by \( \alpha \) in Eq. (14), can take two values, corresponding to an electron being transferred through the left junction or the right junction first followed by an electron tunneling through the other junction. We assume that the coherence between the individual tunnelings can be neglected, which means that the tunneling uncertainty time \( \hbar / \max(eV, kT, \hbar \omega) \), is much smaller than \( e/I \). In this case, the current can be obtained from the transition rate between \( |f\rangle \) and \( |i\rangle \), where \( |i\rangle \) is thermally distributed, but with a fixed number of electrons on the island.

The lifetimes of the intermediate states are given by the imaginary part of \( \Sigma_\alpha \). The real part is neglected since it only gives a small shift of the electrostatic energy. The lifetime obtained from Eq. (13) equals the half of the Fermi’s Golden Rule transition rate for tunneling out of the intermediate state. In the present paper, we restrict ourselves to a two state approximation (denoted by \( N = 0, 1 \)) and therefore the lifetime is given by the transition rate from the intermediate state to the initial state.

By summing over all possible intermediate states while assuming the electronic occupation in the initial state to be in thermal equilibrium we finally obtain for tunneling rate in the positive direction

\[ \gamma^+ = P(0) \int_{-\infty}^{\infty} \frac{d\omega_L}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_R}{2\pi} \times \Gamma_L(\omega_L + eV_L)D_{LR}(\omega_L, \omega_R)\Gamma_R(\omega_R - eV_R), \]  

(16)
where
\[ \Gamma_{\alpha}(\omega) = \frac{G_{\alpha}}{e^2} \frac{\omega}{1 - \exp(-\beta\omega)} \equiv G_{\alpha}f(\omega), \quad (17) \]
and where \( P(0) \) is the probability that the system initially was in the state with \( N = 0 \). We must include this factor since our derivation for the current assumes that the initial state corresponds to the minimum electrostatic energy.

Above, we have introduced a function \( D \), which describes the propagation of the intermediate states. It is defined by
\[
D_{LR}(\omega_1, \omega_2) = 2\pi \sum_n \delta(\omega_1 + \omega_2 - \omega_n) \\
\times \left( \frac{u_L(m)u_R(n - m)}{\Delta^+ + \omega_1 - \omega_m + i\Gamma^-(\omega_1 - \omega_m)/2} \\
+ \frac{u_R(m)u_L(n - m)}{\Delta^- + \omega_2 - \omega_m + i\Gamma^+(\omega_2 - \omega_m)/2} \right)^2. \quad (18)
\]
Here the first term corresponds to an electron first tunneling through the left junction followed by an electron tunneling out of the right junction, while the second term comes from the reverse process. In Eq. (18) \( \omega_m \) is the photon energy in the intermediate state while \( \omega_n \) is the energy which is absorbed (emitted) during the cotunneling process. A non-zero \( \omega_n \) thus represents photon assisted cotunneling. The electrostatic energies in the intermediate states are given by
\[ \Delta^\pm = E(\pm 1) - E(0). \quad (19) \]
The lifetimes in Eq. (18) are
\[ \Gamma_{\alpha}^\pm(\omega) = \sum_{\alpha=L,R} \Gamma_{\alpha}^\pm(\omega), \]
\[ \Gamma_{\alpha}^\pm(\omega) = \sum_n |u_\alpha(n)|^2 \Gamma_{\alpha}(\omega - \omega_n \mp eV), \quad (20) \]
and \( \Gamma^\pm \) is thus the transition rate for an electron to tunnel through one junction in the presence of the oscillating fields. These rates are well-known from previous works on photon assisted tunneling in Coulomb blockade systems.\(^{1,2}\) \( \Gamma_{\alpha} \) is the tunneling rate without the ac field given in Eq. (17).

The reverse rate is obtained by interchanging left and right. We assume that the bias is such that \( V_L = V/2 \) and \( V_R = -V/2 \) (any asymmetry can be absorbed into the voltage on the island gate). We then obtain the final expression for the cotunneling current through the device as
\[
I = \frac{G_L G_R}{e^3} P(0) \int_{-\infty}^{\infty} d\omega_1 \frac{d\omega_2}{2\pi} \int_{-\infty}^{\infty} d\omega_2 \frac{d\omega_2}{2\pi} \\
\times \left[ f(\omega_1 + eV/2)D_{LR}(\omega_1, \omega_2)f(\omega_2 + eV/2) \\
- f(\omega_1 - eV/2)D_{RL}(\omega_1, \omega_2)f(\omega_2 - eV/2) \right]. \quad (21)
\]
Note that a non-zero current can result even for \( V = 0 \) if the time dependencies of the left and right junctions are different. Below we calculate the “pump” current based on this mechanism.
1. Sequential tunneling limit

In the limit of small $\Gamma$ the tunneling current reduces to the expression for the sequential tunneling described by the familiar master equation approach. This is seen as follows.

For small $\Gamma^\pm$, the tunneling rate $\gamma^\pm$ only has contributions from the poles of the function $D$. It can be shown that only diagonal terms of $D$ contributions and $D$ thus reduces to a sum of delta functions with weights $1/\Gamma^\pm(-\Delta^\pm)$, and in the two state approximation we keep only the terms corresponding to tunneling between $N = 0$ and $N = 1$. After integration the current then becomes equal to

\[
I_{\text{sequential}} = \frac{\Gamma_0^-(-\Delta^+)\Gamma_R^+(\Delta^+) - \Gamma_R^-(\Delta^+)\Gamma_L^+(\Delta^+)}{\Gamma^-(\Delta^+)}
\tag{22}
\]

and from Fermi’s Golden Rule it follows that $\Gamma^+(\Delta^+) = \Gamma_{01}$ and $\Gamma^-(\Delta^+) = \Gamma_{10}$, with $\Gamma_{ij}$ being the rate for tunneling from state $N = i$ to $N = j$ including photon assisted processes. From the master equation we have that $P(0) = \Gamma_{10}/(\Gamma_{10} + \Gamma_{01})$ and the sequential tunneling result follows.

C. Harmonic potentials

In this paper, we focus on harmonically varying fields. We define $\epsilon_\alpha(t) = W_\alpha \cos(\omega_0 t + \phi_\alpha)$ and find that the functions $u$ become $u_{(L,R)}(n) = J_{-n}(\alpha_{(L,R)D})$ and $u^*(n) = |u(-n)|^*$, where $J_n$ are the Bessel functions of the first kind. The arguments $\alpha_{LD}$ and $\phi_{LD}$ are given by

\[
\begin{align}
\alpha_{LD}^2 &= \alpha_L^2 + \alpha_D^2 - 2\alpha_L\alpha_D \cos(\phi_L - \phi_D), \tag{23a} \\
\alpha_{LD} \sin \phi_{LD} &= \alpha_L \sin \phi_L - \alpha_D \sin \phi_D, \tag{23b}
\end{align}
\]

where $\alpha_L = W_L/\omega_0$ and $\alpha_D = W_D/\omega_0$ and with similar relations for $L \to R$. Using the weak tunneling limit, where the energy arguments of the lifetimes in $D$ are approximated by the pole values, we obtain after some manipulations

\[
D_{LR}(\omega_1, \omega_2) = 2\pi \sum_n \delta(\omega_1 + \omega_2 - \omega_n) \left| \sum_m J_{m-n}(\alpha_{RD})J_m(\alpha_{LD})e^{im(\phi_{RD} - \phi_{LD})} \right|^2 \left( \frac{1}{\Delta^+ + \omega_1 - \omega_m + i\Gamma_{10}/2} + \frac{1}{\Delta^- - \omega_1 + \omega_m + i\Gamma_{01}/2} \right). \tag{24}
\]

This is the form which is utilized in the numerical results reported in Section IV. The $D$ function obeys the following relationship

\[
D_{RL}(\omega_1, \omega_2, \Delta^\pm, eV) = D_{LR}(\omega_2, \omega_1, \Delta^{\mp}, -eV). \tag{25}
\]
D. Adiabatic limit

In the low frequency limit where the frequency of the driving fields is much smaller than the inverse uncertainty time associated with the cotunneling event, \( \omega_0 \ll \Delta/\hbar \), the frequency dependence in the denominator in the expression for \( D_{LR} \), Eq. (24), drops out. If it is furthermore assumed that the \( eV, kT, W_{L,R,D} \ll \Delta \), the energy denominators may be replaced by \( \Delta^\pm \), and the summation can be performed. This is equivalent to using static result by Averin and Nazarov \(^3\) with a time dependent voltage given by \( eV_L(t) - eV_R(t) = eV + W_L \cos(\omega_0 t + \phi_L) - W_R \cos(\omega_0 t + \phi_R) \). The current becomes in this approximation

\[
I^{(0)}_{\text{adiabatic}} = \frac{G_L G_R \hbar}{12 \pi e^2} V \left[ (2 \pi kT)^2 + (eV)^2 + \frac{3}{2} \tilde{W}^2 \right] \times \left( \frac{1}{\Delta^+} + \frac{1}{\Delta^-} \right)^2,
\]

where \( \tilde{W}^2 = W_L^2 + W_R^2 - 2W_L W_R \cos(\phi_L - \phi_R) \). The two first terms is the result of ref. \(^3\). We see that the this formula gives no contribution when no dc bias is applied.

In the general case, the adiabatic result can be derived from the static cotunneling formula, i.e., with \( D_{LR}(\omega_1, \omega_2) \) setting \( u = 1 \), and \( \omega_0 = 0 \) (which is the formula obtained by Pasquier \textit{et al.} \(^7\)) but allowing for the gate and lead voltages to vary in time. To next leading order in the applied energies \( eV, kT \) or \( W_{L,R,D} \), we obtain after time averaging the formula

\[
I^{(1)}_{\text{adiabatic}} = \frac{G_L G_R \hbar}{24 \pi e^3} [W_{RD}^2 - W_{LD}^2] \times \frac{(\Delta^+ - \Delta^-)(\Delta^+ + \Delta^-)^2}{(\Delta^+ \Delta^-)^3} \times [(2 \pi kT)^2 + 3\tilde{W}^2/4].
\]

In the next section, we calculate the lowest order finite frequency correction to Eq. (27).

E. Perturbative result for photon assisted cotunneling current

Here we derive an perturbative result for the cotunneling current in the case of a “cotunneling pump”, i.e. the current without a dc bias. We assume that both the applied frequency and the temperature are low enough so that we can expand in powers of the energies. In addition, this approximation has the property that one can neglect the broadening effects due to the small finite lifetimes, because it is assumed that the energy is so low that the real transitions (corresponding to zeroes of the real part of the denominators), do not play a role.

In the case of zero dc bias the cotunneling current is given by Eq. (21) with \( V = 0 \). Now by using Eq. (25) and by interchanging \( \omega_1 \) and \( \omega_2 \) in Eq. (21) the difference \( D_{LR} - D_{RL} \) can be written as \( D_{LR}(\Delta^+, \Delta^-) - D_{LR}(\Delta^-, \Delta^+) \). Therefore the pump current vanishes when the gate voltage is set to zero. This is expected because in that case there is no preferred tunneling direction.
Next we expand in powers of the applied frequency $\omega_0$ whereby the integral in Eq. (24) can be performed analytically. After summation over frequencies we obtain the final expression for the pump current (reinserting $\hbar$)

$$I_{\text{PACT,pump}} = \left[ W_{RD}^2 - W_{LD}^2 \right] \frac{G_L G_R \hbar}{24\pi e^3} \times \frac{(\Delta^+ - \Delta^-)(\Delta^+ + \Delta^-)^2}{(\Delta^+ \Delta^-)^3} \times [(2\pi kT)^2 + 3\tilde{W}^2/4 + (\hbar \omega_0)^2].$$

The two first terms is the adiabatic result derived in the previous section. The last term is the lowest order quantum correction to the adiabatic limit.

IV. NUMERICAL RESULTS

In the following, we compare expression (28) with the current through a Coulomb island calculated on the basis of standard master equation approach, the so-called sequential tunneling limit obtained, see Section III B 1. In this calculation we must use the photon assisted sequential tunneling rates. In Fig. 1 we show the different current contributions as a function of frequency for the case where an ac current is applied to the left lead only. At low frequencies the photon assisted cotunneling is much larger than the sequential tunneling current. Even with a finite bias voltage ($b$) this still holds.

V. SUMMARY AND CONCLUSIONS

In summary, we have shown that the coherent photon assisted cotunneling is an important contribution to the charge transfer processes in single electron devices. It must therefore be taken into account when these devices are operated under periodic time dependent conditions, which is indeed the case in many potential applications. The photon assisted contribution that we have pointed out in this paper has not been included previously in models for SET systems, therefore it would be interesting to study experimentally the photon assisted cotunneling process, e.g., by studying a double junction device under the pump conditions, \textit{i.e.} with zero dc bias but asymmetrically biased.

However, since the coherent-photon-assisted cotunneling process is important in a small parameter space only, it may be difficult to separate the competing tunneling mechanisms from the process studied here. In order to observe the photon assisted cotunneling process one should thus bias the device as asymmetrically as possible because it is proportional to the \textit{difference} between the ac amplitudes, as can be seen in the perturbative result in Eq. (28), whereas the usual photon assisted sequential tunneling is additive in the applied ac power. Furthermore, since the cotunneling current stems from a higher order tunneling processes the resistances should not be too small. In the numerical examples presented in Fig. 1, resistances of 0.1 $h/e^2$ were used. In this case the photon assisted cotunneling current dominates for small frequencies but it is important to note that a part of this is due to the adiabatic cotunneling current, \textit{i.e.} it can be explained by a time averaged dc
FIG. 1. The photon assisted cotunneling compared to the sequential tunneling current (dashed curve) and the perturbative result (dotted curve) derived in Section III E in the case where only the left electrode potential is modulated. The parameters are $W_L = 0.5 \ E_C$, $W_R = W_D = 0$, $G_L = G_R = 0.1 \ e^2/h$, $kT = 0.01 \ E_C$, and $\Delta^+ - \Delta^- = 0.2 \ E_C$. Using a typical value for the charging energy $E_C = .4 \ meV$ corresponds to $kT = 50 \ mK$ and the frequency is scanned up to 40 GHz in the figure. In (a) there is no applied voltage whereas in (b) $eV = 0.1 \ E_C$. 

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\textbf{FIG. 1.} The photon assisted cotunneling compared to the sequential tunneling current (dashed curve) and the perturbative result (dotted curve) derived in Section III E in the case where only the left electrode potential is modulated. The parameters are $W_L = 0.5 \ E_C$, $W_R = W_D = 0$, $G_L = G_R = 0.1 \ e^2/h$, $kT = 0.01 \ E_C$, and $\Delta^+ - \Delta^- = 0.2 \ E_C$. Using a typical value for the charging energy $E_C = .4 \ meV$ corresponds to $kT = 50 \ mK$ and the frequency is scanned up to 40 GHz in the figure. In (a) there is no applied voltage whereas in (b) $eV = 0.1 \ E_C$. 
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cotunneling, giving rise to non-zero time average due to the non-linear bias dependence. Since the adiabatic contribution is frequency independent it can in principle be measured in the small frequency limit.

The non-perturbatively expression derived in the this paper generalizes previously derived cotunneling results for the dc case and as in those theories it does not take renormalization effects into account. Such effects are important in the strong tunneling regime and a study of these effects in the time domain is an interesting subject which deserves further work.

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APPENDIX A: TIME DEPENDENT SCATTERING THEORY

In this appendix, the standard $T$-matrix approach is generalized to the case of a periodic time dependent perturbation, $V(t)$, which is assumed to be turned on adiabatically in the distant past, i.e. it includes a factor $\exp(\eta t)$ where $\eta = 0^+$. We want to calculate the probability to find the system at time $t$ in the state $|f\rangle$ given that it started in the initial state $|i\rangle$, which is given by the square modulus of the overlap, $a_{if} = \langle f | i(t) \rangle$.

First consider the time evolution of an initial state $|i(t)\rangle = U(t, t_0)|i(t_0)\rangle$, (A1)

where $U(t, t_0)$ is the time evolution operator. Expanding in powers of $V(t)$ the $n$th order term becomes (for $t_0 \to -\infty$)

$$|i^{(n)}(t)\rangle = (-i)^n e^{-iH_0 t} \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \cdots \int_{-\infty}^{t_{n-1}} dt_n V_I(t_1)V_I(t_2)\cdots V_I(t_n)|i\rangle,$$ (A2)

where $V_I(t)$ is in the interaction picture. Now define the Fourier transforms $V(t) = \sum_n e^{-i\omega_0 nt} V(n)$ and insert these in (A2) which allows us to perform the integrations and obtain

$$|i^{(n)}(t)\rangle = e^{-iE_{0} t + \eta t} \sum_{\{m\}} \exp\left[-i\omega_0 (m_1 + \cdots + m_n) t\right]$$

$$\times G(m_1 + \cdots + m_n) V(m_1) \cdots G(m_{n-1} + m_n) V(m_{n-1}) G_0(m_n) V(m_n)|i\rangle \quad (A3)$$

where $G_0$ is given by Eq. (8) and $\omega_0 = 2\pi/T_0$.

Taking the square modulus of the overlap $a_{if}$, we obtain an expression of the form (emphasizing only the time dependence)

$$|a_{if}(t)|^2 = \sum_{n} \sum_{\{m\}\{m'\}} \exp(-i\omega_0 [m_1 + \cdots + m_n + m'_1 + \cdots + m'_{n'}] t) e^{2\eta t} \cdots \quad (A4)$$
The time averaged overlap is found by integrating over one period assuming that $T_0 \ll 1/\eta$. (This approximation implies that the frequency of the applied ac signal is much larger than the frequency of the tunnel events.) Averaging gives that the sum of the two sets of intermediate photon energies must vanish; below we denote the sum of one set as $j = m_1 + \cdots + m_\ell$. The tunneling rate is obtained as the time derivative of $|d|^2$ and we then obtain for the average rate

$$\bar{\Gamma}_{if} = \sum_j \frac{2\eta e^{2\eta t}}{(E_i - E_f - \omega_0 j)^2 + \eta^2} \left| \langle f | V(j) + \sum_m V(j - m)G_0(m)V(m) \right|^2 + \sum_{m_1,m_2} V(j - m_1 - m_2)G_0(m_1 + m_2)V(m_1)V(m_2)\cdots |i \rangle \right|^2.$$  \hspace{1cm} (A5)

In the limit $\eta \to 0$, we obtain the result quoted in the main text.
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