Virtual states and photon-assisted tunneling

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(Received 16 January 1996)

Perturbation theory is used to understand the transitions that dominate photon-assisted tunneling in coupled quantum-well systems immersed in intense ac fields. In particular, we show that the Bessel functions, widely incorporated into models describing photon-assisted tunneling, correspond to *net n*-photon transition probabilities to virtual states, and that the argument of these Bessel functions is gauge dependent. [S0163-1829(96)50928-8]

In this paper we make several observations that are useful to understand which transitions dominate photon-assisted tunneling (PAT) for coupled systems in the presence of an ac field. More specifically, we show that the Bessel functions, $J_n(\alpha)$, widely incorporated into models describing PAT, do not include transitions that, to lowest order in perturbation theory, yield Fermi's golden rule for direct field-assisted transitions. Here, we will also make two other interesting observations. First, the argument of the Bessel functions is gauge dependent, and therefore requires a careful interpretation for the Bessel function approach to be valid. Second, the square of the Bessel functions corresponds to *net n*-photon transition probabilities to virtual states,[1–4] or alternatively, to the probability of net *n*-photon virtual absorption/emission. For example, a process corresponding to the virtual absorption of 24 photons and emission of 23 photons is incorporated into $J_1(\alpha)$ as are other net one-photon processes. In addition to identifying the corrections to the Bessel function descriptions and providing a more familiar language for the strong-field effects, the perturbative formulation included here should give insight into how scattering and dissipation might be included in future descriptions of photon-assisted tunneling. Scattering and the Bessel function behavior of the PAT channels are believed to play a critical role in the recent observation of absolute negative conductivity (ANC) in semiconductor superlattices.[5]

In Ref. 6 Tien and Gordon (TG) showed that a solution to Schrödinger's equation could be found in the case of an electron in a superconducting tunneling junction coupled to an ac electric field. More specifically, if the perturbation, $eV\cos\omega t$, is defined to be the applied potential difference between two superconducting films, then the relevant solution to the Schrödinger's equation was shown to be

$$|a(t)\rangle = |a\rangle \sum_{n=-\infty}^{\infty} [J_n(\alpha) e^{-i(E_a + n\hbar\omega)t/\hbar}], \qquad (1)$$

where V is a constant, $\alpha = eV/\hbar\omega$, and the $J_n(\alpha)$ are Bessel functions. The $|a\rangle$ and E_a are the unperturbed eigenstate and energy, respectively, of an electron in one of the films. From their model they were able to explain qualitative features of PAT currents observed in superconducting diodes.[6–8] The TG model, in some sense, is the ''standard model'' of PAT and has been used extensively to describe PAT in other systems.[9–25] The TG model is also interesting because it neglects the direct ac-field-induced coupling between the states relevant in the tunneling, and assumes that the Bessel functions in Eq. (1) give the dominant field dependence in the PAT current. Furthermore, Eq. (1) strongly suggests that the Bessel function contribution to the PAT current can be understood in terms of ''virtual states.'' By virtual state, we mean a state that has the same spatial wave function, $\langle r|a\rangle$, as the unperturbed state $|a\rangle$, but has an associated energy that differs from the unperturbed energy E_a by some amount ΔE. Here, we will be interested in the special case that the virtual states are situated at energies given by $E_a + n\hbar\omega$.

Experimentally, PAT has also emerged recently in transport in semiconductor superlattices,[26–30] multiple quantum wells,[31,32] and nanostructures[33–35] in the presence of high-frequency fields. Some of the more recent observations include absolute negative conductance, multiphoton assisted tunneling, and PAT-induced electric-field domains.[5,29] Some of these results[5,8,27,29,30] lend support to the main prediction of their model, namely, the Bessel function dependence of the PAT channels. The predicted Bessel function behavior and the experimental support are particularly remarkable considering the fact that the matrix elements that contribute to the Bessel functions do not include direct field-assisted transitions.

In this paper we derive an expression for the perturbed quantum-well state, but unlike the TG approach, we will not make assumptions about which matrix elements should be neglected in the derivation. The Hamiltonian of interest is simply $H = (H_0 + H') + eV\cos\omega t$, where $H_0 + H'$ is the ac-field-independent part containing H_0, the unperturbed Hamiltonian of a double quantum-well system, and H' an assumed coupling between the quantum wells. Also, $eV\cos\omega t$, is the ac-field interaction energy; unlike TG, here V is a general operator, and we do not make any assumptions about which matrix elements should be neglected. We will consider only two states, one primarily localized in the left well $|a\rangle$, and the other primarily localized in the right well $|b\rangle$, with unperturbed energies E_a and E_b (see Fig. 1). For the sake of generality, we do not specify any other details of the quantum wells or their accompanying states, keeping in mind they might be neighboring wells in a superlattice under a dc bias.[5,26–30] In this two-state subspace, our Hamiltonian can be written as

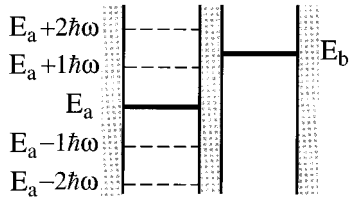

FIG. 1. The two-level double quantum-well system with the relevant virtual states.

$$H=\begin{pmatrix} E_a+eV_{aa}\cos(\omega t) & h_{ab}+eV_{ab}\cos(\omega t) \\ h_{ba}+eV_{ba}\cos(\omega t) & E_b+eV_{bb}\cos(\omega t) \end{pmatrix},$$

where V_{ij}, h_{ij} are the matrix elements $\langle i|V|j\rangle, \langle i|H'|j\rangle$; h_{ij} models the coupling that would lead to tunneling in the absence of the ac field.

We first focus our attention on one-photon absorption and emission, or terms that vary as $e^{\pm i\omega t}$. By standard time-dependent perturbation theory, the first-order correction to the unperturbed state, $|a\rangle e^{-iE_a t/\hbar}$, due to the ac field is given by

$$\begin{aligned}|a^{(1)}(t)\rangle = &-\frac{eV_{aa}}{2}|a\rangle\left[\frac{e^{-i(E_a+\hbar\omega)t/\hbar}}{-\hbar\omega-i\hbar\Gamma}+\frac{e^{-i(E_a-\hbar\omega)t/\hbar}}{\hbar\omega-i\hbar\Gamma}\right] \\ &-\frac{eV_{ba}}{2}|b\rangle\left[\frac{e^{-i(E_a+\hbar\omega)t/\hbar}}{E_b-E_a-\hbar\omega-i\hbar\Gamma}\right. \\ &\left.+\frac{e^{-i(E_a-\hbar\omega)t/\hbar}}{E_b-E_a+\hbar\omega-i\hbar\Gamma}\right], \end{aligned} \tag{2}$$

where Γ is a damping coefficient (see Refs. 2 and 36), taken here as the turn-on time of the ac field. We have ignored the transient response and will let $\Gamma\to 0$ below.

The term containing V_{ba} induces direct ac-field transitions between the states, and is said to cause $|b\rangle$ to be virtually excited if the detuning $E_b-E_a\pm\hbar\omega$ is not zero (or not less than the relevant dephasing time). An alternative description would be to say that V_{ab} mixes the state $|a\rangle$ at energy E_a, with virtual states $|b\rangle$ at energies $E_a\pm\hbar\omega$. This is in close analogy to models with quantized radiation fields,[37] where the states correspond to the quantum well states *plus* the radiation field. Similar interpretations can be used for the V_{aa} term which causes state $|a\rangle$ at E_a to mix with the virtual states $|a\rangle$ at $E_a\pm\hbar\omega$. This excitation will be virtual for $\omega>\Gamma$, since the detuning energy in this case is $E_a-E_a\pm\hbar\omega=\pm\hbar\omega$.

If we neglect V_{ab}, then we notice that the first set of terms in Eq. (2) contains the factor $\alpha=eV_{aa}/\hbar\omega$, and are the first terms in the series expansions of $J_1(\alpha)$ and $J_{-1}(\alpha)$, respectively. This is verified by the series expansion of the Bessel functions, which is given by

$$J_n(\alpha)=\sum_{k=0}^{\infty} J_n^{(2k+n)}(\alpha),$$

$$J_n^{(2k+n)}(\alpha)\equiv\frac{1}{k!(k+n)!}\left(\frac{\alpha}{2}\right)^n\left(\frac{-\alpha^2}{4}\right)^k. \tag{3}$$

Further, the next contribution to one-photon absorption/emission comes from third-order corrections $(n=1,k=1)$, which can be easily described using the virtual-state picture.

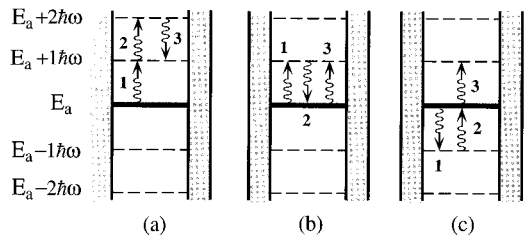

FIG. 2. The third-order diagrams that contribute to $J_1(\alpha)$.

That is, these purely V_{aa} contributions to the absorption require evaluating the three diagrams schematically depicted in Fig. 2. Each diagram corresponds to the virtual absorption of two photons $+\omega,+\omega$, and the virtual emission $-\omega$ of one photon for a net absorption of one photon (see Fig. 2). The explicit amplitudes for each diagram can be obtained with some care by mixing the real and virtual states involved; of course, time-dependent perturbation theory gives the same result.[38] In this case, careful consideration must be given to the damping coefficient Γ (see Refs. 2 and 36) to avoid the divergences that arise in individual diagrams. For example, the three diagrams in Fig. 2 yield

$$\begin{aligned}\lim_{\Gamma\to 0}\left[\right. &\frac{1}{(\omega+i\Gamma)(2\omega+2i\Gamma)(\omega+3i\Gamma)} \\ &+\frac{1}{(\omega+i\Gamma)(2i\Gamma)(\omega+3i\Gamma)} \\ &\left.+\frac{1}{(-\omega+i\Gamma)(2i\Gamma)(\omega+3i\Gamma)}\right]=-\frac{1}{2\omega^3},\end{aligned}$$

in addition to a factor of $(eV_{aa}/2)^3$, so these diagrams combine to give $J_1^{(3)}(\alpha)$. It is important to consider all diagrams in computing the transition amplitudes. Likewise, we could find the higher-order diagrams which, when summed, lead to other terms in Eq. (3). These all add to give $J_1(\alpha)$, and so it is interesting to realize that $J_1(\alpha)$ includes all ''net'' one-photon virtual absorption processes involving V_{aa}. Therefore, it actually includes multiphoton processes of all odd orders, and $J_1(\alpha)$ is accounted for exactly.

It is time consuming, but not difficult, to show that arbitrary orders in perturbation theory give the $J_n^{(2k+n)}(\alpha)$ term, where the superscript $(2k+n)$ denotes the order of the correction. For example, the lowest-order contribution (nth order) to an n-photon process consists of only a single diagram and is the easiest to evaluate. In general, the nth order contribution to the n-photon process contains a factor of $n!(\hbar\omega)^n$ in the denominator and V_{aa}^n in the numerator from the matrix elements. Therefore, the lowest-order contribution to the virtual absorption (emission) of n photons via V_{aa} gives the expected $J_n^{(n)}(\alpha)$.

Of course, other derivations of the Bessel function contribution to the wave function are possible. One could, for example, consider the unitary transformation that ''removes'' the diagonal terms from the Hamiltonian. This transformation in our two-state basis is simply a diagonal matrix $U(t)$, with diagonal elements

$$U_{mm}=e^{-iE_m t/\hbar}\exp\left(-i\int^t dt' eV_{mm}\cos\omega t'\right).$$

The exponential of the sinusoid leads to Bessel functions. After this transformation, the new effective Hamiltonian in the interaction picture will be off-diagonal only,

$$\tilde{H}=U(t)[H'+eV'\cos\omega t]U^{+}(t),$$

where V' is the same as V, but has zero diagonal elements. In this treatment U, and hence the virtual processes involving V_{aa} and V_{bb}, ''dress'' the bare tunnel coupling H' and V'.

What we would like to emphasize here, however, is that from perturbation theory a generalization of Eq. (1) is easily seen to have the following form:[38]

$$|a(t)\rangle=\sum_n \{[J_n(\alpha)+A_n]e^{-i(E_a+n\hbar\omega)t/\hbar}|a\rangle + B_n e^{-i(E_a+n\hbar\omega)t/\hbar}|b\rangle\}. \quad (4)$$

It is important to notice that, in addition to the TG, Bessel function contribution to the perturbed state, we have two additional coefficients A_n and B_n. These coefficients contain terms that include at least one matrix element of the form V_{ba} and/or V_{bb}. Furthermore, it can be seen that the Bessel functions contain no matrix elements connecting the state $|a\rangle$ to the state $|b\rangle$; they only contain the transition matrix element V_{aa}. Thus, the approach of Tien and Gordon is equivalent to treating V_{aa} to all orders, and neglecting the off-diagonal matrix elements. Then after obtaining analytic expressions for the states perturbed by V_{aa}, the coupling between the wells is introduced by an ac-field-independent tunneling probability T_{ab}; in our model h_{ab} plays a similar role. The n-photon-assisted tunneling probability is then given by $J_n^2(\alpha)T_{ab}$. At this point the reader might be alarmed by the fact that it is the B_n coefficient that, at lowest order in perturbation theory, yields Fermi's golden rule for the direct photon-assisted transition between the states. Thus, theories that retain only the Bessel functions neglect a class of transitions which are not restored by introducing the ac-field-independent tunneling probability T_{ab}. Nevertheless, the approach of Tien and Gordon should be a reasonable approximation provided that $J_n^2(\alpha)T_{ab}\gg|B_n|^2$.

It is also important to point out that there is some freedom in choosing how the Bessel functions enter into the formulation discussed above. In particular, since the energy origin can always be chosen arbitrarily, any one time-dependent constant can be added to both E_a and E_b in Eq. (4). Hence, a time-dependent phase (gauge) transformation allows one to consider either V_{aa} or V_{bb} to be equal to an arbitrary value; in particular, at least one of these can be chosen to be zero. The argument of the Bessel functions changes as V_{aa} and V_{bb} change under such transformations, but in the end all calculated quantities should be insensitive to these differences in formulation. This means that it is only $V_{aa}-V_{bb}$ that is physically meaningful, just as only E_b-E_a is physically meaningful. Tien and Gordon have made an implicit selection of the gauge such that V_{aa} is the difference in potential energy between the well that the electron is tunneling from, and the well that the electron is tunneling to. This choice is equivalent to setting $V_{bb}=0$. We stress that for a state $|b\rangle$ localized within the same well as $|a\rangle$, or for *next*-nearest-neighbor transitions, this gauge dependence requires a reinterpretation of α in order to apply the TG model. For example, for a dipole potential, $V=eE_{\rm ac}z\cos\omega t$, TG would write $\alpha=eE_{\rm ac}d/\hbar\omega$, where d is the well spacing. Hence for photon-assisted tunneling between next-nearest neighbors in a superlattice with lattice constant d, we would expect to observe Bessel functions with argument $\alpha=2eE_{\rm ac}d/\hbar\omega$. Furthermore, it is clearly seen that the density of virtual states,[6]

$$\rho_n(E)\equiv\rho(E-n\hbar\omega)J_n^2\left(\frac{edE_{\rm ac}}{\hbar\omega}\right),$$

is a gauge-dependent quantity, where $\rho(E)$ is the density of states without the laser field.

It is not obvious from the outset that the transitions that contribute to the Bessel functions (i.e., the V_{aa} matrix elements) will play an important role in actual experiments. Nevertheless, the observations presented in Refs. 5, 8, 27, 29, and 30 do lend support to the predicted Bessel function dependence of the PAT currents. However, as we have seen, neglecting V_{ab} will not always be appropriate. The results presented here may be useful for extending calculations to include V_{ab}.

In conclusion, we have shown how perturbation theory can be used to understand which transitions dominate photon-assisted tunneling (PAT) for coupled systems in the presence of an ac field. Furthermore, we have shown that the Bessel functions, widely incorporated into models describing photon-assisted tunneling, correspond to net n-photon transition probabilities to virtual states, and that the argument of these Bessel functions is gauge dependent. The framework presented here should be useful in understanding how the Bessel functions become modified in the presence of scattering mechanisms in a first-principles calculation.

Funding was provided by the Army Research Office and the NSF. B.J.K. would also like to thank Gloria Platero for useful discussions.

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