Laser-induced suppression of electron tunneling in a biased asymmetric double quantum well

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Laser-induced suppression of electron tunneling in a biased asymmetric double-quantum-well structure is numerically studied based on the Markoff master equation. A pump pulse is used to prepare initially the electronic wave packet which is irradiated by a cw laser. It is shown that if the bias voltage is adjusted to align the upper two electronic states in the narrow well and the wide well, suppression can occur before dephasing is complete. An analytical expression for the time evolution of the wave packet is derived by a degenerate-two-level model to give a qualitative interpretation to the origin of the electron localization.

I. INTRODUCTION

Grossmann and co-workers[1] have shown that an electron in a quartic double well can be confined to one of the wells by irradiation from a cw laser. An interesting feature of this suppression (or coherent destruction) of the electron tunneling is that the cw laser constructs a proper superposition of the wave functions resulting in electron localization. This work was later refined by Bavli and Metiu,[2] who numerically integrated the Schrödinger equation to obtain the time evolution of the electronic wave packet, explicitly taking into account the pulse rise time and the phase of the cw laser. In this latter work, a claim is made that the extent of the localization is rather sensitive to the parameters of the cw laser.

Since the systems studied so far have potentials with (almost) the symmetric structure, it is unclear whether laser-induced suppression of the electron tunneling can occur in an asymmetric double quantum well (DQW). Furthermore, in condensed systems, there exist dephasing processes which must destroy the superposition state, i.e., electron localization. To investigate these effects on the coherent destruction of electron tunneling, the semiconductor DQW heterostructure[3] offers a convenient setting because such a structure can be tailored to specific purposes.

The observation of coherent dynamics in the DQW requires experimental techniques with subpicosecond-time resolution, because high quality samples typically have dephasing times of a few picoseconds. However, the recent development in femtosecond pulse generation makes it possible to directly observe coherent tunneling of an optically excited electronic wave packet[4] and even interference between the wave packets created by phase-locked pulses.[5] Thus, the semiconductor DQW system has the advantage of experimentally elucidating characteristics of laser-induced suppression of electron tunneling.

For these reasons, we are concerned with the dynamics of an electronic wave packet in a biased asymmetric DQW in the presence of a cw laser. The wave packet is initially prepared by a pump pulse whereby the artificial rise time of the cw laser can be avoided. To see the dephasing effects on the electron localization, the Markoff master equation is used to determine the time evolution of the wave packet. Special attention is paid to seeing how the structure of the DQW as well as the properties of the cw laser affect electron localization. Qualitative interpretation of the laser-induced suppression of the electron tunneling is given based on a degenerate-two-level model.

II. THEORY

Our calculations start from the Markoff master equation,[6] in which the dynamics of the relevant system is described by the Hamiltonian

$$H^t = H + V_s^t + V_p^t = H_s^t + V_p^t \ . \tag{1}$$

The Hamiltonian of the electron in the DQW is H, and the interaction between this electron and the cw laser (the pump pulse) is given by V_s^t (V_p^t), where

$$V_s^t = -\mu E_s^0 \cos(\omega_s t + \phi_s) \tag{2a}$$

and

$$V_p^t = -\mu f(t) E_p^0 \cos(\omega_p t + \phi_p) \tag{2b}$$

with the Gaussian pulse envelope function $f(t)$.

Introducing the double space (Liouville-space) notation,[7,8] the master equation is written as ($\hbar = 1$)

$$\frac{\partial}{\partial t} |\rho(t)\rangle\rangle = -(iL^t + \Gamma)|\rho(t)\rangle\rangle \ , \tag{3}$$

where the Liouvillian is given by

$$L^t = L + K_s^t + K_p^t = L_s^t + K_p^t \ , \tag{4}$$

which corresponds to the Hamiltonian in Eq. (1). The relaxation operator Γ is characterized by the matrix elements

$$\langle\langle mm|\Gamma|\rho(t)\rangle\rangle = \gamma_m(\rho_{mm}(t) - \rho_{mm}^{eq}) \ , \tag{5a}$$

with an equilibrium population of the mth level ρ_{mm}^{eq}, and

$$\langle\langle mn|\Gamma|\rho(t)\rangle\rangle=\gamma_{mn}^{(d)}\rho_{mn}(t),\quad (m\neq n)\ , \tag{5b}$$

where

$$\gamma_{mn}^{(d)}=\frac{\gamma_m+\gamma_n}{2}+\gamma_{mn}^{(pd)}\ , \tag{5c}$$

with the pure dephasing rate $\gamma_{mn}^{(pd)}$.

The formal solution of Eq. (3) is expressed as

$$|\rho(t)\rangle\rangle=\exp[-(iL+\Gamma)t]|\sigma(t)\rangle\rangle\ , \tag{6a}$$

with

$$|\sigma(t)\rangle\rangle=U(t,-\infty)|\sigma(-\infty)\rangle\rangle$$
$$=\exp\left[-i\int_{-\infty}^{t}d\tau\{K_s^\tau(\tau)+K_p^\tau(\tau)\}\right]|\sigma(-\infty)\rangle\rangle\ , \tag{6b}$$

where the operators in the interaction picture are defined by

$$K_s^\tau(t)=\exp[(iL+\Gamma)\tau]K_s^\tau\exp[-(iL+\Gamma)\tau]\ , \tag{7a}$$

and

$$K_p^\tau(t)=\exp[(iL+\Gamma)\tau]K_p^\tau\exp[-(iL+\Gamma)\tau]\ . \tag{7b}$$

The time evolution of $|\sigma(t)\rangle\rangle$ is calculated according to the second-differential scheme[9] generalized to the Liouville-space-time evolution operator

$$|\sigma(t+\Delta t)\rangle\rangle-|\sigma(t)\rangle\rangle=-2i\Delta t\{K_s^t(t)+K_p^t(t)\}|\sigma(t)\rangle\rangle+O(\Delta t)^3\ . \tag{8}$$

The excited electronic wave packet is obtained by

$$W(q,t)=\langle\langle qq|P_e|\rho(t)\rangle\rangle\ , \tag{9}$$

where the projector

$$P_e=\sum|mn\rangle\rangle\langle\langle mn|$$

represents the states concerned and it is summed from the states m,n to all the excited states. If one wants to obtain the occupation probability in some region R, it is given by integrating $W(q,t)$ over this region,

$$P_R(t)=\int_R dq\ W(q,t)\ . \tag{10}$$

III. RESULTS AND DISCUSSION

We adopt the asymmetric DQW structure consisting of an 80 Å $Al_{0.1}Ga_{0.9}As$ middle barrier sandwiched between 190 and 145 Å layers of GaAs, which is the same as used in our earlier paper.[10] For computational simplicity, in this paper we are concerned with intersubband transitions, and assume that only the lowest state is initially (before pumping) populated. To realize such a situation, the extended barrier regions are assumed to be doped with a suitable dopant to give free-electron carriers. When there is no tunneling interaction nor external fields, both the wide (190 Å) well (WW) and the narrow (145 Å) well (NW) have two bound states, say $|W_k\rangle$ and $|N_k\rangle$ ($k=1,2$), respectively. Relevant to the bias voltage and tunneling interaction, those four zero-order states are mixed with each other to construct four eigenstates, say $|n\rangle$ ($n=1$–4). These eigenstates can be numerically obtained according to the procedure shown, e.g., in Ref. 11. In our scheme, the excited wave packet associated with the tunneling dynamics is created by the pump pulse which coherently brings the ground-state population into the two excited states $|3\rangle$ and $|4\rangle$. This wave packet is irradiated by the cw laser. The potential profile together with the energy levels of the eigenstates are illustrated in Fig. 1(a). In this paper, we consider two cases of the bias voltage. One of them, called the resonant case, is the case where the zero-order excited states $|W_2\rangle$ and $|N_2\rangle$ are aligned by adjusting the bias voltage. The other bias is set to be 20% larger in magnitude than that in the resonant case, and this is called the off-resonance case.

Figure 1(b) shows the absorption spectra. Since the ground state in the presence of the bias voltage is almost localized in the NW, and the two absorption peaks have similar intensities, we can see that in the resonant case, the excited states $|3\rangle$ and $|4\rangle$ are approximately represented by the symmetric and asymmetric combinations of the zero-order states $|W_2\rangle$ and $|N_2\rangle$.

First we consider the coherent destruction of electron

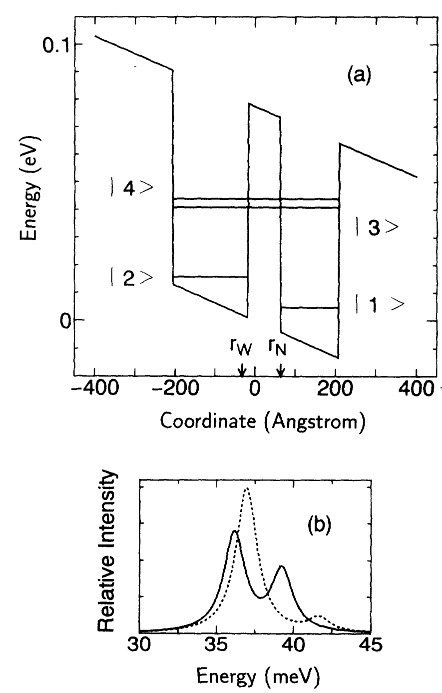

FIG. 1. (a) Schematic picture of the energy diagram of the DQW with four eigenstate levels at the resonant bias voltage of 6.36 kV/cm. (b) Absorption spectra in the resonant (solid line) and off-resonant (dashed line) case of the bias voltage. The values of the relaxation parameters are chosen as $\gamma_1=0.0$, $\gamma_3=\gamma_4=0.06$, $\gamma_{13}^{(pd)}=\gamma_{14}^{(pd)}=0.8$, and $\gamma_{34}^{(pd)}=0.3$ meV.

tunneling in the resonant case and are concerned with the time evolution of the population $P_{NW}(t)$ of the excited wave packet in the NW, $P_{WW}(t)$ in the WW, and $P_{MB}(t)$ in the middle barrier, whose domains of integration in Eq. (10) are, respectively, given by $(r_N, +\infty)$, $(-\infty, r_W)$, and (r_W, r_N) [Fig. 1(a)]. With the pump pulse, a very low intensity (10^5 W/cm^2) is chosen so that the excited population is built up only by a single-photon-absorption process. The temporal peak is set to be $t=0$, and the central frequency is tuned to

$$\omega_p = \frac{\epsilon_3+\epsilon_4}{2} - \epsilon_1 - \Omega_R \ , \tag{11a}$$

with the Rabi frequency

$$\Omega_R = 2|\langle 4|\mu|3\rangle||E_s^0| \ . \tag{11b}$$

The intensity of the cw laser is set as 10^5 W/cm^2, because a more intense laser induces a larger Stark shift which may cause unexpected optical transitions. (The 10^5 W/cm^2 intensity gives a 10.92 meV Rabi frequency.) For the sake of comparison, in Fig. 2(a) we show the time evolution of the excited population in the WW and the NW in the absence of the cw laser. The other parts in Fig. 2 are calculated by employing several values of the frequency of the cw laser. We can see that laser-induced suppression of tunneling also occurs in the asymmetric DQW in our pump-probe excitation scheme. A high power cw laser is needed for establishing the electron localization, though the intensity used in our calculation is stronger than that used in Ref. 2 by three orders of magnitude. The qualitative interpretation to the suppression of the tunneling will be given later by the degenerate-two-level model.

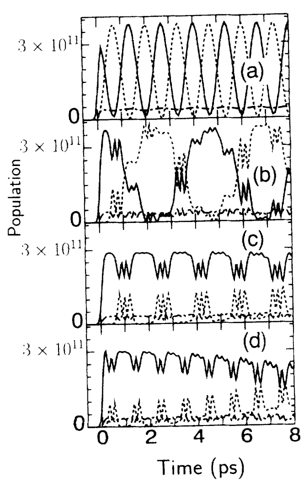

FIG. 2. Time evolution of the population $P_{NW}(t)$ of the excited wave packet in the NW (solid line), $P_{WW}(t)$ in the WW (dashed line), and $P_{MB}(t)$ in the middle barrier (dot-dashed line). A 10^{-5} W/cm^2 pump pulse is used, whose temporal full width at half maximum is 300 fs. The results calculated in the absence of the cw laser are shown in (a). Those in the presence of the cw laser are given in the other figures, with the detuning factor $\Delta=\omega_s-(\epsilon_4-\epsilon_3)$, (b) 0.0, (c) -0.5, and (d) 1.0 meV. The intensity and the phase of the cw laser are, respectively, set to be 10^5 W/cm^2 and $\phi_s=0$.

Figure 3 shows the relaxation effects on the coherent destruction of tunneling. In a typical DQW system, the dephasing rate can be estimated to be in the range 1–0.1 meV from the quantum beat experiments.[4] In the present calculation, we employ a fixed value of 0.8 meV for pure dephasings between the ground and excited states, $\gamma_{13}^{(pd)}$ and $\gamma_{14}^{(pd)}$, while that between the excited states $\gamma_{34}^{(pd)}$ is set to be (a) 0, (b) 0.1, (c) 0.2, and (d) 0.3 meV. The results indicate that localization can take place before the dephasing associated with the tunneling motion is complete. Comparing Figs. 2(a) and 3(a), on the other hand, the dephasing associated with the pump process makes little contribution to the coherent destruction of tunneling.

So far, we have discussed tunneling suppression in the resonance case. Here we show the time evolution of the excited population in the off-resonance case of the bias voltage, which is given in Fig. 4. The calculation is performed by choosing the same intensity 10^5 W/cm^2 for the cw laser, however, no electron localization can be observed. This result is unchanged even when we employ other frequencies of the cw laser, or employ a more intense cw laser, e.g., 10^7 W/cm^2, which gives a Rabi frequency larger than that in the resonance case by a factor of ten.

We are now in a position to give a physical interpretation to the suppression of tunneling based on an analytical expression. For this purpose, we make following approximations and/or assumptions. The intensity of the pump pulse is assumed to be weak enough that the interaction V_p^t can be treated as perturbation together with the rotating-wave approximation (RWA). The validity of

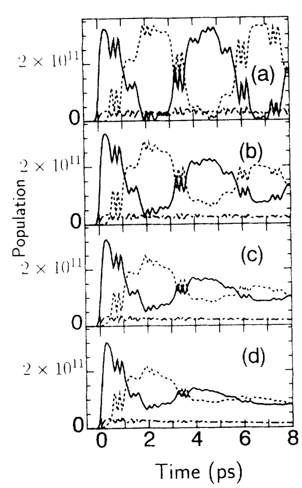

FIG. 3. Dephasing effects on the time evolution of the population in the resonant case of the bias voltage when irradiated by a 10^5 W/cm^2 cw laser with frequency at $\omega_s=\epsilon_4-\epsilon_3$. The phase of the cw laser is set to be zero. The solid, dashed, and dot-dashed lines represent the time evolution of the population of the excited wave packet in each region, $P_{NW}(t)$, $P_{WW}(t)$, and $P_{MB}(t)$, respectively. The pump pulse is the same as that used in Fig. 2. The values of the relaxation rate constants associated with the optical pump are set to be $\gamma_{13}^{(pd)}=\gamma_{14}^{(pd)}=0.8$ meV. The pure dephasing rates associated with the tunneling $\gamma_{34}^{(pd)}$ are chosen as (a) 0.0, (b) 0.1, (c) 0.2, and (d) 0.3 meV, while $\gamma_3=\gamma_4=\frac{1}{5}\gamma_{34}^{(pd)}$ is used to determine the T_1 rates.

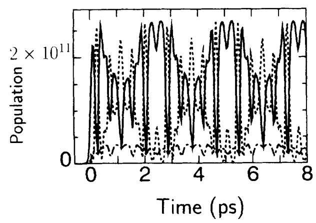

FIG. 4. Time evolution of the population $P_{NW}(t)$ of the excited wave packet (solid line), $P_{WW}(t)$ (dashed line), and $P_{MB}(t)$ (dot-dashed line) in the off-resonant case of the bias voltage when irradiated by a 10^5 W/cm^2 cw laser with frequency at $\omega_s=\epsilon_4-\epsilon_3$ and phase $u\phi_s=0$. The same pump pulse as that used in Fig. 2 is employed. All the relaxation parameters are set, in value, to be zero.

these approximations can be checked by numerical calculations. As for the temporal width of the pump pulse, we make the assumption that it is much shorter than the time scale of the evolution of the excited population. Moreover, we neglect the relaxation terms since suppression occurs before the dephasing is complete. Under these conditions, the dynamics of the excited population is determined by the Schrödinger equation, whose solution is

$$|\psi(t_0)\rangle = U_s(t,t_0)|\psi(t_0)\rangle \ , \tag{12}$$

except for a constant factor. Here the initial condition at time t_0 (at the temporal peak of the pump pulse) is given by

$$|\psi(t)\rangle = \mu_{31}|3\rangle + \mu_{41}|4\rangle \ , \tag{13}$$

along with $\mu_{31}=\langle 3|\mu|1\rangle$ and $\mu_{41}=\langle 4|\mu|1\rangle$, and the time-development operator is

$$U_s(t,t_0)=T\exp\left[-i\int_{t_0}^{t} d\tau H_s^{\tau}\right] . \tag{14}$$

We note that the phase of the pump pulse does not remain in these approximate expressions. Actually, we can see from the numerical results that this is also true in the case of finite-pulse pumping.

Apparently, exact closed-form solutions to Eq. (12) are known only for the case of degenerate energy levels. Rahman[12] has derived the asymptotic solution when

$$\lambda=\frac{|\Omega_R|^2}{(\epsilon_4-\epsilon_3)^2}\rightarrow\infty \ , \tag{15}$$

and pointed out that the two-level system irradiated by a strong laser behaves as a degenerate system. In the resonant and off-resonant cases of the bias voltage, the values of the parameter λ are 12.4 and 2.4, respectively, for the cw laser intensity of 10^5 W/cm^2. To obtain a qualitative understanding, we proceed with our analysis based on this degenerate-level model.[13] It is known that there exists a new basis set

$$|\pm\rangle=\frac{1}{\sqrt{2}}(|3\rangle\pm|4\rangle) \ , \tag{16}$$

in which the Hamiltonian H_s^t is diagonal. These are dressed states in the sense that the matrix elements are

$$\langle\pm|H_s^t|\pm\rangle=\epsilon\pm\tfrac{1}{2}\Omega(t) \ , \tag{17a}$$

and

$$\langle\pm|H_s^t|\mp\rangle=0 \ , \tag{17b}$$

where ϵ could be ϵ_3 or ϵ_4 and

$$\Omega(t)=\Omega_R\cos(\omega_s t+\phi_s) \ . \tag{18}$$

These dressed states give a description of the state vector in Eq. (12) as the superposition

$$|\psi(t)\rangle=C_+(t)|+\rangle+C_-(t)|-\rangle \ . \tag{19a}$$

Here in

$$C_\pm(t)=\exp\{-i[\epsilon(t-t_0)\pm\xi(t)]\}C_\pm(t_0) \ , \tag{19b}$$

along with

$$\xi(t)=\tfrac{1}{2}\int_{t_0}^{t} d\tau\Omega(\tau) \ , \tag{19c}$$

the initial condition $C_\pm(t_0)$ can be determined by Eq. (13).

If the system is prepared in one of these dressed states at time t_0, then subsequent times find no change in the probability. This is the case when we employ the resonant bias voltage. That is, the transition moments μ_{13} and μ_{14} have almost the same magnitude but opposite sign (due to our choice of the phases of the eigenstates), so that the initial state $|\psi(t_0)\rangle$ is approximately expressed by the dressed state $|-\rangle$. Furthermore, as can be seen from the absorption spectra in Fig. 1(b), the eigenstates $|3\rangle$ and $|4\rangle$ are the bonding and antibonding electron states of the DQW system, which means that the dressed state $|-\rangle$ almost localizes in the NW. Contrary to this, the initial state in the off-resonant case is represented by a linear combination of the two dressed states, so that the wave packet continues to oscillate between two wells. The calculated time evolutions of $P_{NW}(t)$ and $P_{WW}(t)$ are shown in Fig. 5(a) in the resonant case of the bias voltage and (b) in the off-resonant case. These model calculations qualitatively reproduce our nu-

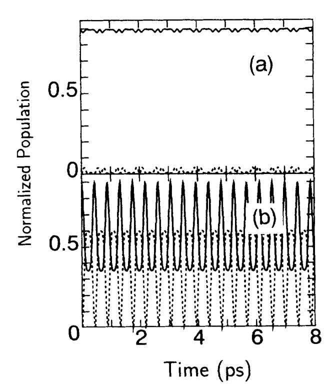

FIG. 5. Time evolution of the population $P_{NW}(t)$ (solid line) and $P_{WW}(t)$ (dashed line) calculated based on the degenerate-two-level model (a) in the resonant case and (b) off-resonant case of the bias voltage. The intensity, frequency, and phase of the cw laser are set as in Fig. 4.

merical results, i.e., the electron is localized by the irradiation of the cw laser in the resonant case, while no suppression of the tunneling can be seen in the off-resonant case.

There exists another problem for the suppression of electron tunneling concerning effects of the phase of the cw laser, which has been pointed out especially by Bavli and Metiu.[2] According to the degenerate-two-level model, the change of the phase is simply equivalent to a change of time origin, and the phase does not seem to play an important role. It should be noticed that in our analytical model, all the excited states are assumed to be resonantly prepared by the pump pulse [Eq. (13)]. In fact, if we assume that the pump pulse has large enough energy width to satisfy such a situation, then the numerical results show that the electron localization has no phase dependency. To realize such an ultrafast optical pumping, we need to consider the interband transition because the pump pulse must have a temporal width less than 100 fs. However, a femtosecond-laser pulse has already been successfully applied to DQW systems to observe electron oscillations between the wells.[4] Therefore, it is expected that by applying a cw laser to the electronic wave packet prepared by a femtosecond-laser pulse, the laser-induced suppression will be observed without the phase problem of the cw laser.

IV. CONCLUSION

We have numerically solved the Markoff master equation to see whether a cw laser can prevent an electron from tunneling in the biased asymmetric DQW structure. In our scheme, the excited electronic wave packet associated with the tunneling motion has been initially generated by the pump pulse. The results have shown that a cw laser of intensity 10^5 W/cm^2 causes laser-induced suppression of tunneling in the resonant case of the bias voltage, while no localization can occur in the off-resonant case. We have given a physical interpretation of these results based on the degenerate-two-level model. According to the numerical results, as well as the approximate analytical expression, if the electronic wave packet is initially prepared by a very short pump pulse whose energy width is large enough to cover the Stark broadened excited states, we expect that the laser-induced suppression of the tunneling can be observed without concern about the phase of the cw laser.

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[1] F. Grossmann, T. Dittrich, P. Jung, and P. Hnggi, Phys. Rev. Lett. **67**, 516 (1991); F. Grossmann, T. Dittrich, and P. Hnggi, Physica **175B**, 293 (1991); F. Grossmann and P. Hnggi, Z. Phys. B **85**, 315 (1991); F. Grossmann, P. Jung, T. Dittrich, and P. Hnggi, Europhys. Lett. **18**, 571 (1992).

[2] R. Bavli and H. Metiu, Phys. Rev. Lett. **69**, 1986 (1992).

[3] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures* (Les Editions de Physique, Les Ulis, 1988).

[4] K. Leo, J. Shah, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, W. Schäfer, and K. Köhler, Phys. Rev. Lett. **66**, 201 (1991); H. G. Roskos, M. C. Nuss, J. Shah, K. Leo, D. A. B. Miller, A. M. Fox, S. Schmitt-Rink, and K. Köhler, *ibid.* **68**, 2216 (1992); K. Leo, J. Shah, T. C. Damen, A. Schulze, T. Meier, S. Schmitt-Rink, P. Thomas, E. O. Göbel, S. L. Chuang, M. S. C. Luo, W. Schäfer, K. Köhler, and P. Ganser, IEEE J. Quantum Electron. **28**, 2498 (1992).

[5] P. C. M. Planken, I. Brener, M. C. Nuss, M. S. C. Luo, and S. L. Chuang, Phys. Rev. B **48**, 4903 (1993); M. S. C. Luo, S. L. Chuang, P. C. M. Planken, I. Grener, and M. C. Nuss, *ibid.* **48**, 11 043 (1993).

[6] W. H. Louisell, *Quantum Statistical Properties of Radiation* (Wiley, New York, 1973), Chap. 6; Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984), Chap. 2.

[7] Y. Ohtsuki and Y. Fujimura, J. Chem. Phys. **91**, 3903 (1989).

[8] S. Mukamel, Phys. Rep. **93**, 1 (1982); M. Schmutz, Z. Phys. B **30**, 97 (1978).

[9] A. Askar and A. S. Cakmak, J. Chem. Phys. **68**, 2794 (1978); R. Kosloff and D. Kosloff, *ibid.* **79**, 1823 (1983).

[10] Y. Ohtsuki, L. N. Pandey, and T. F. George, Chem. Phys. Lett. **196**, 619 (1992).

[11] L. N. Pandey and T. F. George, J. Appl. Phys. **69**, 2711 (1991).

[12] N. K. Rahman, Phys. Lett. A **54**, 8 (1975).

[13] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley, New York, 1990), pp. 264–268.