Nonradiative multiphonon transitions in semiconductor quantum dots

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A mathematical formalism suitable for a treatment of nonradiative multiphonon transitions in semiconductor quantum dots is developed. An analytical expression for the temperature dependence of the transition rate is derived.

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Recently an interest in multiphonon nonradiative transitions has emerged because they might be responsible for the fast intraband relaxation in semiconductor quantum dots.[1] Such transitions seem to be particularly important in lead chalcogenides nanocrystals, where competitive Auger-like processes are inhibited due to very similar electron and hole effective masses in lead salts.[1]

There are two principal methods employed in the theory of nonradiative transitions in solids and molecules. The first is based on the quasiclassical evaluation of the thermoactivated tunneling probabilities and was extensively reviewed by Ioselevich and Rashba[2] and by Abakumov *et al.*[3] The second is the multiphonon transition method first applied to nonradiative processes in the pioneering paper by Huang and Rhys[4] and developed in numerous consecutive studies (for a review of early work see Ref. 5). Both treatments were mainly applied to problems of (i) charge carrier capture by or emission from a deep center, and (ii) self-trapping or autolocalization of quasiparticles resulting in the formation of strongly coupled polarons. In these problems, one of the two electronic states is localized and the other is delocalized. For this class of problems the quasiclassical method was found to be much more flexible than the multiphonon transition one, especially in the case of the self-trapping problems for which the applicability of the multiphonon transition method was questioned.[2] The restrictions of the multiphonon transition method that prevent it from being used in these kinds of problems are the assumption of the parabolicity of the vibrational potential and the coincidence of the vibrational frequencies in the initial and final states. However, when both the final and the initial states of the electron or exciton are localized (which is the case of transitions between different states of an electron bound at a deep center or between different confined states of an exciton in a quantum dot) and the electron-phonon coupling is not too strong, these assumptions are justified and applied in the overwhelming majority of studies. In this paper we choose the multiphonon transition method because of its integrity and validity for the entire temperature range.

In the theory of Huang and Rhys,[4] the so-called Condon approximation was used involving an inconsistent and consequently impermissible application of the perturbation method in its treatment of the electron-phonon interaction diagonal in the electronic states.[6] This inconsistency was first noticed by Kovarskii[7] and later addressed by Ridley.[8] However, as it was recognized by Ridley[9] and Huang,[6] these studies imply "extremely laborious calculations"[9] and involve "very complicated mathematics."[6] A final clarification came with the paper by Huang,[6] who proved an important theorem about the matrix elements of the so-called nonadiabaticity operator (see also Ref. 9). This theorem states that the matrix elements of the nonadiabaticity operator nondiagonal in the electronic states are equal to the analogous matrix elements of the operator of the electron-phonon interaction. Below, we show that this theorem allows one to find the nonradiative transition rate using a rather simple mathematical formalism that is standard for quantum tunneling problems (see, e.g., Ref. 10). The result we obtain is found in the same order in the electron-phonon interaction nondiagonal in the electronic states as the original result by Huang and Rhys[4] but is free of the inconsistency introduced by the Condon approximation.

The Hamiltonian describing multiphonon nonradiative transitions between the two discrete electron (or exciton) levels, with unperturbed energies ε_1 and ε_2, can be represented as $H=H_0+V$. Here

$$H_0 = \sum_\alpha \omega_\alpha b_\alpha^\dagger b_\alpha \tag{1}$$

($\hbar=1$) is the phonon Hamiltonian in the electronic state with the energy ε_2 (Ref. 11) (the index α enumerates different phonon modes),

$$V = \left[\varepsilon_1 + \sum_\alpha \Delta^\alpha (b_\alpha^\dagger + b_\alpha)\right] c^\dagger c + \varepsilon_2 d^\dagger d + \sum_\alpha [\langle 1|A^\alpha|2\rangle c^\dagger d (b_\alpha^\dagger + b_\alpha) + \text{H.c.}], \tag{2}$$

$c^\dagger(d^\dagger)$ is the creation operator of the electronic state of the energy ε_1 (ε_2), $A^\alpha(b_\alpha^\dagger+b_\alpha)$ is the operator of the linear electron coupling with the phonon mode α, and $\Delta^\alpha \equiv \langle 1|A^\alpha|1\rangle - \langle 2|A^\alpha|2\rangle$.[12] In writing such a Hamiltonian, we go beyond the so-called Condon approximation employed in the pioneering paper by Huang and Rhys,[4,13] thus incorporating results of the later studies.[6,9] The nondiagonal matrix elements between the two electronic states result from nonadiabaticity of the electron-phonon system[9] and act as a trigger for the multiphonon nonradiative relaxation processes whose probabilities are determined by the overlap between vibrational wave functions in the upper and lower electronic states governed by the parameter Δ^α. Note that Eq. (2) is similar to the Hamiltonians used in quantum tunneling problems; it empha-

sizes connections between the multiphonon transition method and the method of the quasiclassical evaluation of the thermoactivated tunneling probabilities.

We write the electronic wave function as

$$\Psi(t) = [\hat{u}(t)c^{\dagger} + \hat{v}(t)d^{\dagger}]|0\rangle, \tag{3}$$

where $|0\rangle$ is the electronic vacuum and $\hat{u}(t)$, $\hat{v}(t)$ are the operators acting on the phonon subsystem. The wave function (3) satisfies the Schrödinger equation in the interaction representation [with respect to the Hamiltonian (1)],

$$i\frac{\partial\Psi(t)}{\partial t} = \hat{V}(t)\Psi(t), \tag{4}$$

with the initial conditions

$$\hat{u}(t=0) = 0, \quad \hat{v}(t=0) = 1. \tag{5}$$

Substituting Eqs. (1)–(3) into Eq. (4), we obtain the system of the coupled differential equations,

$$\begin{cases} i\dfrac{\partial\hat{u}}{\partial t} = [\varepsilon_1 + \hat{\mathcal{D}}(t)]\hat{u} + \displaystyle\sum_{\alpha}\langle 1|A^{\alpha}|2\rangle(b_{\alpha}e^{-i\omega_{\alpha}t} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t})\hat{v}, \\ i\dfrac{\partial\hat{v}}{\partial t} = \varepsilon_2\hat{v} + \displaystyle\sum_{\alpha}\langle 2|A^{\alpha}|1\rangle(b_{\alpha}e^{-i\omega_{\alpha}t} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t})\hat{u}, \end{cases} \tag{6}$$

where

$$\hat{\mathcal{D}}(t) = \sum_{\alpha}\Delta^{\alpha}(b_{\alpha}e^{-i\omega_{\alpha}t} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t}). \tag{7}$$

Let us introduce the auxiliary operator $\hat{u}_0(t)$ as the solution of the following Cauchy problem:

$$i\frac{\partial\hat{u}_0}{\partial t} = \hat{\mathcal{D}}(t)\hat{u}_0, \quad \hat{u}_0(t=0) = 1.$$

Using the Feynman procedure for disentangling exponential operators,[14] we obtain

$$\hat{u}_0(t) = \exp\left[\sum_{\alpha}\frac{\Delta^{\alpha}}{\omega_{\alpha}}(1 - e^{i\omega_{\alpha}t})b_{\alpha}^{\dagger} + \sum_{\alpha}\frac{\Delta^{\alpha}}{\omega_{\alpha}}(e^{-i\omega_{\alpha}t} - 1)b_{\alpha} + \sum_{\alpha}\frac{\Delta^{\alpha 2}}{\omega_{\alpha}^{2}}(i\omega_{\alpha}t - i\sin\omega_{\alpha}t)\right]. \tag{8}$$

The first of the equations (6) can then be written as

$$i\frac{\partial}{\partial t}(\hat{u}_0^{-1}\hat{u}) = \varepsilon_1\hat{u}_0^{-1}\hat{u} + \hat{u}_0^{-1}\sum_{\alpha}\langle 1|A^{\alpha}|2\rangle(b_{\alpha}e^{-i\omega_{\alpha}t} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t})\hat{v},$$

and the system (6) with the initial conditions (5) can be represented as a system of equivalent integral equations:

$$\begin{aligned} \hat{u}(t) &= -i\hat{u}_0(t)e^{-i\varepsilon_1 t}\int_0^t dt_0\, e^{i\varepsilon_1 t_0}\hat{u}_0^{-1}(t_0)\sum_{\alpha}\langle 1|A^{\alpha}|2\rangle \\ &\quad\times(b_{\alpha}e^{-i\omega_{\alpha}t_0} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t_0})\hat{v}(t_0), \\ \hat{v}(t) &= e^{-i\varepsilon_2 t} - ie^{-i\varepsilon_2 t}\int_0^t dt_0\, e^{i\varepsilon_2 t_0}\sum_{\alpha}\langle 2|A^{\alpha}|1\rangle \\ &\quad\times(b_{\alpha}e^{-i\omega_{\alpha}t_0} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t_0})\hat{u}(t_0), \end{aligned} \tag{9}$$

In the lowest order in $\langle 1|A^{\alpha}|2\rangle$, $\hat{v}(t) \approx e^{-i\varepsilon_2 t}$. Substituting this into the first of the equations (9), we obtain an explicit expression for $\hat{u}(t)$ to be substituted into the following expression for the transition rate:

$$W_{2\to 1} = \lim_{t\to\infty}\frac{1}{t}\frac{\mathrm{Tr}\{\exp[-\beta H_0]\hat{u}^{\dagger}(t)\hat{u}(t)\}}{\mathrm{Tr}\{\exp[-\beta H_0]\}} \equiv \lim_{t\to\infty}\frac{1}{t}\langle\hat{u}^{\dagger}(t)\hat{u}(t)\rangle, \tag{10}$$

where β is the inverse temperature in energy units. The average here is taken over the initial states of the phonon subsystem at $t=0$. It is assumed that these states are equilibrium states. We also have taken into account that at $t=0$, there is no interaction-induced re-normalization of the phonon states since the electronic level $|1\rangle$ is empty [see Eq. (5)]. In evaluating $W_{2\to 1}$, we encounter a correlation function of the form

$$\begin{aligned} U_{\alpha'\alpha}(t_1,t_0) &= \langle(b_{\alpha'}e^{-i\omega_{\alpha'}t_1} + b_{\alpha'}^{\dagger}e^{i\omega_{\alpha'}t_1})\hat{u}_0(t_1)\hat{u}_0^{\dagger}(t_0) \\ &\quad\times(b_{\alpha}e^{-i\omega_{\alpha}t_0} + b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t_0})\rangle. \end{aligned} \tag{11}$$

It is convenient to rewrite it as

$$U_{\alpha'\alpha}(t_1,t_0) = \frac{d^2}{da_1 da_2}\{\langle\exp[a_1 b_{\alpha'}e^{-i\omega_{\alpha'}t_1}]\exp[a_1 b_{\alpha'}^{\dagger}e^{i\omega_{\alpha'}t_1}]\hat{u}_0(t_1) \times\hat{u}_0^{\dagger}(t_0)\exp[a_2 b_{\alpha}e^{-i\omega_{\alpha}t_0}]\exp[a_2 b_{\alpha}^{\dagger}e^{i\omega_{\alpha}t_0}]\rangle\}|_{a_1=a_2=0}. \tag{12}$$

Using the following properties of arbitrary linear combinations A, B, C, D of the Bose operators b_{α} and $b_{\alpha}^{\dagger}$:

$$\begin{aligned} e^A e^B e^C e^D &= e^{A+B}e^{[A,B]/2}e^C e^D = \cdots \\ &= e^{A+B+C+D}e^{[A,B]/2}e^{[A+B,C]/2}e^{[A+B+C,D]/2}, \end{aligned}$$

$$\langle\exp C\rangle = \exp\left[\frac{1}{2}\langle C^2\rangle\right],$$

and Eq. (12), the function $U_{\alpha'\alpha}(t_1,t_0) = U_{\alpha'\alpha}(t_1 - t_0)$ can be easily evaluated. We obtain

$$U_{\alpha'\alpha}(\tau) = \exp\left\{\sum_{\mu}\frac{\Delta^{\mu 2}}{\omega_{\mu}^{2}}[i\omega_{\mu}\tau - i\sin\omega_{\mu}\tau + (2\bar{n}_{\mu}+1)(\cos\omega_{\mu}\tau - 1)]\right\} \times \left\{\frac{\Delta^{\alpha}\Delta^{\alpha'}}{\omega_{\alpha}\omega_{\alpha'}}[(\cos\omega_{\alpha}\tau - 1) - i(2\bar{n}_{\alpha} + 1)\sin\omega_{\alpha}\tau][(\cos\omega_{\alpha'}\tau - 1) - i(2\bar{n}_{\alpha'}+1)\sin\omega_{\alpha'}\tau] + \delta_{\alpha,\alpha'}[(2\bar{n}_{\alpha}+1)\cos\omega_{\alpha}\tau - i\sin\omega_{\alpha}\tau]\right\}, \quad (13)$$

where $\bar{n}_{\alpha}=(e^{\beta\omega_{\alpha}}-1)^{-1}$ is the Planckian factor. Introducing the Huang-Rhys factor $S=\Sigma_{\mu}(\Delta^{\mu}/\omega_{\mu})^2$, assuming that all the phonon modes have the same frequency ω_0 and using the identity

$$e^{z\cos\theta} = \sum_{l=-\infty}^{\infty} I_l(z)e^{il\theta},$$

where $I_l(z)$ is the modified Bessel function of order l, we can rewrite Eq. (13) in the form

$$U_{\alpha'\alpha}(\tau) = \left\{\frac{\Delta^{\alpha}\Delta^{\alpha'}}{\omega_0^2}[(\bar{n}+1)^2e^{-2i\omega_0\tau} + \bar{n}^2e^{2i\omega_0\tau} + 1 - 2\bar{n}(\bar{n}+1) - 2(\bar{n}+1)e^{-i\omega_0\tau} + 2\bar{n}e^{i\omega_0\tau}] + \delta_{\alpha,\alpha'}[(\bar{n}+1)e^{-i\omega_0\tau} + \bar{n}e^{i\omega_0\tau}]\right\} e^{iS\omega_0\tau}\sum_{l=-\infty}^{\infty} R_l e^{-il\omega_0\tau}, \quad (14)$$

where

$$R_l = \exp[-(2\bar{n}+1)S]\exp\left[\frac{\beta l\omega_0}{2}\right] I_l(2S\sqrt{\bar{n}(\bar{n}+1)}).$$

The summation over l can be replaced by the frequency integration

$$\sum_{l=-\infty}^{\infty} \rightarrow \frac{1}{\omega_0}\int_{-\infty}^{\infty} d\Omega, \quad l\omega_0 \rightarrow \Omega.$$

Then, substituting Eq. (14) into the expression for the transition rate following from Eq. (10):

$$W_{2\rightarrow 1} = \sum_{\alpha,\alpha'}\langle 1|A^{\alpha}|2\rangle\langle 2|A^{\alpha'}|1\rangle\int_{-\infty}^{\infty} d\tau\, U_{\alpha'\alpha}(\tau)e^{i(\varepsilon_2-\varepsilon_1)\tau}, \quad (15)$$

we obtain

$$W_{2\rightarrow 1} = \frac{2\pi}{\omega_0}\{Y[(\bar{n}+1)^2R_{p-2} + \bar{n}^2R_{p+2} + (1-2\bar{n}(\bar{n}+1))R_p - 2(\bar{n}+1)R_{p-1} + 2\bar{n}R_{p+1}] + Z[(\bar{n}+1)R_{p-1} + \bar{n}R_{p+1}]\}, \quad (16)$$

where $p\omega_0=\varepsilon_2-\varepsilon_1+\omega_0 S$ is the energy separation between the two electronic levels (the value of ω_0 should be adjusted so that p is an integer), p is the number of emitted phonons, $Z=\Sigma_{\alpha}|\langle 1|A^{\alpha}|2\rangle|^2$, and $Y=\Sigma_{\alpha}\Sigma_{\alpha'}\langle 1|A^{\alpha}|2\rangle\langle 2|A^{\alpha'}|1\rangle\Delta^{\alpha}\Delta^{\alpha'}/\omega_0^2$.

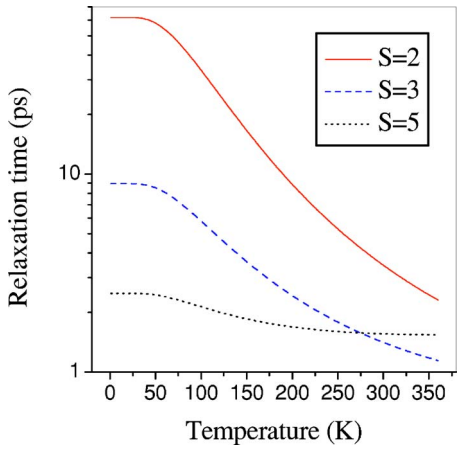

FIG. 1. (Color online) Temperature dependence of the relaxation time for the following values of parameters: $p\omega_0=165$ meV, $p=9$, $Z=(2.5\text{ meV})^2$, $Y=Z\cdot S$, and for different values of the Huang-Rhys factor: $S=2$ (solid line), $S=3$ (dashed line), and $S=5$ (dotted line).

In the limit $p\rightarrow\infty$, the asymptotic behavior of R_p can be reproduced quasiclassically.[15] Therefore, the quasiclassical picture[3,15] can be employed for a qualitative understanding of the results. However, the interplay between different terms in Eq. (16) can be important for certain choices of parameters, and in this aspect our result is different from that of Huang and Rhys.[4] Our formulation of the problem allows one to trace where this difference comes from[13] (see Appendix).

Note that some authors[16–18] totally neglect the matrix elements of the electron-phonon interaction diagonal in the electronic states. Within our formalism this means that for all phonon modes, $\Delta^{\alpha}=0$. In this case, $S=0$ and $R_l=\delta_{l,0}$. Substituting Eq. (14) into Eq. (15), one obtains in this limit the Fermi golden rule, which was the starting point of Refs. 16–18. Only processes involving emission or absorption of a single phonon are possible in this limit leading to a so-called phonon bottleneck. This once again emphasizes the importance of the proper treatment of the electron-phonon interaction diagonal in the electronic states, as it was highlighted by Huang.[6]

In order to illustrate our results, in Fig. 1 we plotted relaxation time corresponding to the rate given by Eq. (16) as a function of temperature for the following values of parameters: $p\omega_0=165$ meV, $p=9$, $Z=(2.5\text{ meV})^2$, $Y=Z\cdot S$, and for different values of the Huang-Rhys factor: $S=2$ (solid line), $S=3$ (dashed line), and $S=5$ (dotted line). At high temperatures, relaxation is due to thermal activation and relaxation time decreases rapidly with temperature increase. At low temperatures, relaxation is dominated by quantum tunneling and relaxation time changes slowly with a temperature increase. The stronger is electron-phonon coupling (the larger is S), the more efficient is quantum tunneling. Therefore, more pronounced temperature dependencies are obtained for smaller values of S.

In conclusion, we have developed a simple mathematical formalism, allowing one to treat nonradiative transitions within the multiphonon transition method. We derived an analytical expression for the temperature dependence of the transition rate under standard assumptions.

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APPENDIX: DERIVATION OF THE HUANG-RHYS FORMULA

In this appendix we show how the original formula of Huang and Rhys[4] can be derived within our formalism.[13] In this case the transition rate is given by an equation similar to Eq. (15),

$$\tilde{W}_{2\to 1} = \sum_{\alpha,\alpha'} B^{\alpha} B^{\alpha' *} \int_{-\infty}^{\infty} d\tau\, \tilde{U}_{\alpha'\alpha}(\tau) e^{i(\varepsilon_2-\varepsilon_1)\tau}, \qquad \text{(A1)}$$

with

$$\tilde{U}_{\alpha'\alpha}(t_1,t_0) = \langle (b^{\dagger}_{\alpha'} e^{i\omega_{\alpha' t_1}} - b_{\alpha'} e^{-i\omega_{\alpha' t_1}}) \hat{u}_0(t_1) \hat{u}^{\dagger}_0(t_0) \times (b_{\alpha} e^{-i\omega_{\alpha} t_0} - b^{\dagger}_{\alpha} e^{i\omega_{\alpha} t_0}) \rangle. \qquad \text{(A2)}$$

For $\tilde{U}_{\alpha'\alpha}(t_1,t_0) = \tilde{U}_{\alpha'\alpha}(t_1 - t_0)$, we obtain

$$\tilde{U}_{\alpha'\alpha}(\tau) = \exp\left\{ \sum_{\mu} \frac{\Delta^{\mu 2}}{\omega^2_{\mu}} [i\omega_{\mu}\tau - i \sin \omega_{\mu}\tau + (2\bar{n}_{\mu}+1)(\cos \omega_{\mu}\tau - 1)] \right\} \times \left\{ \frac{\Delta^{\alpha}\Delta^{\alpha'}}{\omega_{\alpha}\omega_{\alpha'}} [i \sin \omega_{\alpha}\tau + (2\bar{n}_{\alpha}+1)(1-\cos \omega_{\alpha}\tau)][i \sin \omega_{\alpha'}\tau + (2\bar{n}_{\alpha'}+1) \times (1-\cos \omega_{\alpha'}\tau)] + \delta_{\alpha,\alpha'} [(2\bar{n}_{\alpha}+1)\cos \omega_{\alpha}\tau - i \sin \omega_{\alpha}\tau] \right\} \qquad \text{(A3)}$$

[compare to Eq. (13)]. The substitution of Eq. (A3) into Eq. (A1) leads to the original result of Huang and Rhys,[4]

$$\tilde{W}_{2\to 1} = \frac{2\pi}{\omega_0} \{ \tilde{Y}[(\bar{n}+1)^2 R_{p-2} + \bar{n}^2 R_{p+2} + (2\bar{n}(\bar{n}+1) + (2\bar{n}+1)^2) R_p - 2(2\bar{n}+1)(\bar{n}+1) R_{p-1} - 2(2\bar{n}+1)\bar{n} R_{p+1}] + \tilde{Z}[(\bar{n}+1)R_{p-1} + \bar{n} R_{p+1}] \}, \qquad \text{(A4)}$$

where $\tilde{Z} = \Sigma_{\alpha} |B^{\alpha}|^2$, $\tilde{Y} = \Sigma_{\alpha}\Sigma_{\alpha'} B^{\alpha} B^{\alpha' *} \Delta^{\alpha}\Delta^{\alpha'} / \omega_0^2$. Comparing B^{α} to $\langle 1|A^{\alpha}|2\rangle$ we see[13] that this formula underestimates the transition rate by the factor of p^2.[6–9]

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[11] This implies that, on the configuration diagram, the minimum of the vibrational potential for the state $|2\rangle$ lies on the vertical axis.

[12] Strictly speaking, our definitions of the state $|2\rangle$ and eigenstates of the Hamiltonian H_0 imply that $\langle 2|A^{\alpha}|2\rangle = 0$. We keep $\langle 2|A^{\alpha}|2\rangle$ in the definition of Δ^{α} in order to emphasize that the corresponding Huang-Rhys factor (introduced later in the text) is different from the Huang-Rhys factors governing phonon-assisted *optical* transitions in quantum dots.

[13] The results of Ref. 4 can be reproduced if the following changes in the Hamiltonian (2) are made: $\langle 1|A^{\alpha}|2\rangle c^{\dagger} d(b^{\dagger}_{\alpha} + b_{\alpha}) \to B^{\alpha} c^{\dagger} d(b_{\alpha} - b^{\dagger}_{\alpha})$, where $B^{\alpha} \sim \omega_{\alpha} \langle 1|A^{\alpha}|2\rangle / (\varepsilon_2 - \varepsilon_1)$.

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