

Coupled quasiparticle-boson systems: The semiclassical approximation and discrete nonlinear Schrödinger equation

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The validity of the semiclassical approximation is studied for a system comprising one quasiparticle coupled to a boson degree of freedom. Using a two-site Holstein model as an example, it is shown that the semiclassical approximation becomes exact in a nontrivial adiabatic limit. Furthermore, in the model's polaron regime, there exists a hierarchy of time scales that rationalizes the quantum dynamics of the Holstein model. For the single-mode case considered, the discrete nonlinear Schrödinger equation is found to be valid only in a highly limited antiadiabatic regime.

We consider the widely relevant problem of the dynamics of one fermionic or bosonic quasiparticle moving on a lattice and interacting with boson degrees of freedom. One of the recently used approaches to solving the problem has employed the semiclassical approximation (SCA) which assumes that fluctuations of the quasiparticle and boson fields are uncorrelated, and that, consequently, expectation values of products of quasiparticle and boson operators may be factorized. A further approximation has assumed the expectation values of the boson operators to be slaved to those of the quasiparticle operators, and has led to the discrete nonlinear Schrödinger equation (DNLSE). The DNLSE constitutes the foundation of discrete self-trapping which has been invoked to describe localization of energy and its coherent transport, for example, in biological systems or molecular crystals. The quasiparticles are known there as "Davydov-Scott solitons."¹ Our interest in the present paper is in examining the validity of *both* the above approximations.

Questions concerning the applicability of the SCA and/or the DNLSE have been raised by several authors.²⁻⁵ The DNLSE follows unambiguously from the SCA (see, e.g., Ref. 3) in the antiadiabatic limit in which the boson time scale is the fastest one in the system. However, serious doubts have been cast on the possibility of the DNLSE being a consequence of full quantum evolution.^{2,4,5} A thorough study of the quantum system in the antiadiabatic limit has resulted^{4,5} in the conclusion that the DNLSE has a poor range of validity.⁷ This limit has attracted the main focus precisely because the derivation of the DNLSE from the SCA is trivial in this limit.^{1,3,6}

Because many of the results on the semiclassical theory in the literature are concerned with dimers,^{3-5,8} we consider here the Holstein model of the quantum dimer,

$$H = -V\sigma_1 + \lambda\varphi\sigma_3 + \frac{1}{2}\epsilon_0(\pi^2 + \varphi^2), \quad (1)$$

where the Pauli operators are defined in terms of the quasiparticle operators as $\sigma_1 = c_1^\dagger c_2 + c_2^\dagger c_1$, $\sigma_2 = -i(c_1^\dagger c_2 - c_2^\dagger c_1)$, and $\sigma_3 = c_1^\dagger c_1 - c_2^\dagger c_2$. The dimensionless conjugate operators φ and π , obeying the commutation relation

$[\varphi, \pi] = i$, describe a harmonic oscillator with energy $\epsilon_0 = \hbar\omega_0$. The quasiparticle tunnels with a matrix element V (>0) between sites 1 and 2 and interacts with the boson mode φ , the interaction strength being specified by the coupling constant λ . Our interest here is exploring the consequences of this one-mode model at zero temperature.⁹ The properties of Eq. (1) are controlled by two dimensionless parameters χ/V (nonlinearity) and ϵ_0/V (adiabaticity), where $\chi \equiv 2\lambda^2/\epsilon_0$. However, in the adiabatic ($\epsilon_0/V \ll 1$) and the antiadiabatic ($\epsilon_0/V \gg 1$) regimes, the physics of the semiclassical model is determined by χ/V . As a consequence our basic philosophy here is to compare the quantum mechanical and semiclassical dynamics at a given χ/V , because this parameter characterizes the low-energy physics outside the regime $\epsilon_0/V \sim 1$. Specifically, by varying ϵ_0 and with $\chi = \text{const}$, the possible regimes of validity of the semiclassical theory become much more meaningful since the physics that the quantum and semiclassical theories are expected to capture remains the same.

Our main results are threefold. First, we give a simple argument supporting the earlier finding^{4,5,10} that the antiadiabatic limit, leading to the DNLSE according to the usual semiclassical arguments, does *not* produce the correct low-energy physics of the Holstein model. Second, and most importantly, we show that the semiclassical approximation does become *exact* in the nontrivial (adiabatic) limit: $\epsilon_0, \lambda \rightarrow 0$ with $\chi = \text{const}$. Third, we identify a hierarchy of time scales, which rationalize the quantum dynamics in the adiabatic regime and are simply understood in terms of memory functions for the system. In particular, we demonstrate the existence of a characteristic time τ_h up to which semiclassical dynamics is a good approximation. In the polaron-formation parameter regime, τ_h coincides with the polaron-tunneling time τ_T . In our analysis, we emphasize the distinction between the DNLSE and the SCA.¹¹

Hamiltonian (1) leads^{3,12} to the following coupled Heisenberg equations of motion for the quasiparticle and boson operators:

$$\partial_\tau \sigma_1 = -2\lambda\varphi\sigma_2, \quad (2a)$$

$$\partial_\tau \sigma_2 = 2V\sigma_3 + 2\lambda\varphi\sigma_1, \quad (2b)$$

$$\partial_\tau \sigma_3 = -2V\sigma_2, \quad (2c)$$

$$\partial_\tau \varphi = -\epsilon_0 \pi, \quad (2d)$$

$$\partial_\tau \pi = \epsilon_0 \varphi + \lambda \sigma_3, \quad (2e)$$

where $\tau = t/\hbar$ is the scaled time. The SCA consists of replacing φ and π by c numbers in the expectation values of the Heisenberg operators. This is equivalent to the assumption that the *quantum fluctuations* of the quasiparticle and the boson fields are uncorrelated. The DNLSE results from the further assumption that the *expectation values* are completely correlated (slaved). This leads to an instantaneous boson-mediated quasiparticle-quasiparticle interaction:^{1,3,6}

$$\partial_\tau \langle \vec{\sigma} \rangle = \vec{\Omega}_\sigma \times \langle \vec{\sigma} \rangle, \quad (3)$$

where $\vec{\sigma}$ is a vector whose components are the three Pauli operators and $\vec{\Omega}_\sigma = (-2V, 0, -\chi \langle \sigma_3 \rangle)$, which also incorporates the nonlinear term.

To test the DNLSE validity in the antiadiabatic limit, consider Eqs. (2), and take the limit $\epsilon_0, \lambda \rightarrow \infty$ such that $\chi \equiv 2\lambda^2/\epsilon_0 = \text{const}$. The bosonic degree of freedom can be integrated out of the quantum evolution, and

$$\lambda \varphi(\tau) \rightarrow \lambda [\varphi(0) \cos \epsilon_0 \tau + \pi(0) \sin \epsilon_0 \tau] - \frac{1}{2} \chi \sigma_3(\tau), \quad (4)$$

where rapidly oscillating components (relative to $2V$) have been omitted. Substituting Eq. (4) in Eq. (2a) shows clearly that nonlinearities disappear in the limit of no retardation and that the dynamics of the quasiparticle is linear. Thus, in the antiadiabatic limit with a fixed χ , the semiclassical approximation, $\langle \sigma_j \varphi \rangle \approx \langle \sigma_j \rangle \langle \varphi \rangle$, leads to an incorrect result because $2\lambda \varphi \sim -\chi \sigma_3$ and because $\sigma_j \sigma_k = i \sigma_l$ (j, k, l cyclic) in *one*-quasiparticle Fock space (for both bosons and fermions). Even modifying the form of coupling of bosons to the quasiparticle will not produce the DNLSE-type diagonal nonlinear self-interaction. This observation applies equally well for systems with arbitrary number of sites or phonon modes.¹⁰ For more than one quasiparticle, statistics becomes important and, for bosons, nonlinear terms (but not the DNLSE) survive. It is important to appreciate that there *are* antiadiabatic regimes (i.e., finite ϵ_0), where the DNLSE approximately reproduces the low-energy physics of the full quantum mechanical system (2) for sufficiently short times ($\tau \ll \tau_h$). We now turn to this issue of the time domain.

Because no analytical solutions exist for semiclassical and quantum time-evolution equations in the general case, we resort to numerical methods. We focus on the time evolution of $\langle \sigma_3(\tau) \rangle$ and also monitor the correlation function

$$G(\tau) = \langle \{A(\tau) - \langle A(\tau) \rangle\} \{ \sigma_1(\tau) - \langle \sigma_1(\tau) \rangle \} \rangle, \quad (5)$$

which measures the effect of quantum fluctuations (i.e., correlations in the fluctuations of the quasiparticle and boson fields) neglected in the SCA. Here $A \equiv (\lambda/V)\varphi$. The quantum expectation values are computed in a state given by the projected part of the ground state of Hamiltonian (1) that has the quasiparticle at site 1. The semiclassical equations are solved with the corresponding initial condition (see below). We emphasize that $G(\tau)$ is precisely one of the two quantum mechanical “driving forces” contained in Eqs. (2) that must

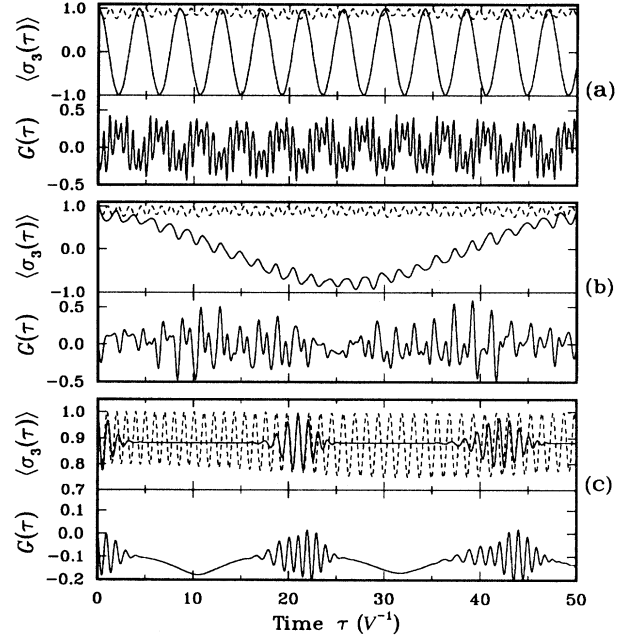


FIG. 1. The exact quantum (solid line) and the semiclassical (dashed line) time evolution of the quasiparticle occupation difference $\langle \sigma_3(\tau) \rangle$, for $\chi = 2\lambda^2/\epsilon_0 = 6V$. Also shown is the quantum correlation function $G(\tau)$. The other parameters are (a) $\epsilon_0 = 10V$, $\lambda = \sqrt{30}V$; (b) $\epsilon_0 = V$, $\lambda = \sqrt{3}V$; and (c) $\epsilon_0 = V/10$, $\lambda = \frac{1}{10}\sqrt{30}V$. Initially, the quasiparticle is assumed to be at site 1; for the detailed initial conditions, see the text following Eq. (15).

be added to the semiclassical equations to make them exact: $\langle A(\tau) \sigma_1(\tau) \rangle = \langle A(\tau) \rangle \langle \sigma_1(\tau) \rangle + G(\tau)$.

In Fig. 1, we summarize a comparison between the semiclassical and fully quantum evolutions of $\langle \sigma_3(\tau) \rangle$ for fixed χ and varying ϵ_0 and λ . Corresponding to the quantum mechanical initial state, we have assumed here that $\langle \varphi(0) \rangle = -\lambda/\epsilon_0$, $\langle \pi(0) \rangle = 0$, $\langle \sigma_3(0) \rangle = 1$, and the other quasiparticle expectation values at $\tau = 0$ are zero. These natural initial values imply $\langle \partial_\tau \pi(0) \rangle = 0$. Our choice of $\chi = 6V$ is large enough to yield self-trapping in the semiclassical case. These results show that the semiclassical description completely fails for $2V, \lambda \ll \epsilon_0$ and $4V \lesssim \chi$, where the quantum dynamics is essentially linear as discussed above. In contrast, for $2V \ll \epsilon_0 \lesssim \lambda$ (implying self-trapping) there is again a region where the semiclassical approach—and indeed the DNLSE—yields an approximate description of the exact quantum mechanical dynamics as shown earlier.⁵ With decreasing ϵ_0 , the quantum dynamics leads to the formation of a new composite particle, the polaron, where the motion of the quasiparticle is slaved to the lattice dynamics. A characteristic signature of this nonlinear dynamics is the appearance of a very small energy scale ϵ_T which corresponds to the reduced polaron bandwidth. The semiclassical description mimics the polaron formation by exhibiting permanent self-trapping at one of the sites, but it cannot capture polaron *tunneling*. As ϵ_0 is further decreased, the period of tunneling τ_T becomes exponentially large and gives only a small correction at $\tau \lesssim \tau_T$. Figure 1 shows that correlations between quantum fluctuations are very important at the onset of the polaron formation. In the strong polaron regime

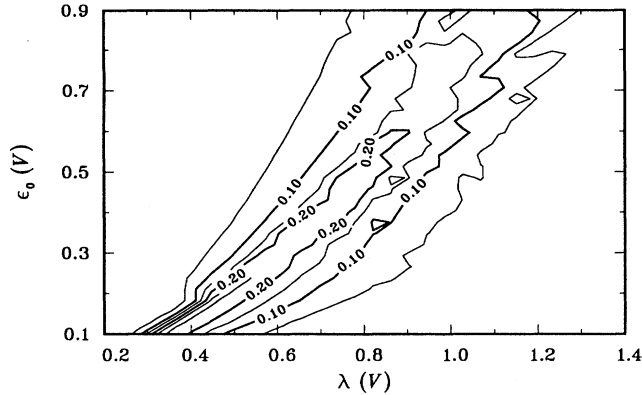


FIG. 2. The magnitude of $\sum_{|\epsilon| \leq \epsilon_m} G(\epsilon)$ on the ϵ_0 - λ plane, shown as a contour plot. Here ϵ_m is the energy of the largest Fourier amplitude of $\langle \sigma_3(\tau) \rangle$. The increment between contour levels is 0.05. Note that the ϵ_0 and λ axes are plotted beginning from a nonzero value.

($\epsilon_T \ll \epsilon_0, 2V$) the correlations become smaller, although they produce on average a continuous quantum force (represented by G) at semiclassical turning points, which drives the system between sites 1 and 2 periodically. During the tunneling processes G changes sign (“direction”), being zero on average. Note again that, in this regime, the quasiparticle and the boson degrees of freedom are strongly correlated. These polaronic correlations remain large for all times τ . Of course $G(\tau)$ is negligibly small in the weak-coupling regime.

In Fig. 2, we plot the absolute value of $\sum_{|\epsilon| \leq \epsilon_m} G(\epsilon)$, where $G(\epsilon)$ is the Fourier amplitude of $G(\tau)$ and ϵ_m is the energy of the largest Fourier amplitude of $\langle \sigma_3(\tau) \rangle$. In the tunneling regime, $\epsilon_m = \epsilon_T$. This figure is very useful for assessing the importance of correlations between quantum fluctuations of the quasiparticle and the boson degree of freedom. In the region where $\sum_{|\epsilon| \leq \epsilon_m} G(\epsilon)$ is large, the semiclassical approximation fails badly, and where it is small, the semiclassical approximation works well. Figure 2 serves therefore as a phase diagram for the validity of the semiclassical theory. Interestingly, the trajectories $\chi = \text{const}$ on the ϵ_0 - λ plane follow approximately the topographic features of $\sum_{|\epsilon| \leq \epsilon_m} G(\epsilon)$, e.g., the ridge in Fig. 2. Thus, it is natural to explore the limit $\epsilon_0, \lambda \rightarrow 0$ with $\chi = \text{const}$ (as above). We can reach a similar picture by studying temporal variations of $G(\tau)$.

The ground and first-excited states of the Hamiltonian (1) can be readily constructed both in the adiabatic regime ($\epsilon_0 \ll V$), for $g^2 \gg V/\epsilon_0$, and in the antiadiabatic regime ($\epsilon_0 \gg V$), for arbitrary $g \equiv \lambda/\epsilon_0$. They are simply the “polaron-tunneling” states $|\Psi_{\pm}\rangle = (|R\rangle \pm |L\rangle)/\sqrt{2}$. The states $|L\rangle = |1, -g\rangle$ and $|R\rangle = |2, g\rangle$ describe the polaron (i.e., the quasiparticle and its correlated cloud of bosons) located at site 1 and 2, respectively. Here $|g\rangle = e^{-ig\pi}|0\rangle$ is a phonon coherent state. The states $|\Psi_{\pm}\rangle$ are split by the energy $\epsilon_T = 2V \exp(-g^2)$, which is the polaron-tunneling energy and determines the tunneling period: $\tau_T = 2\pi/\epsilon_T$. In this limit many properties of the system can be computed analytically. For example, consider $\langle \sigma_3(\tau) \rangle = \langle L | \sigma_3(\tau) | L \rangle$ in the adiabatic regime $\epsilon_0 \ll V$ with $g^2 \gg V/\epsilon_0$. For short times,

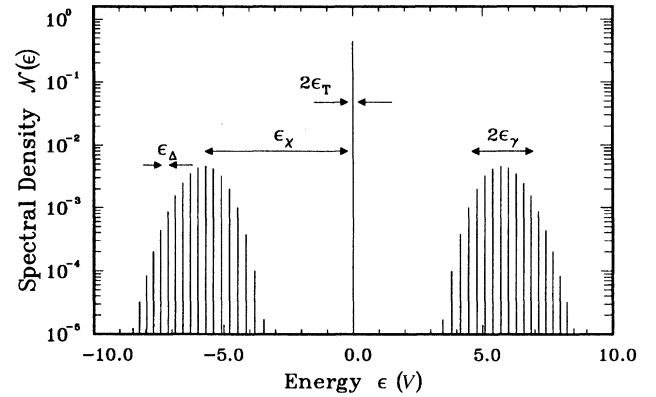


FIG. 3. The spectral density $\mathcal{N}(\epsilon)$ of $\langle \sigma_3(\tau) \rangle$, for $\epsilon_0 = V/10$, and $\lambda = \frac{1}{10}\sqrt{30}V$ ($\chi = 6V$). The energy scales shown are $\epsilon_\chi \approx 6V$, $\epsilon_\gamma \approx 1.1V$, $\epsilon_\Delta \approx 0.3V$, and $\epsilon_0 = 1.1 \times 10^{-10}V$.

$$\langle \sigma_3(\tau) \rangle = (1 + \alpha^{-2})^{-1} + (1 + \alpha^2)^{-1} \cos \epsilon_\chi \tau, \quad (6)$$

where $\epsilon_\chi = \sqrt{\chi^2 + 4V^2}$ and $\alpha = \chi/2V$. Equation (6) has been obtained previously⁵ in the *antiadiabatic* limit. We thus see that $\chi/2V$ emerges as the key parameter in both the adiabatic and antiadiabatic limits. The signal shows fast oscillations with period $\tau_\chi = 2\pi/\epsilon_\chi$ (the generalized Rabi frequency in quantum optics¹³). Additional features appear at times $\tau \gg \tau_\chi$, including the slow polaron tunneling motion, evident as an oscillatory amplitude of the first term in Eq. (6).

Since the oscillator mass μ appears in the parameters as $\lambda \propto \mu^{-1/4}$ and $\epsilon_0 \propto \mu^{-1/2}$, the adiabatic limit $\epsilon_0, \lambda \rightarrow 0$ with $\chi = \text{const}$, can also be regarded as the limit of infinite oscillator mass, $\mu \rightarrow \infty$. In this massive oscillator limit, the semiclassical result for $\langle \sigma_3(\tau) \rangle$ coincides with the quantum mechanical one, Eq. (6). Moving away from the limit, the agreement is only valid for times $\tau \ll \tau_T$ (see below). Denoting by τ_h the characteristic time after which the semiclassical dynamics deviates from the quantum mechanical one due to quantum fluctuations, it is natural to define $\tau_h \sim \tau_T$: here the deviation is signified by the quantum mechanical tunneling between degenerate semiclassical minima. As $g \rightarrow \infty$, $\tau_h \rightarrow \infty$.

As $\epsilon_0, \lambda \rightarrow 0$ such that $\chi = \text{const}$ (i.e., $\mu \rightarrow \infty$), the analytical result (6) becomes increasingly accurate. This is evidenced by the agreement between the semiclassical and quantum results; see Fig. 1(c). In particular, the average value of $\langle \sigma_3 \rangle \approx 0.90$ at times $\tau \ll \tau_T$ is essentially the same for both cases [cf. Fig. 1(c) and Eq. (6)]. Nevertheless, striking differences remain because of two additional time scales, τ_γ and τ_Δ . As a consequence of a finite width $2\epsilon_\gamma$ of the spectral features at the energy ϵ_χ , the quantum fluctuations produce a Gaussian decay, $e^{-(\tau/\tau_\gamma)^2}$, of the signal to its average value in a characteristic time $\tau_\gamma = 2/\epsilon_\gamma$. In the $\mu \rightarrow \infty$ limit, τ_γ can be shown to diverge as $\tau_\gamma \approx \lambda^{-1}$. However because the spectrum is discrete and composed of nearly equally spaced peaks, the signal shows (generally aperiodic) revivals (quantum recurrences) after times $\sim \tau_\Delta$. The time τ_Δ is of order of the boson period: $\tau_\Delta \sim 2\pi/\epsilon_0$. In Fig. 3, we illustrate these time (energy) scales by plotting the exact spectral density of $\langle \sigma_3(\tau) \rangle$ with the same set of parameters as in Fig. 1(c). Note the hierarchy of times: $\tau_\chi = \mathcal{O}(\mu^0)$,

$\tau_\gamma = \mathcal{O}(\mu^{1/4})$, $\tau_\Delta = \mathcal{O}(\mu^{1/2})$, and $\tau_T = \mathcal{O}(e^{c\sqrt{\mu}})$ (c a positive constant). That only τ_χ remains finite as $\mu \rightarrow \infty$ explains why the semiclassical approximation becomes exact in this limit. The slow dispersion of the bursts in Fig. 1(c) continues at longer times and we find numerically that the long-time behavior of $G(\tau)$ [and $\langle \sigma_3(\tau) \rangle$] has additional multiple time scales due to a distribution of τ_Δ 's (cf. semiclassical results),^{3,12} culminating in polaron tunneling.

Memory functions of the generalized-master-equation theory obtained for exciton motion¹⁴ in the many-mode case of Eq. (1) and applied for the explanation of charge mobility observations in molecular crystals,¹⁵ offer a suitable framework for understanding various time scales appearing in the quantum problem. The quantum dynamics is solved by projection operators giving, for example,

$$\partial_\tau \langle \sigma_3(\tau) \rangle + 2 \int_0^\tau d\tau' \mathcal{W}(\tau - \tau') \langle \sigma_3(\tau') \rangle = 0. \quad (7)$$

The memory function \mathcal{W} has been obtained¹⁴ perturbatively in the strong-coupling limit ($\chi/V \gg 1$): for the single-mode case,

$$\mathcal{W}(\tau) = 2V^2 e^{-2g^2(1 - \cos \epsilon_0 \tau)} \cos(2g^2 \sin \epsilon_0 \tau). \quad (8)$$

For short times, \mathcal{W} exhibits rapid oscillations of frequency $\chi \equiv 2g^2 \epsilon_0$ which approximately equals $2\pi/\tau_\chi$ for strong coupling ($\chi \gg 2V$). The decay of \mathcal{W} occurs in a time $1/g\epsilon_0$ which also equals τ_γ in this limit. The fact that the integral of the memory function during this period is nearly zero, and that the memory function is exponentially suppressed for large g to near-zero value explains the “silent run” observed for $\langle \sigma_3(t) \rangle$ [see Fig. 1(c)]. The revival of the memory function after the period $2\pi/\epsilon_0$ of the phonon oscillation is responsible for the quasiparticle revival. The limit $\mu \rightarrow \infty$ reduces $\mathcal{W}(\tau)$ to $2V^2 \cos \chi \tau$. Equation (7) then leads

to Eq. (6) trivially, establishing that the semiclassical description is exact in this adiabatic limit. Moreover, in the strong-coupling limit and for finite μ , the memory-function formalism correctly captures the essence of the four time scales, τ_χ , τ_γ , τ_Δ , and τ_T .¹⁶

We have studied here the quantum and semiclassical correspondence of the *low-energy* dynamics of a coupled quasiparticle-boson problem. In the polaron-tunneling regime, the polaron binding energy gives one characteristic energy scale. Our results are necessarily energy-scale dependent, but similar numerical study can easily be extended to higher energies. We also remark that *spatially extended* systems (for which quasiparticle tunneling generalizes to band formation) could provide intriguing phenomena in the form of length scales describing correlated quasiparticle and lattice distortions, lattice spacing, etc. We speculate, for example, that a characteristic length l_h appears, at which the semiclassical dynamics breaks down.¹⁷

Finally, three cautionary remarks are in order and lead to important directions for future research: (i) With increasing system size, additional time and length scales are introduced that may change conditions for the validity of the SCA and DNLSE. (ii) It is physically important to include coupling to many phonon modes to fully describe memory and a heat bath.^{4,9} (iii) In assessing the utility of various approximations, we must be particularly sensitive to their manifestations in physical observables: Not all quantum retardation effects are equally significant physically. However, for instance in polaron contexts optical and structural experimental probes are becoming sufficiently precise that it is now necessary to go beyond semiclassical or adiabatic approximations for an understanding of the data.¹⁸

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⁷We note in passing a complementary development in quantum optics, where similar limitations have also been found for the “neoclassical” theory of spontaneous emission. See, e.g., G. Agarwal, *Quantum Statistical Theories of Spontaneous Emission and Their Relation to Other Approaches* (Springer-Verlag, Berlin, 1974), and references therein.

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¹⁶The tunneling time τ_T can be deduced by considering the Laplace transform $\mathcal{W}(s)$ in the limit $s \rightarrow 0^+$.

¹⁷One might conjecture that l_h is of the order of one lattice spacing. However, length-scale competition may provide more complex situations.

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