

Change of Decoherence Scenario and Appearance of Localization due to Reservoir Anharmonicity

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Although coupling to a super-Ohmic bosonic reservoir leads only to partial dephasing on short time scales, exponential decay of coherence appears in the Markovian limit (for long times) if anharmonicity of the reservoir is taken into account. This effect not only qualitatively changes the decoherence scenario but also leads to localization processes in which superpositions of spatially separated states dephase with a rate that depends on the distance between the localized states. As an example of the latter process, we study the decay of coherence of an electron state delocalized over two semiconductor quantum dots due to anharmonicity of phonon modes.

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Decoherence of open quantum systems has become one of the central issues of the quantum theory. On one hand, erasure of phase information with respect to a certain basis of states may bring classical behavior out of quantum evolution [1–3]. On the other hand, the ability to control the states of quantum systems may open the way to novel applications, like quantum information processing [4,5], provided that quantum coherence is maintained over a sufficiently long time. Therefore, understanding and reducing decoherence is also of practical importance.

Of special interest is a class of models that allows for erasure of phase information without transitions between the selected basis states. Such a pure dephasing process is an essential ingredient of a measurement, with the basis states selected by the coupling between the measurement device and its environment (the pointer basis) [6] and determining the physical meaning of the measurement [3]. A simple model leading to this kind of behavior is composed of a two-level system and a bosonic bath with the coupling between these two subsystems linear in the bosonic operators and commuting with the Hamiltonian of the system (independent boson model [7]). Pure dephasing effects are also relevant for the short time dynamics of confined carrier states in semiconductor quantum dots (QDs) [8,9]. A class of systems described by such a model (including confined carriers in QDs interacting with phonons) shows only partial dephasing with a finite asymptotic level of coherence [8,10]. This feature of real systems is essential for the possibility of explaining the classical nature of measurement results in terms of dephasing (“einselection”) [3], which is based on the expectation that dephasing processes are asymptotically exponential, as observed in some formal models [11].

The purpose of this Letter is to show that the important property of complete or partial asymptotic dephasing depends not only on the system-reservoir coupling but also on the properties of the reservoir itself. In particular, no real reservoir may be strictly harmonic, one obvious reason being the need to restore the equilibrium, i.e., a sufficient

level of ergodicity. Here, this is accounted for by including an anharmonic coupling between the bosonic modes. This model closely corresponds to real properties of solid-state phonon reservoirs, where also parameters of the model may be inferred from experiments.

The combination of system-reservoir coupling with reservoir anharmonicity leads to a dephasing effect that, for spatially localized systems, may be understood in terms of collisional decoherence: Because of their coupling to the system, reservoir modes undergo a shift of their equilibrium positions which depends on the system state, forming a coherent displacement field [12,13]. If the modes are coupled by anharmonicity the displacement field acts as a scattering potential for other reservoir modes. Since the displacement is state dependent, each scattering event extracts a certain amount of information on the system state, gradually leading to complete dephasing. This is the first essential result of the present work. The second result is related to the special situation when the system is in a superposition of two states corresponding to distinct positions in real space. In such a case, scattering of reservoir modes leads to vanishing of the coherence between the two distant states. This process turns a genuine quantum-delocalized system state into a mixture of two classical-like localized states and is hence referred to as localization [14–16]. It is shown, for a specific model of carrier-phonon interaction in semiconductor QDs, that the rate of localization grows with the separation between the two spatial locations. This confirms the intuitive expectation that coherence on large distances should be more fragile.

Let us consider the Hamiltonian $H = H_0 + H_1$, where H_0 is the two-level independent-boson Hamiltonian

$$H_0 = -\frac{1}{2}E\sigma_z + \sum_k \hbar\omega_k b_k^\dagger b_k + \frac{1}{2}\sigma_z \sum_k F_k^* (b_k + b_{-k}^\dagger), \quad (1)$$

with $F_k^* = F_{-k}$, and H_1 describes the third order anharmonic coupling between various phonon modes

$$H_1 = \frac{1}{6} \sum_{k_1 k_2 k_3} w_{k_1, k_2, k_3} \delta_{k_1 + k_2 + k_3 = 0} A_{k_1} A_{k_2} A_{k_3}, \quad (2)$$

where $A_k = b_k + b_{-k}^\dagger$ and the anharmonic constants $w_{k_1, k_2, k_3} = w_{-k_1, -k_2, -k_3}^*$ are symmetric under permutation of indices. The polarization (branch) index of the boson modes is implicit in k .

We define the unitary operator

$$\mathbb{W} = |0\rangle\langle 0| \otimes W^\dagger + |1\rangle\langle 1| \otimes W, \quad (3)$$

where $W = \exp[(1/2)\sum_k g_k^* b_k - \text{H.c.}]$, $g_k = F_k/(\hbar\omega_k)$. In terms of the new operators $\beta_k = \mathbb{W}b_k\mathbb{W}^\dagger = b_k + \frac{1}{2}\sigma_z g_k$, the Hamiltonian H_0 is diagonal, $H_0 = -(1/2)E\sigma_z + \sum_k \hbar\omega_k \beta_k^\dagger \beta_k$. Using the exact diagonalization by the operator \mathbb{W} one can find the evolution of the off-diagonal element of the reduced density matrix (in the original basis), $|\rho_{01}(t)| = |\rho_{01}(0)| \exp[-2 \int d\omega \times \coth(\hbar\omega/k_B T) J(\omega) \sin^2(\omega t/2)/(\hbar\omega)^2]$, where $J(\omega) = \sum_k |F_k|^2 \delta(\omega - \omega_k)$. For spectral densities sufficiently regular at low frequencies, $J(\omega) \sim \omega^n$, $n \geq 2$ (super-Ohmic reservoirs) and for gapped reservoirs ($J(\omega) = 0$ around $\omega = 0$), $|\rho_{01}|$ reaches a finite asymptotic value (corresponding to partial dephasing) [8,10]. On the other hand, if $J(\omega) \sim \omega$ (Ohmic reservoirs), ρ_{01} decays exponentially for long times [17]. It turns out, however, that Ohmic independent boson models show infrared divergences that cause problems on the formal level [10]. Below we will see that dephasing generated by a super-Ohmic reservoir becomes complete (exponential at long times) if the reservoir is anharmonic.

In terms of the transformed operators β the anharmonic Hamiltonian H_1 becomes

$$\begin{aligned} H_1 = & \frac{1}{6} \sum_{k_1 k_2 k_3} w_{k_1, k_2, k_3} \delta_{k_1 + k_2 + k_3 = 0} \\ & \times (\mathcal{A}_{k_1} \mathcal{A}_{k_2} \mathcal{A}_{k_3} - 3\sigma_z g_{k_1} \mathcal{A}_{k_2} \mathcal{A}_{k_3} \\ & + 3g_{k_1} g_{k_2} \mathcal{A}_{k_3} - \sigma_z g_{k_1} g_{k_2} g_{k_3}), \end{aligned}$$

where $\mathcal{A}_k = \beta_k + \beta_{-k}^\dagger$. The first term is the anharmonic coupling between the new modes, the third describes a shift of the oscillator equilibria, the fourth is a shift of the energy levels that may be included in the energy E .

Of interest here is the second term that describes two-phonon absorption, emission, and scattering. Only the latter may lead to energy-conserving processes that do not involve real transitions between system states. Using commutation relations and symmetries of the anharmonic coefficients one may write the relevant (scattering) part of the anharmonic Hamiltonian

$$\begin{aligned} H_1^{(s)} = & -\sigma_z \sum_{k, q} w_{q-k, -q, k} g_{q-k} \\ & \times [\beta_q^\dagger \beta_k - \delta_{q, k} n_k + \delta_{q, k} (n_k + 1/2)]. \quad (4) \end{aligned}$$

The last term may again be included into the energy E (it

vanishes in the limit of infinite reservoir volume). It should be noticed that Eq. (4) is nondiagonal in phonon modes which allows for real phonon scattering processes (unlike the model of Ref. [18]). A similar scattering Hamiltonian may be obtained by including higher exciton levels into a purely harmonic model [19].

It is known that scattering on a heavy Brownian particle leads to decoherence and localization of the quantum state of the latter [14–16]. In order to see that the same is true in the present case of scattering of bosonic modes on a two-state quantum system and to extract the corresponding long-time behavior we write the evolution equation with the interaction Hamiltonian (4). Assuming that the time scales of the dephasing process are longer than those related to the reservoir memory and initial dephasing one may consistently describe the long-time dynamics in the Markov limit. From the resulting Lindblad equation one finds the solution describing exponential pure dephasing at long times with the rate [20]

$$\begin{aligned} \frac{1}{T_2} = & 2 \int_{-\infty}^{\infty} dt \langle B^\dagger(t) B(0) \rangle \\ = & 4\pi \sum_{k, q} |w_{q-k, -q, k}|^2 |g_{q-k}|^2 n_k (n_k + 1) \delta(\omega_q - \omega_k), \end{aligned} \quad (5)$$

where $B(t) = \sum_{k, q} w_{q-k, -q, k} g_{q-k} [\beta_q^\dagger \beta_k e^{i(\omega_q - \omega_k)t} - \delta_{q, k} n_k]$ are reservoir operators with vanishing equilibrium average. The dephasing rate is finite at $T > 0$, which should be contrasted with the harmonic case, where the dephasing effect vanishes in the Markov limit. Thus, reservoir anharmonicity has qualitatively changed the decoherence properties of the system.

The rate given by Eq. (5) is consistent with the scattering picture described in the introduction. A boson scatters from the state k to q . The scattering amplitude is proportional to the magnitude of the displacement field, governed by the system-reservoir coupling constants g_{q-k} , and to the anharmonic coupling $w_{q-k, -q, k}$. The momentum transfer in such an event cannot exceed the inverse size of the displacement field which will be reflected by a cutoff in g_{q-k} (see below). The scattering probability depends on the occupations of the initial and final states. Finally, since no real transitions between system states are allowed, the scattering must be elastic, as expressed by the energy-conserving Dirac delta.

In order to see if this effect may be of importance under realistic conditions, let us now study the specific case of a single electron in a pair of semiconductor quantum dots, as in the recent coherent manipulation experiment [21]. The decay of coherence between the localized states corresponds in this case to a localization process, hence T_2 may be referred to as the localization time. Since we are interested in the dephasing due to anharmonicity, we disregard phonon-assisted tunneling between the states ($\sigma_{x, y}$ coupling) which might appear only if the states overlap

(such terms obviously lead to exponential decoherence; the anharmonicity effects for such real transition processes were studied elsewhere [22,23]). The Hamiltonian is therefore

$$H_{\text{QD}} = \sum_{i=0,1} |i\rangle\langle i| [\epsilon_i + \sum_{\mathbf{k}} f_{\mathbf{k}}^{(i)*} (\tilde{b}_{\mathbf{k}} + \tilde{b}_{-\mathbf{k}}^\dagger)] + \sum_{\mathbf{k}} \hbar\omega_{\mathbf{k}} \tilde{b}_{\mathbf{k}}^\dagger \tilde{b}_{\mathbf{k}},$$

where $|0\rangle, |1\rangle$ are the basis states (each localized in one of the two dots), ϵ_i are the energies of the two states, $\tilde{b}_{\mathbf{k}}$ are phonon operators (with respect to the unperturbed equilibrium), and $f_{\mathbf{k}}^{(i)}$ are coupling constants. This Hamiltonian is transformed to the form of Eq. (1) by the canonical transformation (shift) of the phonon modes, $\tilde{b}_{\mathbf{k}} = b_{\mathbf{k}} + (f_{\mathbf{k}}^{(0)} + f_{\mathbf{k}}^{(1)})/(2\hbar\omega_{\mathbf{k}})$. The effective system-reservoir coupling constants are then $F_{\mathbf{k}} = f_{\mathbf{k}}^{(0)} - f_{\mathbf{k}}^{(1)}$. This shift modifies also the anharmonic Hamiltonian H_1 but, since the transformation is independent of system state, no extra coupling will appear. The new linear and quadratic terms in H_1 may be removed by rediagonalizing the phonon Hamiltonian, which produces negligible higher order corrections to the couplings.

In polar semiconductors, the strongest lattice displacement (polaron [7]) is related to longitudinal optical (LO) phonons, which are also subject to strong anharmonic coupling to acoustic phonons [24,25]. Therefore, the present discussion will be restricted to the scattering of longitudinal acoustic (LA) phonons on the LO displacement field. We assume dispersionless LO modes with frequency $\Omega = 54 \text{ ps}^{-1}$ (all values for GaAs [26]). The electron wave functions will be modeled by isotropic Gaussians of size L and the QDs will be displaced by a distance D along z [27]. The physical coupling constants are [23]

$$f_{\mathbf{k}}^{(0,1)} = \frac{e}{\hbar k} \sqrt{\frac{\hbar\Omega}{2V\epsilon_0\tilde{\epsilon}}} e^{-(Lk/2)^2} e^{\pm i(k_z D/2)}. \quad (6)$$

Hence,

$$|g_{\mathbf{k}}|^2 = \frac{|f_{\mathbf{k}}^{(0)} - f_{\mathbf{k}}^{(1)}|^2}{(\hbar\Omega)^2} = \frac{2e^2}{k^2 \hbar^3 V \epsilon_0 \tilde{\epsilon} \Omega} e^{-(Lk)^2/2} \sin^2 \frac{k_z D}{2}, \quad (7)$$

where e is the electron charge, V the normalization volume of the phonon modes, ϵ_0 the vacuum dielectric constant, and $\tilde{\epsilon} = 70$ the lattice part of the relative dielectric constant. The Gaussian momentum cutoff reflects the momentum conservation and momentum-position uncertainty for an electron wave packet of size L .

In an anharmonic process, an LO phonon interacts with two LA phonons with linear dispersion $\omega_{\mathbf{k}} = ck$ up to the Debye wave vector $k_{\text{D}} = 11 \text{ nm}^{-1}$, where $c = 5150 \text{ m/s}$ is the speed of sound. For this process, the general form of the coupling is $w_{\mathbf{q}-\mathbf{k},\mathbf{q},\mathbf{k}} = (w_0/\sqrt{V})\sqrt{qk}$ [28], where \mathbf{q}, \mathbf{k} pertain to the LA phonons and we neglected the dependence on the LO phonon momentum in the narrow range of

its relevant values $k \lesssim 1/L \ll k_{\text{D}}$. Using the measured lifetime $\tau_0 = 9.2 \text{ ps}$ of the LO phonon at $\mathbf{k} = 0$ [24] one finds, using the Fermi golden rule with the anharmonic Hamiltonian (2), $w_0^2 = \frac{64\pi\hbar^2 c^5}{\tau_0 \Omega^4}$.

Substituting the above result along with Eq. (7) into Eq. (5) one finds after some algebra [20]

$$\frac{1}{T_2} = \frac{64}{\pi^2} \frac{e^2 (k_{\text{B}} T)^5}{\tau_0 \hbar^6 \Omega^5 \epsilon_0 \tilde{\epsilon} c} \int_0^\infty \frac{dx}{x} e^{-x^2} \left(1 - \frac{\sin \alpha x}{\alpha x}\right) \times \left[\phi(x_{\text{D}}) - \phi\left(\frac{x x_{\text{D}}}{\sqrt{2} k_{\text{D}} L}\right) \right], \quad (8)$$

where $x_{\text{D}} = (\hbar c k_{\text{D}})/(k_{\text{B}} T)$, $\alpha = \sqrt{2} D/L$, and

$$\phi(x) = \int_0^x du u^5 \frac{e^u}{(e^u - 1)^2}.$$

The dependence of the dephasing time T_2 on temperature and distance between the basis states is shown in Fig. 1. Note that at moderate temperatures T_2 is much longer than the initial dephasing and reservoir memory times ($\sim 1 \text{ ps}$) so that the results are consistent. For low temperatures the localization rate is limited by the number of occupied initial states and the resulting allowed final states (due to energy conservation). As a result, one finds $T_2 \sim T^{-7}$ and the effect is extremely weak for sub-Kelvin temperatures [see Fig. 1(a)]. However, already at $T \sim 20 \text{ K}$ the dephasing rate is of the order of nanoseconds and becomes comparable to typical coherent manipulation times on such structures. At $\sim 100 \text{ K}$ the dephasing time drops to several picoseconds even for closely spaced dots. This temperature dependence is much stronger than in the quantum Brownian motion [3,29]. This is not astonishing, since this feature depends on the reservoir density of states and the physical nature of the coupling so that no universality can be expected here. In fact, similar strong temperature dependence has been found for localization processes due to scattering of light on dielectric balls and on free electrons [14].

At all temperatures the localization rate is increased by over an order of magnitude when the distance D between the dots grows from nanometers to micrometers. This distance dependence is shown in detail in Fig. 1(b). It turns

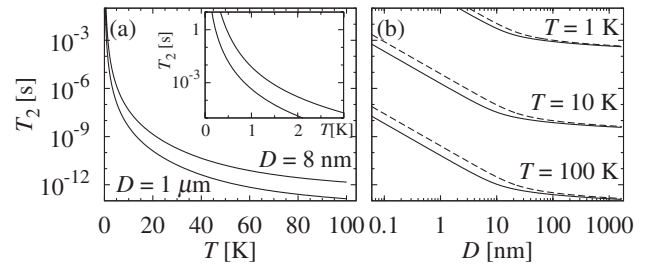


FIG. 1. Left: the dependence of the localization time on temperature for $L = 4 \text{ nm}$ (inset shows the low-temperature sector). Right: the dependence on the distance between the states for $L = 4 \text{ nm}$ (solid lines) and $L = 8 \text{ nm}$ (dashed).

out that the dephasing rate grows rather fast ($\sim D^2$) as long as the wave functions overlap. This dependence is a general feature in the regime of “ineffective single scattering event” [14]. When the states get separated, the increase of the localization rate continues, although it becomes only logarithmic. This is related to the crossover to the regime of spatially distinct states where a single scattering event is sufficient to extract the position information. The dependence on the dot size L becomes less and less important as the distance grows, as shown by the comparison between the dot sizes of 4 and 8 nm. This should be contrasted with the harmonic model [30], where the dependence on the separation saturates while that on the size does not. Therefore, the present results cannot be fully explained by merely invoking the known effect of increased carrier-phonon coupling for separated carriers. The logarithmic asymptotic behavior is unexpected on the grounds of a general discussion [15,16]; here it results from the long range nature of carrier-phonon interaction, manifested by the long wavelength singularity in Eq. (6).

The presented results show that anharmonicity of a super-Ohmic bosonic reservoir leads to a qualitative change in the dynamics of an open system. Exponential (Markovian) pure dephasing appears even though only partial decoherence was present without anharmonicity. For typical coupling properties, the dephasing rate depends very strongly on temperature. In the case of a superposition between two spatially separated states the rate of the resulting localization grows also with the spatial distance between the two states.

It should be stressed that the dephasing (localization) mechanism described here is inherent to physical properties of the system and appears universally for any localized states embedded in a translationally invariant bosonic reservoir with a certain degree of anharmonicity. As such, it sets material-dependent limits to system coherence, independent on any design improvements that might eliminate noise sources that dominate dephasing in the current experiments [21].

The present result is of importance to a few areas. First, it describes an additional dephasing mechanism that must be taken into account both in design of devices relying on quantum coherence and in interpretation of experiments. Apart from the localization effect discussed here, the anharmonic scattering mechanism will contribute, e.g., to the broadening of the zero-phonon line in QD spectroscopy [31,32]. Second, the dependence of dephasing on the distance in space may affect scalability of quantum computing schemes and applicability of concatenation techniques used in quantum fault-tolerant architectures [33]. Third, appearance of dephasing in models with nonsingular, super-Ohmic coupling may be of importance for emerging of classicality from quantum evolution [3].

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