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. On the other hand, the ability to control the states of quantum systems may open the way to novel applications, like quantum information processing. A simple model leading to this kind of behavior is composed of a two-level system and a bosonic bath interacting with phonons. 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. 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 decoherence. 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 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. It is shown, for a specific model of carrier-phonon interaction in semiconductor QDs, that the rate of localization grows with the anharmonicity of the reservoir, where also parameters of the model may be inferred from experiments.

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.

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.
We define the unitary operator

\[ \mathbb{W} = |0\rangle\langle 0| \otimes W^\dagger + |1\rangle\langle 1| \otimes W, \]

where \( W = \exp[(1/2) \sum_k g_k^* b_k - \text{H.c.}]. \) In terms of the new operators \( \beta_k = \mathbb{W} a_k \mathbb{W}^\dagger = b_k + \frac{1}{2} \sigma_z g_k, \) 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 non-diagonal element of the reduced density matrix (in the original basis), \( |\rho_{01}(t)| = |\rho_{01}(0)| \exp[-2i \omega \text{coth}(\hbar \omega / k_B T) J(\omega) \sin^2(\omega t/2)/(\hbar \omega)^2], \)

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 \( (J(\omega) = 0 \text{ around } \omega = 0), |\rho_{01}| \) reaches a finite asymptotic value (corresponding to partial dephasing) \[ 4. \]

On the other hand, if \( J(\omega) \sim \omega \) (Ohmic reservoirs), \( \rho_{01} \) 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 \( \beta \) the anharmonic Hamiltonian \( H_1 \) becomes

\[ H_1 = \frac{1}{6} \sum_{k,k_1,k_2,k_3} w_{k_1,k_2,k_3} \delta_{k_1+k_2+k_3=0} (A_{k_1} A_{k_2} A_{k_3} - \sigma_z g_k A_{k_2} A_{k_3} + 3g_k g_k g_k A_{k_3} - \sigma_z g_k g_k g_k A_{k_3}), \]

where \( A_k = \beta_k + \beta^\dagger_k. \) 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

\[ H_1^{(s)} = -\sigma_z \sum_{q,k} w_{q-k,-q,k} g_{q-k} \]

\[ \times \left[ \beta_k^\dagger \beta_k - \delta_{q,k} n_k + \delta_{q,k} (n_k + 1/2) \right]. \]

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 non-diagonal in phonon modes which allows for real phonon scattering processes (unlike the model of Ref. \[ 12. \]) A similar scattering Hamiltonian may be obtained by including higher exciton levels into a purely harmonic model \[ 12. \]

It is known that scattering on a heavy Brownian particle leads to decoherence and localization of the quantum state of the latter \[ 14, 15, 16. \] 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 \[ 41. \] 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 \[ 20. \]

\[ \frac{1}{T_2} = 2 \int_{-\infty}^{\infty} dt \langle B(t) B(0) \rangle \]

\[ = 4\pi \sum_{k,q} |w_{q-k,-q,k}|^2 |g_{q-k}|^2 n_k (n_k + 1) \delta(\omega_q - \omega_k), \]

where \( B(t) = \sum_{k,q} w_{q-k,-q,k} g_{q-k} [\beta_k^\dagger \beta_k e^{i(\omega_q - \omega_k)t} - \delta_{q,k} N_k] \) 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 \( (\sigma_x, \sigma_y) \) 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

\[ H_{QD} = \sum_{i=0,1} |i\rangle\langle i| \sum_k \epsilon_i + \sum_k f^{(i)}_k (\tilde{b}_k + \tilde{b}^\dagger_k) \]

\[ + \sum_k \hbar \omega_k \tilde{b}^\dagger_k \tilde{b}^\dagger_k, \]
where $|0\rangle, |1\rangle$ are the basis states (each localized in one of the two dots), $\epsilon_i$ are the energies of the two states, $b_k$ are phonon operators (with respect to the unperturbed equilibrium), and $f^{(i)}_k$ are coupling constants. This Hamiltonian is transformed to the form of Eq. 11 by the canonical transformation (shift) of the phonon modes, $\tilde{b}_k = b_k + (f^{(0)}_k + f^{(1)}_k)/(2\hbar \omega_k)$. The effective system-reservoir coupling constants are then $F_k = f^{(0)}_k - f^{(1)}_k$. 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 re-diagonozing the phonon Hamiltonian, which produces negligible higher order corrections to the couplings.

In polar semiconductors, the strongest lattice displacement (polaron) is related to longitudinal optical (LO) phonons, which are subject to strong anharmonic coupling to acoustic phonons. 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 $\Omega = 54$ ps$^{-1}$ (all values for GaAs). The electron wave functions will be modelled by isotropic Gaussians of size $L$ and the QDs will be displaced by a distance $D$ along $z$.

The physical coupling constants are 

$$f^{(0,1)}_k = \frac{e}{\hbar k} \sqrt{\frac{2\hbar \Omega}{V \varepsilon_0 \varepsilon}} e^{-\frac{(k \xi)^2}{2}} e^{i \frac{k z D}{2}}.$$  \hspace{1cm} (6)

Hence

$$|g_k|^2 = \frac{|f^{(0)}_k - f^{(1)}_k|^2}{(\hbar k)^2} \frac{2e^2}{k^2 \hbar^2 V \varepsilon_0 \varepsilon \Omega} e^{-\frac{(k \xi L)^2}{2}} \sin^2 \frac{k z D}{2},$$  \hspace{1cm} (7)

where $e$ is the electron charge, $V$ the normalization volume of the phonon modes, $\varepsilon_0$ the vacuum dielectric constant, and $\xi = 70$ is 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 $\omega_k = c k$ up to the Debye wave vector $k_0 = 11$ nm$^{-1}$, where $c = 5150$ m/s is the speed of sound. For this process, the general form of the coupling is

$$w_{q-k,q-k} = (w_0/\sqrt{V}) \sqrt{q k} \xi,$$

where $q, k$ pertain to the LA phonons and we neglected the dependence on the LO phonon momentum in the narrow range of its relevant values $k \lesssim 1/L \ll k_0$. Using the measured lifetime $\tau_0 = 9.2$ ps of the LO phonon at $k = 0$, one finds, using the Fermi golden rule with the anharmonic Hamiltonian, $w_0^2 = \frac{13 \pi \hbar^2 e^2}{101 \Omega}$. Substituting the above result along with Eq. 11 into

$$\frac{1}{T_2} = \frac{64 \pi^2 e^2 (k_B T)^5}{\tau_0 \hbar^2 \Omega^2 \varepsilon_0 \varepsilon c} \int_0^\infty dx \frac{x^2}{x} e^{-x^2} \left( 1 - \frac{\sin \alpha x}{\alpha x} \right) \frac{\sin \alpha x}{\alpha x} \int_0^\infty dx \left[ \phi(x) - \frac{(x D)}{\sqrt{2k_0 D}} \right],$$

where $x_D = (\hbar c k_0 T) / (k_B T)$, $\alpha = \sqrt{2D}/L$, and

$$\phi(x) = \int_0^x du u^5 \frac{e^u}{(e^u - 1)^2}.$$  \hspace{1cm} (8)

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 ($\sim 1$ 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. 1b). However, already at $T \sim 20$ K the dephasing rate drops to several picoseconds even for closely spaced dots. This temperature dependence is much stronger than in the quantum Brownian motion. 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.

At all temperatures the localization rate is increased by 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. 1b. It turns 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”. 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 \cite{20}, 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 \cite{15, 16}; here it results from the long range nature of carrier-phonon interaction, manifested by the long wavelength singularity in Eq. \ref{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 \cite{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 \cite{20, 31}. 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 \cite{32}. Third, appearance of dephasing in models with non-singular, super-Ohmic coupling may be of importance for emerging of classicality from quantum evolution \cite{3}. The author is grateful to M. Axt, T. Kuhn, L. Jacak, and A. Jamtka for discussions and to the Alexander von Humboldt Foundation and Polish MNI (PBZ-MIN-008/P03/2003, PB 2 P03B 08525) for support.

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Derivation of the $T_2$ time

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This auxiliary material explains how the long time scale behavior of the system was extracted in the Markov limit and gives the technical details of derivation of the final formula [Eq. (8)] for the dephasing times in the QD localization example.

The system evolution in the Markovian limit is found following the usual approach [? ]. The Hamiltonian $H^{(s)}_1$ (discarding the last term) is written in the interaction picture in the form

$$H^{(s)}_1(t) = -\sigma_z \otimes B(t),$$

where $B(t)$ is the reservoir operator defined below Eq. (5). We assume that the initial state of the system is separable in terms of the shifted operators $\beta_k$,

$$\rho(0) = \rho_S(0) \otimes \rho_B,$$

where $\rho_B$ is the thermal equilibrium state of the reservoir. Physically, this corresponds to preparing the system in such a way that the polaronic displacement field is formed by adiabatically following the system evolution. This assumption corresponds to typical timescales in the system of two QDs used as an example in the paper but, in any case, the long-time behavior cannot depend on the way the initial state was prepared.

The evolution equation for the density matrix of the total system is written in the usual integral form and the reduced density matrix of the two-level system is then obtained by tracing out the reservoir degrees of freedom,

$$\dot{\rho}_S(t) = -\int_0^t d\tau T_{RB} \left[ H^{(s)}_1, \left[ H^{(s)}_1, \rho(\tau) \right] \right],$$

where we used the fact that the operator $B(t)$ has a vanishing average at thermal equilibrium so that the 1st order term vanishes.

At this point, a series of standard approximations is introduced, leading to the Born-Markov equation for the evolution of the two-level system. First, one assumes that the perturbation of the reservoir state due to the joint evolution of the two subsystems is weak. It may then be assumed that at each instant of time the system state is separable, with the reservoir at thermal equilibrium, $\rho(s) = \rho_S(s) \otimes \rho_B$, which leads to the Nakajima-Zwanzig equation

$$\dot{\rho}_S(t) = -\int_0^t d\tau T_{RB} \left[ H^{(s)}_1, \left[ H^{(s)}_1, \rho_S(\tau) \otimes \rho_B \right] \right] = \int_0^t d\tau \text{Re} \langle B(t-\tau)B(t) \rangle (\sigma_z \rho_S(\tau) \sigma_z - \rho_S(\tau)), $$

where we use the fact that, for a hermitian operator $B(t)$, one has $\langle B(t)B(t') \rangle = \langle B(t')B(t) \rangle^*$. Next, one assumes that the evolution of the two-level system is very slow and its state changes negligibly during the reservoir memory time $\tau_M$ over which the correlation function $\langle B(\tau)B \rangle$ is essentially nonzero. This allows us to replace $\rho_S(\tau)$ by $\rho_S(t)$. Finally, at times $t \gg \tau_M$ the lower integration limit may be set to $-\infty$. Using the fact that $\rho_B$ is a stationary state, the Born-Markov evolution equation for the reduced density matrix of the two-level system may be written as

$$\dot{\rho}_S(t) = \frac{1}{2} \gamma [\sigma_z \rho_S(t) \sigma_z - \rho_S(t)],$$

where

$$\gamma = 4 \text{Re} \int_0^{\infty} \langle B(t)B \rangle dt = 2 \int_{-\infty}^{\infty} \langle B(t)B \rangle dt.$$

It is easy to see that the diagonal elements of $\rho_S(t)$ remain constant, while the non-diagonal ones decay with the time constant $T_2 = 1/\gamma$. The final formula of Eq. (5) follows in a straightforward manner by explicitly calculating the correlation function using the definition of the operator $B(t)$, as given below Eq. (5) in the paper.

This result may be directly used to calculate the $T_2$ time constant for the localization process used as an example in the paper. Inserting the coupling constants $g_{q-k}$ and $w_{q-k,-q,k}$ into Eq. (5) and changing both summations into integrals according to the usual formula (for normalization volume $V$), $\sum_k \to V \int d^3k/(2\pi)^3$, one gets

$$\gamma = \frac{1}{T_2} = \frac{8\varepsilon^2 e^5}{\pi^4 T_0 V \hbar \varepsilon_0} \int d^3k \int d^3q \frac{kq}{(k-q)^2} \frac{e^{-k^2/2q^2}}{\sin^2 \frac{kz-qzD}{2} \delta(ck-cq)nk(nq+1)}.$$
Let us introduce the new variable \( p = k - q \). Next, we express \( q \) in spherical coordinates \((q, \theta, \phi)\) in the reference system with the \( z' \) axis along \( p \) and \( p \) itself in spherical coordinates \((p, \vartheta, \varphi)\) in the original (absolute) reference frame.

In the new variables the Dirac delta is transformed into
\[
\delta(ck - cq) = \frac{1}{c}\delta(q - |q + p|) = \frac{\delta(p + 2q \sin \theta)}{c|\sin \theta|}.
\]

The result becomes
\[
\gamma = \frac{32e^2c^5}{\pi^2\tau_0\Omega^5\hbar\varepsilon_0\varepsilon} \int_{q<k_D} dq q^2 \int_{-\pi/2}^{\pi/2} d\theta \sin \theta \int_{|q + p|<k_D} dp p^2 \int_{-\pi/2}^{\pi/2} d\vartheta \cos \vartheta
\times \frac{q(p + q)}{p^2} e^{-\frac{1}{4}(Lp)^2} \sin^2 \frac{pD \sin \vartheta \delta(p + 2q \sin \theta)}{2c|\sin \theta|} n_{q + p}(n_q + 1),
\]

where the integration with respect to \( \phi, \varphi \) has been performed. It is now possible to integrate over \( p \) and \( \vartheta \). Introducing the new variables
\[
x = \frac{hqc}{kB_T}, \quad t = \frac{\sqrt{2kB_TLx}}{\hbar c \sin \theta}
\]
we can write the result in the form
\[
\gamma = \frac{64e^2c^4}{\pi^2\tau_0\Omega^5\hbar\varepsilon_0\varepsilon} \left( \frac{k_BT}{\hbar c} \right)^5 \int_0^{x_D} dx x^5 \frac{e^x}{(e^x - 1)^2} \int_0^{\sqrt{k_BT}Lx/x_D} dt e^{-t^2} \left( 1 - \frac{\sin \alpha t}{\alpha t} \right),
\]
where \( x_D = \hbar c k_D / (kB_T) \) and \( \alpha = \sqrt{2D/L} \). This formula may be integrated by parts to yield
\[
\gamma = \frac{64e^2c^4}{\pi^2\tau_0\Omega^5\hbar\varepsilon_0\varepsilon} \left( \frac{k_BT}{\hbar c} \right)^5 \left[ \phi(x_D) \int_0^{\sqrt{k_BT}L} dx x e^{-x^2} \left( 1 - \frac{\sin \alpha x}{\alpha x} \right) - \int_0^{\sqrt{k_BT}L} dx x e^{-x^2} \left( 1 - \frac{\sin \alpha x}{\alpha x} \right) \phi \left( \frac{xD}{\sqrt{2k_BT}} \right) \right].
\]

Since \( k_D L \gg 1 \) the upper limits of both integrals may be extended to \( \infty \), which leads to the form of Eq. (8).