Heat pumping with optically driven excitons

Erik M. Gauger[1,*] and Joachim Wabnig[1,2]
[1]*Department of Materials, University of Oxford, Oxford OX1 3PH, United Kingdom*
[2]*Cavendish Laboratory, University of Cambridge, Cambridge CB3 0HE, United Kingdom*
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We present a theoretical study showing that an optically driven excitonic two-level system in a solid-state environment can act as a heat pump by means of repeated phonon emission or absorption events. We derive a master equation for the combined phonon bath and two-level system dynamics and analyze the direction and rate of energy transfer as a function of the externally accessible driving parameters in the coherent control regime. We discover that if the driving laser is detuned from the exciton transition, cooling the phonon environment becomes possible.

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In semiconductor quantum dots (QDs), the ground state and the state containing a trapped electron-hole pair (exciton) form a two-level system (2LS) which is a popular implementation of a qubit. Unlike their atomic counterparts, such excitonic qubits are inextricably coupled to the lattice dynamics of the surrounding material.[1–5] Longitudinal-optical phonons give rise to excitonic polarons[3] and are known to play an important role for ultrafast excitation.[6,7] However, in the much slower coherent control regime that is so interesting for quantum information processing, optical phonons only contribute negligibly to the dephasing, and the coupling to longitudinal-acoustic (LA) phonons via the deformation potential is predicted to be dominant at low temperatures.[7,8] In recent experimental studies, the influence of LA phonons on the coherent QD dynamics has been measured by the tunnel charge through the QD (proportional to the excitonic population) following optical excitation,[9] and also through the direct coupling of the QDs dipole to optical modes in microcavities.[10,11] These studies confirm that a 2LS model with a perturbative treatment of the LA phonon interaction provides an excellent description in this regime.

Previous theoretical studies of the exciton-phonon interaction in the Rabi regime have fully discarded all information about the state of the phonon bath.[6–8,12–14] Here we develop a technique for tracking the number of excitations in the environment, allowing us to analyze the net rate of absorbed or released bath energy, and we show that a continuously driven excitonic qubit constitutes a controllable two-way heat pump (see Fig. 1). Exploiting this cooling effect would help with gaining further experimental insight into the exciton-phonon coupling, crucial for managing decoherence of semiconductor charge qubits, as well as providing an easy preparatory qubit initialization step for quantum information processing.

I. MODEL

We consider a self-assembled QD (such as InGaAs encased in GaAs substrate) illuminated by a laser beam with frequency ω_l, which is nearly resonant with the crystal ground state to exciton transition. In a frame rotating with the laser frequency and within the rotating wave approximation (RWA), the system is governed by the Hamiltonian ($\hbar = 1$),

$$H_S = \Delta/2\sigma_z + \Omega/2\sigma_x, \tag{1}$$

where the detuning Δ describes the energy difference between the basis states in the rotating frame, Ω is the Rabi frequency, coupling the two basis states, and where $\sigma_x = |g\rangle\langle e| + |e\rangle\langle g|$ and $\sigma_z = |e\rangle\langle e| - |g\rangle\langle g|$ are the Pauli pseudospin operators defined with respect to the QD ground state $|g\rangle$ and single-exciton state $|e\rangle$.

Let us further assume that the excitonic qubit is coupled to a bath of phonons, resulting in the total Hamiltonian,

$$H = H_S + H_B + H_I, \tag{2}$$

where $H_B = \sum_{\mathbf{q}} \omega_{\mathbf{q}} \hat{a}^\dagger_{\mathbf{q}} \hat{a}_{\mathbf{q}}$ is the free Hamiltonian of the phonon modes with $\hat{a}^\dagger_{\mathbf{q}}$ and $\hat{a}_{\mathbf{q}}$ being the creation and annihilation operators of a phonon in mode $\mathbf{q}$ with frequency $\omega_{\mathbf{q}}$. The exciton-phonon interaction term is generically given by[1,6]

$$H_I = \sigma_z \sum_{\mathbf{q}} g_{\mathbf{q}} (\hat{a}^\dagger_{\mathbf{q}} + \hat{a}_{\mathbf{q}}), \tag{3}$$

where the $g_{\mathbf{q}}$ are coupling constants. Moving into the interaction picture with respect to H_S and H_B, we obtain for the transformed interaction Hamiltonian,

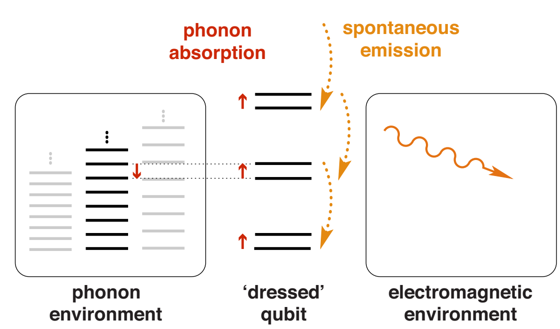

FIG. 1. (Color online) The driven exciton acts as a heat pump between the phonon and the electromagnetic environment. If the driving is detuned from the exciton transition, the laser-dressed eigenstates (see Ref. 14) are composed of different amounts of the ground and excited state. Cooling is possible when spontaneous emission repeatedly takes the system into its lower eigenstate, from where it can only absorb phonons.

$$\tilde{H}_I(t) = \tilde{\sigma}_z(t)\sum_{\mathbf{q}} g_{\mathbf{q}}(\hat{a}_{\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}}t} + \hat{a}_{\mathbf{q}} e^{-i\omega_{\mathbf{q}}t}), \quad (4)$$

where $\tilde{\sigma}_z(t)$ denotes the transformed σ_z operator.

For monitoring phonon excitations in the QDs solid-state environment, we adapt a technique from the literature on observing single-electron charge transport[15–17] by performing a gauge transformation which adds phase markers to the interaction Hamiltonian. Crucially, these do not alter the system's dynamics. We define the phonon-number specific density matrix, i.e., an object that contains information about both the qubit dynamics and the number of phonons in the environment,

$$\rho_m = \mathrm{tr}_E(P_m\varrho), \quad (5)$$

where tr_E denotes the trace over the phonon modes, ϱ is the density matrix of the combined phonon-qubit system, and P_m is a projection operator,

$$P_m = \frac{1}{2\pi}\int_0^{2\pi} d\lambda e^{-im\lambda}\mathcal{E}_\lambda, \quad \mathcal{E}_\lambda = e^{i\lambda\hat{N}}$$

with the total phonon number operator $\hat{N}=\Sigma_{\mathbf{q}}\hat{a}_{\mathbf{q}}^\dagger\hat{a}_{\mathbf{q}}$, and the gauge transformation operator $\mathcal{E}_\lambda$. The projection operator P_m projects out all parts of the wave function with a phonon number different from m (for a detailed discussion, see Ref. 16). The probability of having emitted m phonons into the environment is then obtained by the matrix trace of the phonon-number specific density matrix,

$$p_m = \mathrm{tr}(\rho_m). \quad (6)$$

It is convenient to introduce the Fourier-transformed density matrix as

$$\rho_\lambda = \sum_m e^{im\lambda}\rho_m = \mathrm{tr}(\varrho_\lambda)$$

with the definition $\varrho_\lambda=\mathcal{E}_{\lambda/2}\varrho\mathcal{E}_{-\lambda/2}^\dagger$. This particular choice corresponds to an initial state with a definite phonon number as in Ref. 15. It can be shown that the full density matrix ϱ_λ in the interaction picture obeys the von Neumann equation,

$$\dot{\varrho}_\lambda(t) = -i[\tilde{H}_\lambda(t)\varrho_\lambda(t) - \varrho_\lambda(t)\tilde{H}_{-\lambda}(t)] = \mathcal{L}_\lambda(t)\varrho_\lambda(t),$$

defining the superoperator $\mathcal{L}_\lambda(t)$, where the interaction Hamiltonian $\tilde{H}_\lambda=\mathcal{E}_{\lambda/2}\tilde{H}_I\mathcal{E}_{-\lambda/2}^\dagger$ has acquired phase markers on the phonon creation and annihilation operators,

$$\tilde{H}_\lambda = \tilde{\sigma}_z(t)\sum_{\mathbf{q}} g_{\mathbf{q}}(e^{-i(\lambda/2)}\hat{a}_{\mathbf{q}}^\dagger e^{i\omega_{\mathbf{q}}t} + e^{i(\lambda/2)}\hat{a}_{\mathbf{q}}e^{-i\omega_{\mathbf{q}}t}), \quad (7)$$

written in a shorter notation as $\tilde{H}_\lambda=\tilde{\sigma}_z(t)B_\lambda(t)$. The phase factor $\exp(-i\lambda/2)$ then keeps track of the creation of phonons while $\exp(i\lambda/2)$ tracks annihilation processes.

We proceed along the standard path of deriving a master equation (ME),[18] and obtain an integrodifferential equation for the reduced density matrix of the system ρ_λ,

$$\dot{\rho}_\lambda(t) = \mathrm{tr}_E\int_0^t ds\mathcal{L}_\lambda(t)\mathcal{L}_\lambda(s)\varrho_\lambda.$$

After the Born-Markov approximation,[18] the resulting Markovian ME reads

$$\dot{\rho}_\lambda(t) = -\int_0^\infty ds\sum_{i,j} G_{ij}^\lambda(s)\mathcal{S}_i(t)\mathcal{S}_j(t-s)\rho_\lambda(t),$$

where we have defined the superoperators $\mathcal{S}_1(t)X=\tilde{\sigma}_z(t)X$, $\mathcal{S}_2(t)X=X\tilde{\sigma}_z(t)$ and the environment correlation functions $G_{11}^\lambda(s)=\langle B_\lambda(s)B_\lambda(0)\rangle$, $G_{12}^\lambda(s)=-\langle B_{-\lambda}(0)B_\lambda(s)\rangle$, $G_{21}^\lambda(s)=-\langle B_{-\lambda}(s)B_\lambda(0)\rangle$, and $G_{22}^\lambda(s)=\langle B_{-\lambda}(0)B_{-\lambda}(s)\rangle$. To simplify the further algebraic evaluation, we express the operator $\tilde{\sigma}_z(t)$ in terms of system eigenoperators, yielding

$$\tilde{\sigma}_z(t) = \sum_{\omega\in\{0,\Lambda\}}(e^{-i\omega t}P_\omega + e^{i\omega t}P_\omega^\dagger), \quad (8)$$

where $\Lambda=\sqrt{\Omega^2+\Delta^2}$ now denotes the spacing between the system eigenstates $|-\rangle=\cos\theta|g\rangle-\sin\theta|e\rangle$ and $|+\rangle=\cos\theta|e\rangle+\sin\theta|g\rangle$ of Hamiltonian (1) with $2\theta=\arctan\Omega/\Delta$. In this basis, $P_0=\cos 2\theta(|-\rangle\langle-|-|+\rangle\langle+|)/2$ and $P_\Lambda=\sin 2\theta|-\rangle\langle+|$.

By introducing the Hermitian operator $\tilde{\mathcal{P}}=2P_0+e^{-i\Lambda t}P_\Lambda+e^{i\Lambda t}P_\Lambda^\dagger$ and abbreviating $\tilde{\mathcal{Q}}=e^{-i\Lambda t}P_\Lambda$, we obtain the following interaction picture ME after some straightforward algebra:

$$\dot{\rho}_\lambda = \Gamma_\downarrow[e^{-i\lambda}(\tilde{\mathcal{Q}}\rho_\lambda\tilde{\mathcal{P}}^\dagger + P\rho_\lambda\tilde{\mathcal{Q}}^\dagger) - \tilde{\mathcal{P}}^\dagger\tilde{\mathcal{Q}}\rho_\lambda - \rho_\lambda\tilde{\mathcal{Q}}^\dagger\tilde{\mathcal{P}}] + \Gamma_\uparrow[e^{i\lambda}(\tilde{\mathcal{P}}^\dagger\rho_\lambda\tilde{\mathcal{Q}} + \tilde{\mathcal{Q}}^\dagger\rho_\lambda\tilde{\mathcal{P}}) - \tilde{\mathcal{P}}\tilde{\mathcal{Q}}^\dagger\rho_\lambda - \rho_\lambda\tilde{\mathcal{Q}}\tilde{\mathcal{P}}^\dagger],$$

where the rates are given by $\Gamma_\downarrow=J(\Lambda)[n(\Lambda)+1]/2$ and $\Gamma_\uparrow=J(\Lambda)n(\Lambda)/2$ when the phonon bath ρ_E is in a thermal state.[19] Further,

$$J(\omega) = 2\pi\sum_{\mathbf{q}}|g_{\mathbf{q}}|^2\delta(\omega-\omega_{\mathbf{q}}) \quad (9)$$

is the spectral density of phonon modes and $n(\omega)=[\exp(\beta\omega)-1]^{-1}$ denotes the thermal occupancy of a phonon mode with frequency ω.

Upon going back to the Schrödinger picture, the operators $\tilde{\mathcal{P}}$ and $\tilde{\mathcal{Q}}$ lose their time-dependent phase factors, turning into $\mathcal{P}=\sigma_z$ and $\mathcal{Q}=P_\Lambda$, respectively. We can return to the number representation with the transformation,

$$\rho_m = \frac{1}{2\pi}\int_0^{2\pi} d\lambda e^{-im\lambda}\rho_\lambda.$$

This yields a set of coupled differential equations for the evolution of phonon-specific density matrices. After a RWA [implemented by setting $\mathcal{P}=\mathcal{L}$, expected to be valid whenever $\Lambda \gg J(\Lambda)$ (Refs. 18 and 20)], we finally obtain a ME in diagonal Lindblad form

$$\dot{\rho}_m = -i[H_S(t),\rho_m] + \Gamma_\downarrow(2P_\Lambda\rho_{m+1}P_\Lambda^\dagger - P_\Lambda^\dagger P_\Lambda\rho_m - \rho_m P_\Lambda^\dagger P_\Lambda) + \Gamma_\uparrow(2P_\Lambda^\dagger\rho_{m-1}P_\Lambda - P_\Lambda P_\Lambda^\dagger\rho_m - \rho_m P_\Lambda P_\Lambda^\dagger). \quad (10)$$

This equation describes the joint phonon-qubit dynamics in the case where the phonon environment is only weakly disturbed from thermal equilibrium by the excitonic qubit.

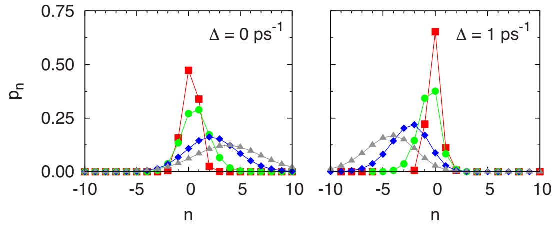

FIG. 2. (Color online) The distribution of p_n at different points of time in the presence of radiative decay. The constant excitation pulse uses $\Omega=1\ \text{ps}^{-1}(\approx 2/3\ \text{meV})$ and a detuning Δ as indicated in the plots (in ps^{-1}). The decay rate is fixed at $\Gamma=0.1\ \text{ps}^{-1}$. The different colors/symbols correspond to time as follows: red squares=1.2, green circles=10, blue diamonds=40, gray triangles =70, all in units of full Rabi cycles, $2\pi/\sqrt{\Omega^2+\Delta^2}$.

II. PHONON-ASSISTED TRANSITIONS

We now apply Eq. (10) to an excitonic qubit that is optically driven by a pulse of constant intensity Ω. The initial state at $t=0$ is the system ground state with zero excitations in the environment, $\rho_n(0)=\delta_{n,0}|g\rangle\langle g|$. For simplicity, we define the spectral density phenomenologically, $J(\omega)=\alpha\omega^3\exp(-\omega^2/\omega_c^2)$, where α describes the effective electron-phonon coupling strength and ω_c is the high-frequency phonon cutoff. For relatively weak driving with a peak Rabi frequency Λ well below both the electron and the hole cutoff, we can neglect the exponential cutoff term altogether. Setting $\alpha=1/4\ \text{ps}^2$ yields a coupling strength that is consistent with the magnitude of the GaAs deformation potential reported in the literature.[6,21]

The structure of Eq. (10) permits the emission or absorption of no more than a single phonon: The system is initialized in the state $|g\rangle\langle g|=(|-\rangle+|+\rangle)(\langle -|+\langle +|)/2$, i.e., in an equal superposition of system eigenstates. The Lindblad operator P_Λ induces a transition from $|+\rangle$ to $|-\rangle$ while $P_\Lambda^\dagger$ raises population from $|-\rangle$ to $|+\rangle$. Once a decay process has happened, we find the system in the $|-\rangle$ state, meaning it cannot decay again.

Provided the excitation is sufficiently long, the population ratios thus tend to a Boltzmann distribution, as is obvious from the phonon emission rate proportional to $n(\Lambda)+1$ and the absorption rate proportional to $n(\Lambda)$,

$$\lim_{t\to\infty}\frac{\text{tr}[\rho_0(t)|+\rangle\langle +|]}{\text{tr}[\rho_1(t)]}=\lim_{t\to\infty}\frac{\text{tr}[\rho_{-1}(t)]}{\text{tr}[\rho_0(t)|-\rangle\langle -|]}=e^{-\beta\Lambda}.$$

So far, the only perturbation to the system has been caused by the coupling to the phonon bath, resulting in single phonon emission and absorption. Realistically, other dissipative processes will be present in any physical system. We shall include these using additional phenomenological noise operators, such as pure dephasing and radiative decay of the exciton. We model these processes with an additional Lindblad dissipator[18] on the right-hand side of Eq. (10),

$$\mathcal{D}\rho=\Gamma\left[L\rho L^\dagger-\frac{1}{2}(L^\dagger L\rho+\rho L^\dagger L)\right] \tag{11}$$

with respective Lindblad operators $L=\sigma_z$ and $L=\sigma_-$, and where Γ is the dephasing or decay rate. These operators do not preserve the system's eigenstates; consequently, under their action, phonon-assisted transitions become possible in any of the ρ_n subspaces. This leads to a dynamic equilibrium, where phonon-assisted transitions keep occurring after the transient (coherent) evolution of $\rho=\Sigma_n\rho_n$ has subsided. Our theoretical model is well suited for illustrating this behavior: Fig. 2 presents the p_n distribution at different points of time for the case of optical decay.

The pure dephasing σ_z operator randomizes the phase between $|g\rangle$ and $|e\rangle$, thus balancing the population of $|-\rangle$ and $|+\rangle$. Consequently, once the steady state has been reached, phonon emission always occurs with a faster rate then absorption, and the distribution is therefore shifted in the direction of increasing n, meaning the average number of emitted phonons increases. On the other hand, the distribution can move in either direction for optical decay from $|e\rangle$ to $|g\rangle$. For $\Delta=0$, $|g\rangle$ consists of an equal superposition of $|-\rangle$ and $|+\rangle$ while it contains a larger $|-\rangle$ component for $|\Delta|>0$. Under these latter circumstances, it is possible for phonon absorption to permanently dominate over emission, shifting the distribution in the direction of decreasing n, as shown in the right panel of Fig. 2. In this case, thermal energy is removed from the QDs bulk surroundings and released into the wider environment by spontaneous photon emission (depicted in Fig. 1).

III. HEAT TRANSFER RATE

To quantify this observation, we consider only the radiative decay operator of Eq. (11) in the following. We proceed by calculating the rate of phonon emission or absorption, which is given by[17]

$$\langle \dot{n}(t)\rangle=\frac{d}{dt}\sum_m mp_m=i\frac{d}{d\lambda}\text{tr}(\dot{\rho}_\lambda)\bigg|_{\lambda=0}$$
$$=2\,\text{tr}[\Gamma_\downarrow\mathcal{Q}\rho(t)\mathcal{Q}^\dagger-\Gamma_\uparrow\mathcal{Q}^\dagger\rho(t)\mathcal{Q}], \tag{12}$$

where $\rho(t)$ is obtained by integrating Eq. (10) inclusive of the

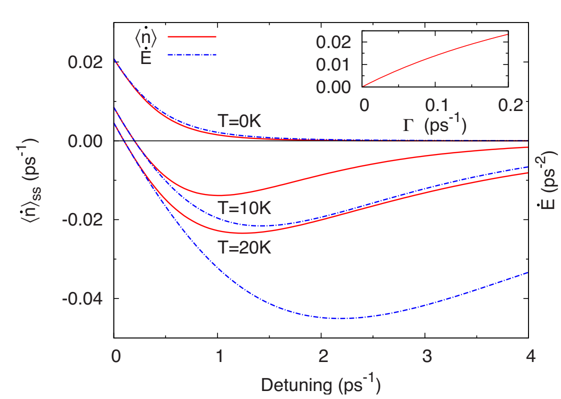

FIG. 3. (Color online) The rate of phonon-induced transitions $\langle\dot{n}\rangle_{ss}$ (solid red) and the rate of energy transfer $\dot{E}$ (dashed blue), both as a function of the detuning for $\Omega=1\ \text{ps}^{-1}$. The rates shown here are the steady-state values of Eq. (12) for the decay rate $\Gamma=0.1\ \text{ps}^{-1}$. A positive sign indicates net phonon emission whereas a negative corresponds to net phonon absorption. The inset shows $\langle\dot{n}\rangle_{ss}$ for fixed $\Delta=1\ \text{ps}^{-1}$ as a function of Γ at $T=10$ K.

Lindblad dissipator [Eq. (11)] with $L=\sigma_-$ and disregarding the indices m of ρ_m.

Figure 3 presents the steady-state value $\langle \dot{n} \rangle_{ss}$ of Eq. (12) as a function of Δ. As expected, net phonon absorption is only possible at finite temperature for off-resonant excitation. It is a natural question to ask at which rate energy is transferred to or from the surroundings of the system. Considering the quantity $\dot{E}=\sqrt{\Omega^2+\Delta^2}\langle \dot{n} \rangle_{ss}$ shows that the number of absorbed phonons decreases for a larger detuning, yet their energy is greater, shifting the Δ that achieves optimal heat transfer. The inset of Fig. 3 shows that the process is limited by the radiative decay time Γ of the system. A saturation only occurs when the spontaneous emission rate becomes faster than that of phonon-mediated transitions but this regime would require an unrealistic optical lifetime on the order of a picosecond or less.

We proceed by estimating the achievable cooling rate for realistic parameters in SI units. Using $\Omega_0=\Delta=1$ ps^{-1} corresponds to a phonon energy of $\Lambda=1.49\times 10^{-22}$ J. With approximately 0.02 absorbed phonons per picosecond (at $T=20$ K), we obtain a theoretical energy transfer rate of roughly 3×10^{-12} J/s. Neglecting heating effects, this achieves a temperature reduction on the order of 1 K/s for a micrometer cube of GaAs.[22] Of course, any real sample will also be subject to heating processes, e.g., by the laser illumination and by thermal contact to its surroundings. While it is difficult to estimate the precise rate of heating, it could plausibly exceed the cooling rate in bulk samples. However, by incorporating the QD on a nanopillar or a cantilever, the thermal coupling to the bulk could be reduced to make net cooling possible.[23] An experiment to show the cooling effect could proceed in the following way: insert a cooling cycle before the standard sequence for observing Rabi oscillations and study the influence of the length of the cooling cycle on coherence times. If cooling is successful one would expect increased coherence times.

IV. SUMMARY

We have shown that a single excitonic QD, an experimentally well-studied system, can act as a heat pump. Adapting a counting statistic approach from the context of charge transfer, we have discussed how energy can be removed from the QDs environment by means of repeated phonon absorption in conjunction with spontaneous photon emission. This effect does not rely on the structure of the spectral density, making our analysis applicable to similar systems with a different coupling mechanism to a bath of harmonic oscillators. This opens up the possibility of experimental investigation of quantum heat pumps as well as the prospect of environment preparation for the use of excitonic systems in quantum information processing.

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*erik.gauger@materials.ox.ac.uk

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[22] Using a mass density of $\mu=5.3\times 10^3$ kg/m^3 and a specific heat of 350 J/(kg K) gives a heat capacity of 1.85×10^{-12} J/K for a micrometer cube of GaAs.

[23] Provided the Born-Markov approximation is still justified, this only requires a substitution of $J(\omega)$ to account for the modified phonon spectrum.