

# Possible spontaneous energy flow against a temperature step

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## Abstract

Modification of the Van Hove and Davies mathematically rigorous weak-coupling scaling for open quantum systems leads to a slow-particle (excitation) description where transfer can be well competed by bath-assisted relaxation. It yields spontaneous exciton diffusion, i.e. also energy flow between two macroscopic systems, going possibly even against temperature step.

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Except for early challenging universal validity of the Second law of thermodynamics by Maxwell construction of his demon [1,2], the first modern and serious analysis of this law is due to Feynman [3]. His idea was a proper transformation of a noise existing and influencing the systems in question owing to the existence of external baths into uni-directional motion, usable work etc. The mechanism is based on the idea of the ratchet and pawl but as a detailed analysis by Feynman [3] himself shows, no real violation of the Second law is in practice possible [4]. Under different names, the problem remains, however, topical - see, e.g., [5] and citations therein. Since 1997, models appear within the rigorous quantum theory of open systems that contradict standard thermodynamics [6–8]. Even before, serious challenge to the Second law appeared on the level of experiment [9], with conclusions now fully supported by rigorous quantum theory [10]. In 1998, the first theoretical model working cyclically and producing usable work at the cost of just heat from the bath without compensation was reported [11–13]. Since 1999, possibility of violation of the Second law of the macroscopic thermodynamics in the strict quantum regime is independently being reported by also other groups, in both theory and experiment [14–16]. In 2000, public discussion started about challenges to the Second law [17–19]. Sheehan [20] recently published experimental data confirming that the Duncan’s paradoxical system [18] potentially able to violate the second law could in principle be realized. Also other principles of the standard thermodynamics have already been challenged [21]. Likely necessary conditions for such an unexpected behaviour are so far, however, quantum regime (or at least some specific quantum effect included) and possibly very specific effects of strong or intermediate couplings. The point is that on the classical level, the above mechanisms cease to work and the validity of the Second law seems to be justified [22]. Similarly, in the zeroth order of the system-bath coupling (see any standard textbook of statistical thermodynamics), no violations of the second law appear. All this makes detailed investigation of relation of the rigorous quantum theory of open systems to thermodynamic principles highly desirable.

Here, we report on similar results obtained for rather a trivial model of two electronic two-level systems, with their levels described as exciton-less and single-exciton states. Each of the electronic systems has its own bath of phonons (with, in the limiting sense, infinite number of

modes) forming, together with them, corresponding subsystems 1 or 2 modeling two macroscopic bodies. Interaction of the excitons with their own baths leads to finite exciton life-time effects. Contact between these (electronic+phonon) subsystems is provided solely by the coherent exciton transfer term. The Hamiltonian  $H$  of all the (electron+phonon) complex consists of those of the (electronic+phonon) subsystems  $H_j$ ,  $j = 1, 2$  complemented by the contact term, i.e.

$$H = H_1 + H_2 + J(a_1^\dagger a_2 + a_2^\dagger a_1),$$

$$H_j = \epsilon_j a_j^\dagger a_j + \sum_{\kappa} \hbar \omega_{\kappa} b_{j\kappa}^\dagger b_{j\kappa} + \frac{1}{\sqrt{N}} \sum_{\kappa} g_{j\kappa} \hbar \omega_{\kappa} (a_j + a_j^\dagger)(b_{j\kappa} + b_{j\kappa}^\dagger), \quad j = 1, 2. \quad (1)$$

The exciton creation (annihilation) operators  $a_j^\dagger$  ( $a_j$ ),  $j = 1, 2$  are assumed to fulfil the Pauli relations  $\{a_1, a_1^\dagger\} = \{a_2, a_2^\dagger\} = 1$ ,  $[a_1, a_2^\dagger] = 0$ ,  $\{a_1, a_1\} = \{a_2, a_2\} = [a_1, a_2] = 0$  etc. Phonon frequencies  $\omega_{\kappa}$  are, for simplicity, assumed the same for both the baths. Finally,  $\epsilon_1 \neq \epsilon_2$  are the exciton energies while  $N$  is the number of the phonon modes (finite before taking the baths thermodynamic limit) in each bath separately.

We fully accept the mathematic theory by Davies [23–25] but refrain here from using the Van Hove - Davies identification of the perturbation with the system coupling to the bath. (Notice that though we speak about perturbation, the Davies theory is exact.) This identification implies scaling

$$g \rightarrow 0, \quad \tau \rightarrow +\infty, \quad g^2 \tau = \text{const.} \quad (2)$$

forming mathematical basis of the weak coupling theories. Here  $g$  is a small parameter of coupling of the electronic (exciton) system to the phonon bath ( $g_{1\kappa}, g_{2\kappa} \propto g$ ) and  $\tau$  is a new time unit in terms of which the rescaled (‘macroscopic’) time is introduced. Scaling (2) yields always relaxation where the bath-assisted processes including dephasing are by the definition assumed to be infinitely slower than in-phasing processes connected with the hopping (transfer) term  $\propto J$  in (1). This is thus the regime where natural finite exciton life-time broadening of energy levels  $\epsilon_j$ ,  $j = 1, 2$  in, e.g., the Golden-Rule transfer rate formulae becomes irrelevant. Let us remind that with (2), the exciton relaxation goes always to a canonical state whose exciton density matrix is diagonal in the

basis of eigenstates of the electronic Hamiltonian  $H_S = \sum_{j=1}^2 \epsilon_j a_j^\dagger a_j + J(a_1^\dagger a_2 + a_2^\dagger a_1)$ . This would mean existence of covalent bonding between excited states  $a_j^\dagger |vac\rangle$ ,  $j = 1, 2$  hindering exciton  $1 \leftrightarrow 2$  flows.

Instead, in order to have a possibility to include the line broadening whose importance will be seen below, we must have the in-phasing and dephasing commensurable. That is why we include into the Davies perturbation, in addition to the system-bath coupling, also the coherent transfer term  $\propto J$ . In other words, our scaling reads

$$g \rightarrow 0, J \rightarrow 0, \tau \rightarrow +\infty, g^2\tau = \text{const}, \frac{g^2}{J} = \text{const}. \quad (3)$$

The ratio  $g^2/|J|$  could be here arbitrary but finite and nonzero. Application of (3) causes, as a consequence of thus obtained commensurability of in-phasing and dephasing, violation of conditions for applicability of, e.g., standard equilibrium statistical mechanics as the dephasing processes (which are owing to the system interaction with the bath) are thus *not* any more assumed vanishingly weak as compared to all (specifically, in our case, transfer) processes inside the exciton system. Hence, the stationary density matrix of the exciton system can no longer be expected canonical, i.e.  $\propto \exp(-\beta H_S)$ . This could make the very notion of equilibrium dubious.

Readers who believe in standard Golden Rule formula with the  $\delta$ -function expressing the energy-conservation law broadened by finite life-time effects can skip the following reasoning, going directly to formula (9). All others may find details of the scaling procedure (3) in, e.g., [26] where the treatment (except for another identification of perturbation) technically follows the algebra of [24] but includes also the procedure of getting rid of the Poincaré recurrences [27] (what leads to appearance of irreversibility). This is achieved by including the thermodynamic limit of the baths. The result which is free of any approximation can be explained, without going into technical details, in words as follows: After projecting off the baths (phonons above), a closed equation for the density matrix  $\rho(t)$  of only the electron system is obtained which has a form of the Liouville equation

$$\frac{d}{dt}\rho(t) = \frac{i}{\hbar}[H_S, \rho(t)] + \mathcal{R}\rho(t). \quad (4)$$

It has been assumed here that the density matrix of the complex ‘system+bath’ is initially factorizable  $\rho_{S+B}(t=0) = \rho(t=0) \otimes \rho_B$  (otherwise, (4) would have to be complemented by an inhomogeneous term). With, e.g., the canonical form  $\rho_B \propto \exp(-\sum_{j=1}^2 \beta_j \sum_{\kappa} \hbar \omega_{\kappa} b_{j\kappa}^{\dagger} b_{j\kappa})$ , initial bath temperatures  $T_j = 1/(\beta_j k_B)$  ( $j = 1, 2$ ) can be well defined. For small enough  $|J|$ , the exciton systems first thermalize with their baths, i.e.  $T_j$  fast acquire the meaning of temperatures of the whole  $j$ -th (exciton+bath) subsystem. As for the (linear) relaxation superoperator  $\mathcal{R}$ , it describes the bath- (i.e. phonon-) assisted relaxation to eigenstates of  $H_S|_{J=0}$ . This point is quite important because with scaling (2), one would obtain the weak-coupling form of (4) where the relaxation would be to (the canonical density matrix which is diagonal in) eigenstates of  $H_S|_{J \neq 0}$  [25,28]. This difference can be easily understood because with (2), both  $\mathcal{R} \propto g^2$  (relaxation is a bath-assisted process) while in our case, also  $J \propto g^2$ . Hence, since with (3) only second order terms survive in (4), one must set  $J = 0$  inside  $\mathcal{R}$  (though not in the first term on the right hand side of (4)).

We now specify the basis we work in. In order to make the treatment as simple as possible, we ignore double excited states with both electron levels excited. These terms may be always easily re-introduced. So, the (localized) basis in the exciton Hilbert space consists of states  $|0\rangle$  (excitonless state),  $|1\rangle = a_1^{\dagger}|0\rangle$  and  $|2\rangle = a_2^{\dagger}|0\rangle$ . Omitting some uncoupled and for us unimportant site off-diagonal elements of  $\rho(t)$ , we get from (4) [29]

$$\frac{d}{dt} \begin{pmatrix} \rho_{00}(t) \\ \rho_{11}(t) \\ \rho_{22}(t) \\ \rho_{12}(t) \\ \rho_{21}(t) \end{pmatrix} = \begin{pmatrix} \mathcal{A} & \mathcal{B} \\ \mathcal{C} & \mathcal{D} \end{pmatrix} \cdot \begin{pmatrix} \rho_{00}(t) \\ \rho_{11}(t) \\ \rho_{22}(t) \\ \rho_{12}(t) \\ \rho_{21}(t) \end{pmatrix}. \quad (5)$$

Here the blocks

$$\mathcal{A} = \begin{pmatrix} -\gamma_{1\uparrow} - \gamma_{2\uparrow} & \gamma_{1\downarrow} & \gamma_{2\downarrow} \\ \gamma_{1\uparrow} & -\gamma_{1\downarrow} & 0 \\ \gamma_{2\uparrow} & 0 & -\gamma_{2\downarrow} \end{pmatrix}, \quad \mathcal{B} = \begin{pmatrix} 0 & 0 \\ iJ/\hbar & -iJ/\hbar \\ -iJ/\hbar & iJ/\hbar \end{pmatrix},$$

$$\mathcal{C} = \begin{pmatrix} 0 & iJ/\hbar & -iJ/\hbar \\ 0 & -iJ/\hbar & iJ/\hbar \end{pmatrix}, \quad (6)$$

and  $2 \times 2$  block  $\mathcal{D}$  is diagonal with diagonal matrix elements  $-\frac{1}{2}(\gamma_{1\downarrow} + \gamma_{2\downarrow}) + i(\epsilon_2 - \epsilon_1)/\hbar$  and  $-\frac{1}{2}(\gamma_{1\downarrow} + \gamma_{2\downarrow}) - i(\epsilon_2 - \epsilon_1)/\hbar$ . Above,

$$\gamma_{j\uparrow} = \frac{2\pi}{\hbar} \frac{1}{N} \sum_{\kappa} (g_{j\kappa} \hbar \omega_{\kappa})^2 \frac{1}{\exp(\beta_j \hbar \omega_{\kappa}) - 1} \delta(\hbar \omega_{\kappa} - \epsilon_j) = e^{-\beta_j \epsilon_j} \gamma_{j\downarrow}, \quad j = 1, 2 \quad (7)$$

are the usual Golden-Rule bath-assisted transfer rates.

First of all, we now need a proper expression for the  $1 \rightarrow 2$  exciton flow  $I$ . One can start from elementary quantum mechanical expression for the particle flow or from the exciton total balance calculating  $\frac{d}{dt} \langle a_j^\dagger a_j \rangle$ . In the latter case, the result is nothing but the second or third equation of (5). Trivial interpretation of individual terms therein immediately gives

$$I = \frac{2J}{\hbar} \Im \rho_{12}. \quad (8)$$

Stationary solution for  $\rho_{12}$  obtained from (5) then yields in (8), after keeping just leading (lowest order in  $g$ ) terms,

$$\begin{aligned} I &\approx \frac{2\pi}{\hbar} J^2 \cdot \frac{1}{\pi} \frac{\frac{\hbar}{2}(\gamma_{1\downarrow} + \gamma_{2\downarrow})}{[\frac{\hbar}{2}(\gamma_{1\downarrow} + \gamma_{2\downarrow})]^2 + [\epsilon_2 - \epsilon_1]^2} [\rho_{11} - \rho_{22}] \\ &= \frac{2\pi}{\hbar} J^2 [\rho_{11} - \rho_{22}] \cdot \int_{-\infty}^{+\infty} \frac{1}{\pi} \frac{\frac{\hbar}{2} \gamma_{1\downarrow}}{[\frac{\hbar}{2} \gamma_{1\downarrow}]^2 + [\epsilon - \epsilon_1]^2} \cdot \frac{1}{\pi} \frac{\frac{\hbar}{2} \gamma_{2\downarrow}}{[\frac{\hbar}{2} \gamma_{2\downarrow}]^2 + [\epsilon - \epsilon_2]^2} d\epsilon. \end{aligned} \quad (9)$$

Several facts are worth mentioning:

- The second equality in (9) corresponds to the assumed elastic form of the exciton transfer term  $\propto J$  in (1): The exciton transfer proceeds horizontally in energy, at arbitrary energy  $\epsilon$  in the overlap region of tails of broadened exciton levels  $\epsilon_1$  and  $\epsilon_2$ .
- Stationary forms of the exciton site occupation probabilities  $\rho_{jj} = \exp(-\beta_j \epsilon_j) / (1 + \exp(-\beta_j \epsilon_1) + \exp(-\beta_j \epsilon_2))$  in (9) as they result from (5) differ from the respective statistically correct forms  $1 / (1 + \exp(\beta_j \epsilon_j))$ ,  $j = 1, 2$  (Fermi-Dirac distributions for  $J \rightarrow 0$ ) solely because of the above omission of the double-excited (two-exciton) state  $a_1^\dagger a_2^\dagger |0\rangle$ . Reintroducing it

turns  $\rho_{jj}$  into the respective Fermi-Dirac forms but leaves the form of (9) unchanged. So, (9) is nothing but the proper balance Golden Rule transfer rate with proper inclusion of the energy level broadening. This yields really a small space for speculations about correctness of the above approach.

- In connection with that, one should realize that formula (9) is, in case of a single bath at least, fully standard (e.g. formula (6.8.27) of [30]). What is above between our formulae (4) and (6) may be thus viewed as only its rigorous derivation for the case of our two baths.
- Owing to term  $\rho_{11} - \rho_{22}$  in (9), the exciton flow goes in the direction from the local level (site) with the higher stationary exciton population to that one with the lower one. For, e.g.,  $\epsilon_1 < \epsilon_2$  and  $\beta_1 = \beta_2$ , the flow is positive in the direction  $1 \rightarrow 2$ .
- The same picture survives if we prescribe different initial temperatures  $T_1 \equiv 1/(\beta_1 k_B) \neq T_2 \equiv 1/(\beta_2 k_B)$  to our baths 1 and 2 forming parts of our subsystems 1 and 2. Specifically, as far as  $0 < \epsilon_1 < \epsilon_2$  and  $T_1 < T_2 < T_1 \epsilon_2 / \epsilon_1$ , the stationary population  $\rho_{11}$  of site 1 is still greater than  $\rho_{22}$  of site 2. Hence the flow remains positive, going thus *against* the temperature step from site 1 to site 2.

The exciton (as an electronic excitation) bears energy ( $\epsilon_j$  when  $g \rightarrow 0$ ) what implies also an energy flow from subsystem 1 to subsystem 2 though  $T_1 < T_2 < T_1 \epsilon_2 / \epsilon_1$ . This clearly contradicts the Clausius (1850) [31] formulation of the Second law stating that no spontaneous energy (heat) flow against temperature gradient is possible. We can see only one excuse that could save validity of the Clausius form of the second law in our case: It would be that the  $1 \rightarrow 2$  forward flow and the backflow  $2 \rightarrow 1$  would in average take place at different energies  $\epsilon$ . This possibility could easily be beaten off showing that the energy flow  $Q$  can be written, in a correspondence with (9), as

$$\begin{aligned}
 Q &= \frac{2\pi}{\hbar} J^2 [\rho_{11} - \rho_{22}] \cdot \int_{-\infty}^{+\infty} \frac{1}{\pi} \frac{\frac{\hbar}{2} \gamma_{1\downarrow}}{[\frac{\hbar}{2} \gamma_{1\downarrow}]^2 + [\epsilon - \epsilon_1]^2} \cdot \frac{1}{\pi} \frac{\frac{\hbar}{2} \gamma_{2\downarrow}}{[\frac{\hbar}{2} \gamma_{2\downarrow}]^2 + [\epsilon - \epsilon_2]^2} \epsilon d\epsilon \\
 &= \frac{2\pi}{\hbar} J^2 [\rho_{11} - \rho_{22}] \cdot \frac{\hbar}{2\pi} \frac{\epsilon_2 \gamma_{1\downarrow} + \epsilon_1 \gamma_{2\downarrow}}{[\frac{\hbar}{2} (\gamma_{1\downarrow} + \gamma_{2\downarrow})]^2 + [\epsilon_2 - \epsilon_1]^2}.
 \end{aligned} \tag{10}$$

Derivation of (10) is more complicated as we need, for the definition of  $Q$ , energy balance at sites 1 and 2. It is, however, not unique in such a case to say what is, e.g., the  $J$ -induced renormalization of already broadened exciton energies  $\epsilon_j$ . This problem, on the other hand, disappears once any (one or the second) of the two energy levels becomes practically unbroadened (negligible  $\gamma_{1\downarrow}$  or  $\gamma_{2\downarrow}$ ). In such a case, derivation of (10) by calculating either  $\frac{d}{dt}\langle H_1 \rangle$  or  $\frac{d}{dt}\langle H_2 \rangle$  is really possible and formula (10) is really obtained as a correct asymptotic formula for at least  $\gamma_{2\downarrow}\epsilon_1 \ll \gamma_{1\downarrow}\epsilon_2$  (or  $\gamma_{1\downarrow}\epsilon_2 \ll \gamma_{2\downarrow}\epsilon_1$ ) and (owing to preserving just the leading term in (9))  $|J|/\hbar \ll \text{Min}(\gamma_{1\downarrow}, \gamma_{2\downarrow})$ . The latter condition ensures that, according to (5), the electronic two-level systems very fast thermalize (irrespective of their initial states) with their respective baths (accepting their temperatures  $T_1$  and  $T_2$ ) before the much slower (but still commensurable)  $1 \leftrightarrow 2$  stationary exciton and energy transfer appears, and that the  $J$ -induced perturbation of the stationary exciton site occupation probabilities gets unimportant.

To comprehend the effect physically, one should realize basic points discussed above:

- The on-energy-shell (i.e. elastic) exciton transfer is possible between (among) even different exciton levels owing to finite widths of the exciton levels caused by finite life-time effects.
- Stationary exciton concentration will be lower at sites where the exciton excitation energy is higher even if a slightly higher, e.g., local temperature is ascribed to such sites.
- As standard diffusion processes going in the direction of decreasing concentration, such net exciton transfers, i.e. also energy flows, are therefore possible prevailingly in the direction *to* such sites with lower exciton concentration, going thus contingently even against moderate temperature steps.

The last point, when translated to the language of exciton-mediated energy flows, however, contradicts (in our case the Clausius formulation of) the Second law of the macroscopic thermodynamics [31]. This is in accordance with analogous statements made in previous independent, theoretical as well as experimental papers [6–8,11–16]. In contrast to these works, however, the present mechanism is physically easily understandable, the model is rather trivial, and its treatment is



based on just notoriously known very simple formulae that can be here again rigorously justified. Concluding: We insist that the above derivation is sufficiently rigorous to serve as a basis for a relevant discussion. However, even those who might feel internal inhibitions to admit that can hardly overlook the fact that conclusions stemming from standard mathematical formalism of quantum mechanics (specifically the Golden Rule formula) as applied in formulae (9-10) may in some specific situations become hardly reconcilable with standard thermodynamics.

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