Dimer as a challenge to the second law[*]

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Abstract. A simple system consisting of an asymmetric dimer cooperating with two baths is presented as a challenge to the second law of thermodynamics. This requires specific coupling of the dimer to the baths and a specific thermodynamic regime.

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1 Introduction

In view of experimental [1–3], theoretical [4–7] (see also [8,9] for extended list of references) as well as combined evidence [10], it is difficult to disregard or ignore *a priori* all challenges to the second law of thermodynamics as erroneous, not reproducible, or misleading. Furthermore, a number of other paradoxical systems exist where experimental evidence speaks in favour of the second law violations though decisive experiments have not been performed so far [11,12]. Growing interest in the problem that would deepen our understanding of Nature and potentially redefine all of thermodynamics underscores the importance of finding simple systems that do not mask the relevant processes by technical complexity. Many attempts have been made, but their results are still not conclusive. Single quantum Brownian particle in a harmonic confining potential and coupled linearly to a bath of noninteracting harmonic oscillators is perhaps still too simple for such purposes [13]. Here we report that a simple quantum particle on a pair of non-equal states – an asymmetric dimer – may under suitable conditions behave in a way challenging the second law. The necessary conditions include a specific type of strong coupling of the dimer to a pair of baths.

2 Model

The Hamiltonian for the proposed system reads

$$H = H_S + H_B + H_{S-B}, \tag{1}$$

[*] This articles has been submitted by V. Capek a few weeks before he passed away and it is presumably his last paper. It has been edited by D.P. Sheehan (University of San Diego, USA).

where Hamiltonians for a single particle on a dimer, the pair of baths, and the system-bath coupling read, respectively:

$$H_S = \sum_{j=1}^{2} \epsilon_j a_j^\dagger a_j + J(a_1^\dagger a_2 + a_2^\dagger a_1),$$

$$H_B = \sum_k [\hbar\omega_k b_k^\dagger b_k + \hbar\Omega_k B_k^\dagger B_k] \equiv H_B^I + H_B^{II},$$

$$H_{S-B} = \frac{1}{N} \sum_{k_1 \neq k_2} g_{k_1,k_2} \hbar\sqrt{\omega_{k_1}\omega_{k_2}} a_1^\dagger a_1$$

$$\times (b_{k_1} + b_{k_1}^\dagger)(b_{k_2} + b_{k_2}^\dagger)$$

$$+ \frac{1}{\sqrt{N}} \sum_k G_k \hbar\Omega_k (a_1^\dagger a_2 + a_2^\dagger a_1)(B_k + B_k^\dagger)$$

$$\equiv H_{S-B}^I + H_{S-B}^{II}. \tag{2}$$

Operators $a_j^\dagger$ and a_j are the standard (Fermi- or Bose-like) particle creation and annihilation operators; $\epsilon_1 < \epsilon_2$ are local particle site-energies and J is the hopping (resonance or transfer) integral. The two baths of harmonic phonons involved (baths I and II) are distinguished by corresponding phonon frequencies ω_k and Ω_k, and also by the respective creation $b_k^\dagger$, $B_k^\dagger$ and annihilation operators b_k, B_k. N is the number of the phonon modes in each of the baths separately.

We are interested in the density matrix $\rho(t) \equiv \rho_S(t)$ of our system, *i.e.* the dimer. Its definition reads as $\rho_S(t) = \mathrm{Tr}_B \rho_{S+B}(t)$ where $\rho_{S+B}(t)$ is the density matrix of the whole complex 'system + bath' (the bath itself consisting of two baths as specified above). Time dependence of $\rho_{S+B}(t)$ is determined, in addition to the initial condition, by the Liouville - von Neumann equation

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\rho_{S+B}(t) = \frac{1}{\hbar}[H, \rho_{S+B}(t)] \equiv \mathcal{L}\rho_{S+B}(t) \tag{3}$$

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\begin{pmatrix}\rho_{11}(t)\\\rho_{22}(t)\\\rho_{12}(t)\\\rho_{21}(t)\end{pmatrix}=\begin{pmatrix}-\mathrm{i}\Gamma_\uparrow & \mathrm{i}\Gamma_\downarrow & -\frac{J}{\hbar} & \frac{J}{\hbar}\\[4pt]\mathrm{i}\Gamma_\uparrow & -\mathrm{i}\Gamma_\downarrow & \frac{J}{\hbar} & -\frac{J}{\hbar}\\[4pt]-\frac{J}{\hbar} & \frac{J}{\hbar} & \frac{\epsilon_1-\epsilon_2}{\hbar}-\mathrm{i}\Gamma-\frac{\mathrm{i}}{2}(\Gamma_\uparrow+\Gamma_\downarrow) & \frac{\mathrm{i}}{2}(\Gamma_\uparrow+\Gamma_\downarrow)\\[4pt]\frac{J}{\hbar} & -\frac{J}{\hbar} & \frac{\mathrm{i}}{2}(\Gamma_\uparrow+\Gamma_\downarrow) & \frac{\epsilon_2-\epsilon_1}{\hbar}-\mathrm{i}\Gamma-\frac{\mathrm{i}}{2}(\Gamma_\uparrow+\Gamma_\downarrow)\end{pmatrix}\times\begin{pmatrix}\rho_{11}(t)\\\rho_{22}(t)\\\rho_{12}(t)\\\rho_{21}(t)\end{pmatrix}. \quad (14)$$

where $\mathcal{L}\ldots = [H,\ldots]/\hbar$ is the Liouville superoperator. The basic problem is that $\rho_{S+B}(t)$ contains too much information, which makes it difficult to follow its time-dependence in practice. Therefore, at this stage it is common to resort to some of the projection techniques; we shall do this now.

3 Davies theory and results

In what follows, we first apply an exact mathematical result obtained by Davies – see [14], Theorem 1.2. (Discussion of applicability of the Davies theory may be found below.) Davies' theorem states that, in the physical notation [15],

$$\lim_{\lambda\to 0}\sup_{0\le\lambda^2 t\le a}||\rho(t)-\mathrm{e}^{-\mathrm{i}(\mathcal{L}_0+\langle\mathcal{L}_1\rangle+i\lambda^2\mathcal{K})t}\rho(0)||=0,$$

$$\mathcal{K}\ldots=-\int_0^{+\infty}\mathrm{d}x\,\mathrm{Tr}_B\left(\mathrm{e}^{\mathrm{i}\mathcal{L}_0 x}\mathcal{L}_1\mathrm{e}^{-\mathrm{i}\mathcal{L}_0 x}(1-\mathcal{P})\mathcal{L}_1(\ldots\otimes\rho^B)\right),$$

$$\langle\mathcal{L}_1\rangle\ldots=\mathrm{Tr}_B(\mathcal{L}_1\ldots\otimes\rho^B). \qquad (4)$$

Several aspects of this deserve explanation.

– The Hamiltonian has been split

$$H = H_0 + \lambda H_1, \qquad (5)$$

with the corresponding definitions

$$\mathcal{L}_0\ldots=\frac{1}{\hbar}[H_0,\ldots], \quad \lambda\mathcal{L}_1\ldots=\frac{1}{\hbar}[\lambda H_1,\ldots] \qquad (6)$$

Here λ is a scaling parameter.

– Integral $\int_0^{+\infty}\mathrm{d}x\ldots$ in the definition of $\mathcal{K}$ in (4) is taken in the thermodynamic limit of the bath. This limit is not indicated explicitly, but it is important in that it ensures the convergence of the integral as well as time-irreversible behavior of the system.

– $\mathcal{P}\ldots=\mathcal{P}^2\ldots$ is a projection superoperator fulfilling

$$\mathcal{P}\mathcal{L}_0=\mathcal{L}_0\mathcal{P}. \qquad (7)$$

With the two choices of H_0 below, (7) is well satisfied with our choice of $\mathcal{P}\ldots$ as the Argyres-Kelley projector [16]

$$\mathcal{P}\ldots=\rho^B\,\mathrm{Tr}_B(\ldots), \qquad (8)$$

– Finally, the initial condition

$$\rho_{S+B}(0)=\rho(0)\otimes\rho^B \qquad (9)$$

has been assumed. This means that the system and bath are initially statistically independent, with ρ^B being the initial density matrix of the bath. In order to formally introduce temperature, we take ρ^B as

$$\rho^B=\frac{\exp\{-\beta_I H_B^I-\beta_{II}H_B^{II}\}}{\mathrm{Tr}_B\exp\{-\beta_I H_B^I-\beta_{II}H_B^{II}\}}. \qquad (10)$$

Hence $T_I=1/(k_B\beta_I)$ and $T_{II}=1/(k_B\beta_{II})$ are the initial temperatures of baths I and II with k_B the Boltzmann constant.

The meaning of the statement (4) becomes evident once we realize that $\rho(t)$ is the density matrix of the system with its time-dependence determined from (3). So, the statement (4) means that the time development of $\rho(t)$ is not, under the above assumptions and in the above limiting sense, distinguishable from that one given by equation

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\rho(t)=[\mathcal{L}_0+\langle\mathcal{L}_1\rangle+i\lambda^2\mathcal{K}]\rho(t). \qquad (11)$$

Now, a standard choice is usually made corresponding to the van Hove scaling given by

$$H_0=H_S+H_B, \quad \lambda H_1=H_{S-B}. \qquad (12)$$

This choice then yields (11) as the weak-coupling master equation [17] (in fact, these are Redfield equations without the secular Redfield approximation [18–20]). In the weak coupling regime the bath-induced processes are infinitely slower than inherent internal transfer processes in the system; in our case, these are the coherent transfer processes caused by the coherent transfer term $J(a_1^\dagger a_2+a_2^\dagger a_1)$ in H_S in (2). This regime, however, is not what we are here interested in; in fact, in the weak coupling regime nothing particular happens. Instead, we are interested in the situation when the coherent transfer process rates are comparable to the bath assisted process rates indicated by both H_{S-B}^I and H_{S-B}^{II}. Hence, as far as we are forced by technical reasons to use the scaling ideas, the only and proper choice of H_0 and λH_1 compatible with the above regime is that one in which

$$H_0=\sum_{j=1}^2\epsilon_j a_j^\dagger a_j+H_B, \quad \lambda H_1=J(a_1^\dagger a_2+a_2^\dagger a_1)+H_{S-B}. \qquad (13)$$

This yields from (4) the set of equations for site elements of the density matrix $\rho(t)$

see equation (14) above

Here

$$\Gamma = \frac{2\pi}{\hbar N^2} \sum_{k_1 \neq k_2} |g_{k_1 k_2}|^2 \hbar^2 \omega_{k_1} \omega_{k_2} n_B^I(\hbar\omega_{k_1})$$

$$\times [1 + n_B^I(\hbar\omega_{k_2})]\delta(\hbar\omega_{k_1} - \hbar\omega_{k_2}),$$

$$\Gamma_\uparrow = \frac{2\pi}{\hbar N} \sum_k |G_k|^2 (\hbar\Omega_k)^2 n_B^{II}(\hbar\Omega_k)\delta(\epsilon_2 - \epsilon_1 - \hbar\Omega_k),$$

$$\Gamma_\downarrow = \frac{2\pi}{\hbar N} \sum_k |G_k|^2 (\hbar\Omega_k)^2 [1 + n_B^{II}(\hbar\Omega_k)]\delta(\epsilon_2 - \epsilon_1 - \hbar\Omega_k),$$

$$n_B^I(z) = [\exp(\beta_I z) - 1]^{-1}, \quad n_B^{II}(z) = [\exp(\beta_{II} z) - 1]^{-1}. \tag{15}$$

In these expressions for Γ's, one can recognize the standard Golden Rule bath-assisted transfer rates. Their positions on the right hand side of (14) are standard and fully correspond to what we know about their role in transfer and bath-induced dephasing processes. The bad news to critics of this approach is: Except for the difference between $\Gamma_\uparrow$ and $\Gamma_\downarrow$ found in (15), owing to the spontaneous processes with respect to bath II, (14) fully corresponds to what we would get from standard models, for example, from the stochastic Liouville equation approach [21].

It is now easy to find the stationary solution to (14) and to use it to express the net rate

$$Q = (\epsilon_2 - \epsilon_1)[\Gamma_\downarrow \rho_{22} - \Gamma_\uparrow \rho_{11}] \tag{16}$$

of energy emitted into (or, for $Q < 0$, absorbed from) bath II as a consequence of the existence of coupling term H_{S-B}^{II} of the dimer to the bath. (Eq. (16) follows easily from the physical meaning of the quantities involved, but it can also be directly derived from the heat balance condition.) The result reads

$$Q =$$

$$\frac{2\Gamma(J/\hbar)^2(\epsilon_2 - \epsilon_1)[\Gamma_\downarrow - \Gamma_\uparrow]}{(\Gamma_\downarrow + \Gamma_\uparrow)[(\epsilon_2 - \epsilon_1)^2/\hbar^2 + \Gamma^2 + \Gamma(\Gamma_\downarrow + \Gamma_\uparrow)] + 4\Gamma(J/\hbar)^2}. \tag{17}$$

Thus, in general, we have $Q \neq 0$ expressing, for $\epsilon_2 > \epsilon_1$, the net heat flow from bath I to bath II. Several facts are worth stressing:

- The effect is purely quantum with respect to bath II. Disregarding the corresponding spontaneous processes, *i.e.* setting $\Gamma_\downarrow = \Gamma_\uparrow$, yields zero energy (heat) flow to bath II; that is $Q = 0$.
- The effect is based on a competition between the coherent and incoherent (bath-II assisted) channels. Respectively, these are given by terms $J(a_1^\dagger a_2 + a_2^\dagger a_1)$ in H_S and $\frac{1}{\sqrt{N}}\sum_k G_k \hbar\Omega_k(a_1^\dagger a_2 + a_2^\dagger a_1)(B_k + B_k^\dagger) \equiv H_{S-B}^{II}$ in (2). Setting either $J = 0$ or $G_k = 0$ (*i.e.* $\Gamma_\uparrow = \Gamma_\downarrow = 0$) yields zero heat flow into bath II. We point out that:
  - these two mechanisms (coherent and incoherent channels) appear not to have been considered previously as simultaneously competing or cooperating; and

- the coherent mechanism is elastic (*i.e.* symmetric with respect to the $1 \leftrightarrow 2$ transitions) while the incoherent bath-II assisted one is inelastic, *i.e.* asymmetric, preferring $2 \rightarrow 1$ transitions with net heat flow going *into* bath II (because of the assumed inequality $\epsilon_2 > \epsilon_1$).

The simultaneously competing coherent and incoherent transfer mechanisms $1 \leftrightarrow 2$ are crucial to this model in that each of the mechanisms alone would lead to establishing definite phase relations between probability amplitudes of finding the particle at sites 1 and 2. This would imply a kind of a chemical bonding that would make the particle distribution stiff, thus hindering particle and heat transfer.

- As expected, one obtains $Q = 0$ when $\Gamma = 0$, *i.e.* when there is no heating of the particle at site 1 (at the cost of heat contents of bath I). Furthermore, because of energy-time uncertainty relations, the moving particle *may* be on-site heated. Such an on-site particle heating combined with two competing mechanisms of the particle transfer to another site has, to our best knowledge, not considered been considered before.

Two points are noteworthy. First, for the final effect, the regime considered is critical (see Eq. (14)). In the weak-coupling regime, the 1-2 covalent bonding owing to the term $\propto J$ in H_S would again make the particle distribution stiff, hindering any heat flow transmitted by the particle. Technically, this would appear in the square matrix on the right hand side of (14). Secondly, in this system $Q > 0$, irrespective of temperatures $T_I > 0$ and $T_{II} > 0$ of the individual baths. So, for $T_{II} > T_I$, we have a spontaneous heat transfer from our colder bath I to the warmer bath II. This transfer is spontaneous; that is, it is not aided from outside. Both the baths are macroscopic and standard thermodynamics is applicable. However, this spontaneous heat flow from colder to warmer macroscopic bodies violates the Clausius form of the second law of thermodynamics. This behavior is allowed within the rigorous quantum theory of open systems by Davies.

4 Discussion

Although this model and its treatment are rigorous, several apparently open issues should be discussed.

- The upper time limit beyond which the Davies theory could fail is finite, as indicated by the finite constant a in (4). This does not present a problem, given that it can be extended to arbitrarily long times (or, equivalently, to arbitrarily low frequencies). Such extensions are standard devices in kinetic theory. For example, one can cite numerous successful transport predictions starting from the Boltzmann equation based on just such assumptions. Of course, this provides no explicit justification in this case.
- Recently, Novotný [22] proposed an exactly solvable model in which an uncritical application of

conclusions of the Davies theory [1] yields results contradicting the exact solution obtained by the nonequilibrium Green function method. Unlike (2), the Novotný model (Hamiltonian of the system + bath, together with their interaction) is bilinear in creation and annihilation operators. Such models (*e.g.*, those of harmonic phonons) are exactly diagonalizable, and therefore not applicable to the inherently nonequilibrium model discussed here. Diagonizable models for fermions, for example, include the exact eigenstates given by Slater determinants composed of properly chosen counterparts of molecular orbitals and those of the Bloch-like states from the periodic solid state theory. Particles in such states, in effect, do not scatter; therefore, there is no effective bath-induced diffusion in the model of [22] and, consequently, no long-time diffusion regime corresponding to (11). While the Novotny model is *mathematically* correct, in essence, it has built-in second law compliance.

One should stress that the effect and model of [8] criticized by Novotný is intimately connected with the effect of diffusion. In fact, Novotný in [22] introduced his formal bath by simply a formal procedure of splitting sites into two groups defining the bath and the system. This cannot change the coherent character of propagation of particles considered. So, the model of [22] cannot describe diffusion and, therefore, cannot be critically compared with [8]. Similarly, this is the case for our model above. Despite this, Novotny's model remains as a challenge to the Davies mathematical theory, and in connection with the problem treated here, it shows uncertainties that remain concerning applicability of any limiting theory to finite coupling situations, in particular when assumptions underlying the theory cannot be simply verified. Hence, independent verification of the above conclusions on the existence of the spontaneous heat flow from our colder to the warmer baths would be highly desirable. This could take the form of theory or experiment.

The most direct theoretical verification is perhaps provided by the Time-Convolutionless Generalized Master Equation (TCL-GME) theory [23–27]. Using the Argyres-Kelley projector (8), this approach yields, the basic equation in form of

$$i\frac{d}{dt}\rho(t) = [\mathcal{L}_0 + \langle\mathcal{L}_1\rangle + i\lambda^2\mathcal{K}^{\text{TCL}-\text{GME}}(t)]\rho(t). \quad (18)$$

to be compared with (11). Here, we have again applied the initial condition (9). The relaxation superoperator $\lambda^2\mathcal{K}^{\text{TCL}-\text{GME}}(t)$ is (owing to initial transient effects) necessarily time-dependent, however, with increasing time and with a genuine bath (not artificial one as in [22]), it fast acquires a time-independent form. A necessary condition is that the real time t exceeds a (presumably) well identifiable bath-dephasing time t_{deph}. Then (18) reproduces the result of (11). With (13),

with $t \gg t_{deph}$, and upon disregarding higher-order effects in λ, $\lambda^2\mathcal{K}^{\text{TCL}-\text{GME}}(t)$ reads, however,

$$\lambda^2\mathcal{K}^{\text{TCL}-\text{GME}} \ldots =$$
$$-\int_0^\infty \text{Tr}_B\Big(\mathcal{L}_1 \exp\{-i\mathcal{L}_0\tau\}(1-\mathcal{P})\mathcal{L}_1\mathcal{P}$$
$$\times \exp\{i\mathcal{L}_0\tau\}\,d\tau(\rho^B \otimes \ldots)\Big) + \mathcal{O}(\lambda^4). \quad (19)$$

Structurally, the form of the relaxation superoperator given in (4) and by (19) are evidently different. Hence, for general and more complicated models, two different predictions on the time-development of the system relaxation may be provided by (4) and (18–19). We have verified, on the other hand, that this is fortunately *not* the case of our model (2). In other words, to the lowest (second) order in λ and with $t \gg t_{deph}$, (18-19) reproduces (14), reproducing thus the above conclusion on violation of the second law of thermodynamics in the Clausius formulation.

One thing should be stressed: The TCL-GME method respects the fact that time t could be, in experiment, taken in principle arbitrarily large (frequency could be arbitrarily low). On the other hand, experimental coupling constants (*e.g.* λ in our case) are fixed. Thus, consequent theories should resort (if at all) to expansions in powers of the coupling constants at most at the last stage, after taking the long-time (low-frequency) limit. Our TCL-GME approach is definitely of this type. Hence, it is resistive to objections of the previous type regarding the finiteness of a. Nonetheless, uncertainties remain concerning finite values of the coupling constants; hence, it would be desirable to find exactly solvable limiting cases and models where:

- expansions in powers of λ could be avoided; and
- finite values of λ could be used throughout the time interval of the relaxation process, until a stationary (though still not necessarily equilibrium) state is achieved.

Unfortunately, such methods and models do not yet exist in full generality. One additional argument is worth mentioning here: The most relevant and crucial feature of (14) – irrespective of the method used to derive it – that is technically responsible for values of $Q \neq 0$ and the violability of the Second law is the relaxation among local eigenstates of $H_S|_{J=0}$ superimposed on a free propagation in terms of $H_S|_{J\neq0}$. From the point of view of the Davies theory, this can be understood as a consequence of choosing, in (13) and in accordance with the physical regime investigated, the perturbation as including both the bath-free and the bath-assisted transfer processes. So, an element of subjectivity in the choice could be made responsible for the second law challenge obtained. This manner of treating competing processes is, has been known for years as the only allowed choice corresponding to the above regime, and it has been since successfully used many times with verified effects [28]. Absence of the $J-$ induced (*i.e.* coherent) contribution to the bath-assisted relaxation processes in, *e.g.*, the relaxation superoperator $\lambda^2\mathcal{K}^{\text{TCL}-\text{GME}} \ldots$ [2]

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[1] In fact, Novotný did not verify the assumptions of the Davies theory.

[2] Notice absence of J in all the formulae (15).

could be well justified in the singular reservoir limit in which $t_{deph} \to 0$, all $|g_{k_1,k_2}| \to +\infty$, $|G_k| \to +\infty$, but such that all the transfer rates Γ's remain finite and comparable with $J/\hbar$. Then all the $J-$dependent contributions automatically disappear from the relaxation superoperator $\lambda^2 \mathcal{K}^{\mathrm{TCL-GME}} \ldots$ because of the infinitely short extent of all the integrations on the time axis.

In the end, one should realize that all methods mentioned here rely upon standard approaches to modelling in physics and chemistry in which smaller terms in the Hamiltonian, Liouvillean etc. (*e.g.*, presumably less important reaction channels) are omitted and only dominating ones are considered. This approach might, in long time limit, be treacherous [29]. If so, however, the consequences would be far reaching, going even beyond physics or natural science in general. In any event, the final word must come from experiment.

Note

This is the final article written by Vlada Capek before his death in October, 2002. Vlada was one of the principal exponents of the emerging field of second law challenges. Through examination of unresolved foundational issues and counter-examples to its absolute status, this new field attempts to understand the second law in more fundamental terms than has yet been achieved.

More memorable to me than Vlada's prodigious and original contributions to physics, however, were his sharp wit, quick laugh, intellectual courage, and indominable spirit.

He was a lion. He will be missed.

Daniel P. Sheehan,
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