Journal of Statistical Physics, Vol. 92, Nos. 5/6, 1998

Regularization of Quantum Relative Entropy in Finite Dimensions and Application to Entropy Production

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Received October 10, 1996; final May 29, 1998

The fundamental concept of relative entropy is extended to a functional that is regular-valued also on arbitrary pairs of nonfaithful states of open quantum systems. This regularized version preserves almost all important properties of ordinary relative entropy such as joint convexity and contractivity under completely positive quantum dynamical semigroup time evolution. On this basis a generalized formula for entropy production is proposed, the applicability of which is tested in models of irreversible processes. The dynamics of the latter is determined by either Markovian or non-Markovian master equations and involves all types of states.

KEY WORDS: Quantum relative entropy; irreversible processes; entropy production; open quantum systems.

I. INTRODUCTION

Relative entropy is a concept of central importance in various fields, such as mathematical probability theory.[1,2] and classical or quantum statistical mechanics.[3,4] In quantum theory relative entropy can be regarded as being of a more fundamental nature than the von Neumann entropy. This point of view has been adopted in the excellent texts by Ohya and Petz,[5] and by Thirring.[4] It is true indeed that the relative entropy functional must be used in the proofs of many important and partly surprising properties of the von Neumann entropy itself as, for example, strong subadditivity.[3,6]

In the theory of stochastic processes it is usually assumed that the amount of information which is contained in a given probability density decreases in the course of time. The relative entropy is a measure for the

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0022-4715/98/0900-1115$15.00/0

loss of information between the initial and the final states. A similar interpretation applies to irreversible quantum dynamical processes close to thermodynamic equilibrium where the second law of thermodynamics is obeyed. This is the case in the so-called weakly irreversible regime which is described by the quantum version of the phenomenological Onsager theory, and where a linear relationship between generalized forces and fluxes is valid.[7–9] The important quantity of entropy production is then given by a time-derivative of relative entropy if the dynamics is determined by a completely positive quantum dynamical semigroup.[8, 10, 11]

For more general irreversible processes, in particular those very far from equilibrium and of a strongly non-Markovian nature, thermodynamic considerations break down, but the von Neumann entropy still remains a useful concept because of its information theoretic interpretation for single states. Moreover, to classify pairs of states on the basis of their relative information content or, else, to measure the change of information during a dynamical evolution, relative entropy again is a useful quantity. It is positive for all pairs and can be used to establish a generalized formula for entropy production. On the other hand, some rather inconvenient properties must be analyzed in more detail.

First of all, relative entropy $Q(\rho, \sigma)$ is an ordered functional of two states ρ and σ that takes on different values for reversed order. This spoils its desired interpretation as representing something like a distance in state space.[5] Furthermore, in finite dimensions, i.e., for N-level systems, the maximum value of entropy is finite but relative entropy can become infinite, namely, whenever the second state is non-faithful (non-invertible). State space is a compact convex set, embedded in a real vector space, with the pure states as extremal points. A peculiar situation arises for two states ρ and σ lying very close together, where σ is assumed to be pure. Then $Q(\rho, \sigma)$ takes on the value $+\infty$, whereas $Q(\sigma, \rho)$ almost equals zero. This testifies to a dependence on directions in state space, and is of practical significance in relation to the second law of thermodynamics where during any state change from ρ to σ the degree of mixture and, consequently, the entropy[4] can only increase. On the other hand, for a general time evolution between these two states far from the weakly irreversible regime the above-mentioned asymmetry seems to be completely unrealistic since either of states ρ, σ can be chosen to be the initial or final state. Moreover, in case of quantum dynamical semigroup evolution one can even determine the two different infinitesimal generators for such twin processes. Pure final states are not at all unrealistic but occur, for instance, in various problems of spectroscopy whenever the emission of quanta from an excited level to a ground state is considered. Initial and final state have entropy zero, and relative entropy is infinite. Hence there is no obvious way to calculate entropy production from time-dependent relative entropy as in the weakly irreversible case. For this reason a functional which is different from but similar to relative entropy has been proposed earlier.[12] It has the drawback that some of the important properties of the latter must be abandoned. Nevertheless, reasonable analytical results for photon emission in a two-level system as well as for free induction decay have been obtained.

We believe that the theory should still be improved. A few more considerations must be taken into account and should support our point of view. One could argue that for processes outside the weakly irreversible regime there would not be any need of the relative entropy functional. From a purely mathematical point of view some suitable entropy-like Lyapunov functional might be sufficient to determine entropy production. However, several physical reasons will favour an extrapolated version of relative entropy even far from equilibrium. First of all, according to all experience general irreversible quantum dynamics with unique final state will always show two characteristic time intervals. A very instructive example is given by quantum collapse and revival dynamics in the damped Jaynes-Cummings model in quantum optics.[33] A first regime is characterized by a posssibly strong non-Markovian behavior followed necessarily by a second Markovian regime in the long-time tale. This is ultimately the reason why for many processes a weak-coupling (van Hove) limit is possible at all in that the first non-Markovian interval either shrinks to zero or becomes so small that its effect can be taken into account by so-called slip initial conditions.[26] It is in this Markovian tail that use of the relative entropy functional is a must, as has been shown in various papers.[10, 11, 32] It does not seem likely that a Lyapunov functional chosen for the first interval would fit to this regime. Second, and certainly most important, a Lyapunov functional will contain considerably less information on essential details of the underlying physics in the non-Markovian region. This will be clear, for instance, from Fig. 4 where wiggles in the relative entropy as a function of time clearly show that it is not Lyapunov but reproduces a complicated variation of entropy itself as due to strong interaction with the reservoir. With caution can one call this a damped exchange of entropy back and forth between system and reservoir. The caution is due to the Araki-Lieb inequality[4] for the von Neumann entropy which implies that for two coupled systems there are bounds but no exact conservation law for the sum of partial entropies. As a consequence, it can happen that entropy increases (or decreases) simultaneously in both systems.[33] In any case this oscillating phenomenon is completely absent in Markovian dynamics and, according to the concepts proposed later, considerably enhances the value of entropy production. It is obvious that in this way a marked distinction between Markovian and non-Markovian characteristics of dynamics is provided. It is difficult to see how a Lyapunov approach could reproduce the same information.

All the above arguments support our proposal to base calculations on a suitably regularized concept for relative entropy. On the one hand, all relevant properties of the conventional functional should be retained and, on the other hand, the asymmetry and the singularities should be suppressed. We shall demonstrate how this can be achieved for Hilbert spaces of finite dimensions.

In Section II.A we collect all important properties of the ordinary functional for relative entropy. This provides the necessary basis for a comparison with the regularized version, which will be presented in Section II.B. Section III is then devoted to applications. First, an extended formula for entropy production will be proposed. Subsequently, this generalized concept is used to study three examples of irreversible dynamical processes ranging from simple Markovian to complicated non-Markovian.

II. RELATIVE ENTROPY

In order to fix notation and to state basic properties we first give an account of known theorems on the ordinary relative entropy functional. Then we proceed to the proposal for a regularized version in finite dimensions.

A. Ordinary Relative Entropy

State space of an open quantum system is the positive convex cone Γ of self-adjoint and normalized trace-class operators ρ, so $\operatorname{Tr} \rho = 1 \; \forall \rho \in \Gamma$. The convex set Γ has extremal points[4, 13] represented by one-dimensional orthogonal projectors $\{p_i\}$ in terms of which the spectral decomposition for any $\rho \in \Gamma$ is given by

$$\rho = \sum_{i=1}^{\infty} \lambda_i p_i, \qquad 0 \leqslant \lambda_i \leqslant 1, \quad \sum_{i=1}^{\infty} \lambda_i = 1, \quad p_i = p_i^*, \quad p_i p_k = \delta_{ik} p_k \tag{1}$$

The absolute von Neumann entropy is then given by

$$S(\rho) = -\operatorname{Tr} \rho \ln \rho = -\sum_{i=1}^{\infty} \lambda_i \ln \lambda_i \geqslant 0 \tag{2}$$

A related functional for pairs of states is usually defined as

$$Q(\rho, \sigma) = \operatorname{Tr} \rho(\ln \rho - \ln \sigma) \geqslant 0, \qquad \forall \rho, \sigma \in \Gamma \tag{3}$$

The so-called relative entropy Q provides a measure for the entropy of one state relative to another state. Note that the minus sign in (3) does not affect nonnegativity but under the spectral restriction in (1) there is the asymmetry

$$Q(\rho, \sigma) \neq Q(\sigma, \rho) \tag{4}$$

Thus, Q does not have a distance-like property in Γ. In terms of normalized eigensolutions

$$\rho \mathbf{x}^{(i)} = \lambda_i \mathbf{x}^{(i)}, \qquad \sigma \mathbf{y}^{(i)} = \mu_i \mathbf{y}^{(i)} \tag{5}$$

one finds explicitly for Q

$$Q(\rho, \sigma) = \sum_i \lambda_i \left(\ln \lambda_i - \sum_k |(\mathbf{x}^{(i)} \cdot \mathbf{y}^{(k)})|^2 \ln \mu_k \right) \tag{6}$$

For pairs with $[\rho, \sigma] = 0$ we get

$$Q(\rho, \sigma) = \sum_i \lambda_i \ln \left(\frac{\lambda_i}{\mu_i} \right) \tag{7}$$

From formulas (6) and (7) it is manifest that $Q = \infty$ whenever one or more eigenvalues of σ are zero or, in other words, whenever σ is a non-faithful state.

In the following we list further important properties of Q.

(1) The mapping $(\rho, \sigma) \to Q(\rho, \sigma)$ is unitarily invariant, jointly convex and lower semicontinuous,[3, 4, 16, 17]

$$Q(U\rho U^*, U\sigma U^*) = Q(\rho, \sigma), \qquad UU^* = U^*U = 1 \tag{8}$$

$$Q(\alpha\rho_1 + \beta\rho_2, \alpha\sigma_1 + \beta\sigma_2) \leqslant \alpha Q(\rho_1, \sigma_1) + \beta Q(\rho_2, \sigma_2), \qquad \alpha + \beta = 1 \tag{9}$$

Lower semicontinuity is an essential analytical property of convex functions.[4, 14, 15]

(2) If we consider a composition of two independent systems and form states $\Omega = \rho \otimes \tau$ and $\Theta = \sigma \otimes \tau$ then the value of Q remains unaltered,[4, 20]

$$Q(\Omega, \Theta) = Q(\rho, \sigma) \tag{10}$$

(3) For a composition of two interacting systems with Hilbert spaces $\mathscr{H}_1$ and $\mathscr{H}_2$ two different total density operators Ω and Θ acting on

$\mathscr{H} = \mathscr{H}_1 \otimes \mathscr{H}_2$ are no longer tensor products. The corresponding operators restricted to $\mathscr{H}_1$ are obtained by taking partial traces, $\rho = \mathrm{Tr}_2(\Omega)$ and $\sigma = \mathrm{Tr}_2(\Theta)$. In this case, Q has the contractive property[4, 20]

$$Q(\rho, \sigma) \leqslant Q(\Omega, \Theta) \tag{11}$$

More general, if $\mathscr{A}$ is the operator algebra of a finite quantum system and φ and ω are states on $\mathscr{A}$ with $\varphi(a) = \mathrm{Tr}(\rho a)$, $\omega(a) = \mathrm{Tr}(\sigma a)$, $a \in \mathscr{A}$, one may consider a restriction to a subalgebra $\mathscr{B} \subset \mathscr{A}$ with corresponding states $\varphi/\mathscr{B}$ and $\omega/\mathscr{B}$. Again, one proves[5] that

$$Q(\varphi/\mathscr{B}, \omega/\mathscr{B}) \leqslant Q(\varphi, \omega) \tag{12}$$

(4) Another fundamental contractive property exists if the time evolution of ρ and σ is determined by a quantum dynamical semigroup Λ_t, $t \geqslant 0$, and if the dual Λ_t^* acting on the operator algebra of observables is completely positive.[9, 18, 19] Then for $\rho_t = \Lambda_t \rho$ and $\sigma_t = \Lambda_t \sigma$ one can prove[20] that

$$Q(\rho_t, \sigma_t) \leqslant Q(\rho, \sigma) \tag{13}$$

This inequality is of particular importance if σ is the unique final destination state of Λ_t, $\lim_{t \to \infty} \Lambda_t \rho = \sigma$, or, else, $\Lambda_t \sigma = \sigma$, and, therefore, $Q(\rho_t, \sigma) \leqslant Q(\rho, \sigma)$. Under these conditions entropy production P_Λ is determined by Q according to refs. 10 and 11

$$P_\Lambda(\rho, \sigma) = -\left[\frac{d}{dt} Q(\Lambda_t \rho, \sigma)\right]_{t=0} \tag{14}$$

Consequently, for faithful states and finite dimensions P_Λ is entirely determined by the infinitesimal generator L of $\Lambda_t = \exp[Lt]$ through

$$P_\Lambda(\rho, \sigma) = \mathrm{Tr}\, L(\rho)(\ln \sigma - \ln \rho) \tag{15}$$

Again, $P_\Lambda(\rho, \sigma)$ is jointly convex, a stability property originating ultimately from the one provided by the concavity of $S(\rho)$.

(5) There is a lower bound on Q in terms of the trace norm[5]

$$Q(\rho, \sigma) \geqslant \tfrac{1}{2} \|\rho - \sigma\|^2 \tag{16}$$

where the norm of an operator A is defined by $\|A\| = \mathrm{Tr}(A^* A)^{1/2}$. A somewhat cruder but more explicit bound can be found by making use of the ordered sets of eigenvalues $\{\lambda_1 \geqslant \lambda_2 \cdots\}$ of ρ and $\{\mu_1 \geqslant \mu_2 \cdots\}$ of σ. One gets

$$Q(\rho, \sigma) \geqslant \frac{1}{2}\left(\sum_{i=1}^{\infty} |\lambda_i - \mu_i|\right)^2 \tag{17}$$

The idea that Q might represent something like a distance in state space is obviously suggested by the above semi-inequalities.

(6) An upper bound can also be established by using a special case of Lieb's concavity theorem.[16] This leads to the result

$$Q(\rho, \sigma) \leqslant \mathrm{Tr}(\rho^2 \sigma^{-1}) - 1 \tag{18}$$

(7) The relevance of Q for thermodynamic stability can be seen as follows.[4] For different isothermal states ρ the free energy $F_\beta(\rho)$ attains its minimum for the canonical Gibbs state $\sigma = \exp[-\beta H]/Z$, H the Hamiltonian, β the inverse temperature and Z the partition function. Thus, $[F_\beta(\rho) - F_\beta(\sigma)]$ is positive whenever $\rho \neq \sigma$. It is interesting to note that under these conditions one proves that

$$Q(\rho, \sigma) = \beta[F_\beta(\rho) - F_\beta(\sigma)] \geqslant 0 \tag{19}$$

B. Regularized Relative Entropy

The following considerations are restricted to a Hilbert space $\mathscr{H}$ with $\dim \mathscr{H} = N < \infty$, so that the states are represented by $N \times N$-matrices. The search for a regularized functional $R(\rho, \sigma)$ which avoids any singularities, is guided by the fact that the von Neumann entropy takes on a unique maximum for the central state ξ,

$$\xi = \frac{1}{N}\mathbf{1}_N, \qquad S(\xi) = \ln N \tag{20}$$

Therefore we require that R possesses also an upper bound, which is proportional to $S(\xi)$. Furthermore, it is still desirable that R provides a kind of relative measure as symmetric as possible in the two arguments. Finally, and most crucially, R should preserve as many properties of the ordinary relative entropy as possible. All this should then yield an acceptable candidate for an entropy production in irreversible processes, in particular those involving also non-faithful states.

The original idea for the functional Q was based on Klein's inequality[4, 13, 21] which, in a somewhat more general formulation, states that for any two nonnegative operators A and B there exists a nonnegative form $F(A, B)$ given by

$$F(A, B) = A \ln A - A \ln B - A + B \geqslant 0, \qquad A, B \geqslant 0 \tag{21}$$

For density operators the right-hand side reduces directly to Q upon taking a trace. It is essential to recognize that for invertible B there exists an upper bound on F. Therefore, we replace the ordinary density operators by invertible operators. We introduce spectrally shifted operators $\tilde{\rho}$ and normalized operators $\hat{\rho}$ by

$$\tilde{\rho} = \rho + \mathbf{1}_N, \qquad \hat{\rho} = c_N \tilde{\rho}, \qquad c_N = (1 + N)^{-1}, \qquad \operatorname{Tr} \hat{\rho} = 1 \tag{22}$$

and define the regularized version R of the relative entropy by

$$R(\rho, \sigma) \triangleq \alpha_N Q(\hat{\rho}, \hat{\sigma}) \tag{23}$$

We emphasize that a positive spectral shift does not affect the important convexity properties. The proportionality factor α_N is fixed by the requirement that for any pure state $\rho_o = \rho_o^2$ relative to the central state, R takes on its maximum value. The constraint

$$R(\rho_0, \xi) = S(\xi) = \ln N \tag{24}$$

leads to the result

$$\alpha_N = \frac{(N+1) \ln N}{(N+1) \ln(N/(N+1)) + 2 \ln 2} \tag{25}$$

Whereas there is an upper bound by construction no further modifications are necessary to derive the lower bound on R. Note first that Q is homogeneous of degree one and we can write

$$R(\rho, \sigma) = \alpha_N c_N Q(\tilde{\rho}, \tilde{\sigma}) \tag{26}$$

It is thus sufficient to consider $Q(\tilde{\rho}, \tilde{\sigma})$. For any difference $(\rho - \sigma)$ we define p to be the spectral projection on the nonnegative part of the spectrum, and q to be the complement such that $p + q = \mathbf{1}_N$. In terms of the traces

$$x = \operatorname{Tr}(p\rho), \qquad y = \operatorname{Tr}(p\sigma), \qquad 0 \leqslant x, y \leqslant 1 \tag{27}$$

the norm can be written as

$$\|\rho - \sigma\| = 2(y - x) \tag{28}$$

where, without loss of generality, we have assumed $y \geqslant x$. Consider now the two commuting states

$$\tilde{\rho}_p = \begin{pmatrix} 1+x & 0 \\ 0 & 2-x \end{pmatrix}, \qquad \tilde{\sigma}_p = \begin{pmatrix} 1+y & 0 \\ 0 & 2-y \end{pmatrix} \tag{29}$$

and the inequality

$$f(x, y) \geqslant 0 \tag{30}$$

for the function f defined by

$$\begin{aligned} f(x, y) &= Q(\tilde{\rho}_p, \tilde{\sigma}_p) - \frac{2}{3}(x-y)^2 \\ &= (1+x)\ln\left(\frac{1+x}{1+y}\right) + (2-x)\ln\left(\frac{2-x}{2-y}\right) - \frac{2}{3}(x-y)^2 \end{aligned} \tag{31}$$

The proof of (30) goes by elementary analysis, and shows also that the equality sign applies to the unique case $x = y$. From (28) and (31) one concludes that

$$(6Q[\tilde{\rho}_p, \tilde{\sigma}_p])^{1/2} \geqslant 2(y-x) = \|\rho - \sigma\| \tag{32}$$

Since projectors p and q generate an Abelian subalgebra, relation (12) applies in the form

$$Q(\tilde{\rho}, \tilde{\sigma}) \geqslant Q(\tilde{\rho}_p, \tilde{\sigma}_p) \tag{33}$$

The desired final result is then found to be

$$R(\rho, \sigma) \geqslant \tfrac{1}{6}\alpha_N c_N \|\rho - \sigma\|^2 \tag{34}$$

Owing to our regularization it is now possible to calculate the relative entropy between two different pure states with projectors $\rho = p = p^2$ and $\sigma = q = q^2$ and $[p, q] = 0$. For instance, for $N = 2$ one finds

$$R(p, q) = (\ln 2)^2/\ln(\tfrac{32}{27}) \tag{35}$$

whereas $Q(p, q) = \infty$ follows directly from (7). The general formula for $N \geqslant 2$ reads

$$R(p, q) = \frac{(\ln 2)(\ln N)}{(N+1)\ln(N/(N+1)) + 2\ln 2} \tag{36}$$

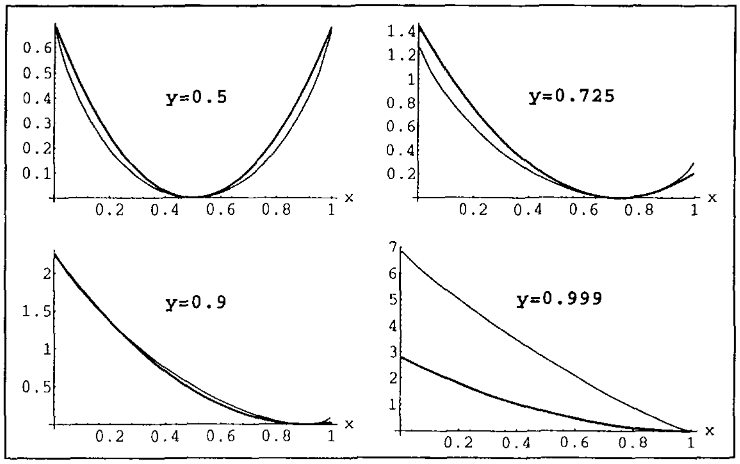

Fig. 1. Comparison between $Q(x, y)$ [thin line] and $R(x, y)$ [thick line] for various final states.

A comparison between the ordinary relative entropy $Q(\rho, \sigma)$ and the regularized version $R(\rho, \sigma)$ shows the following qualitative trends. First of all, for σ not too close to a non-faithful state the function values are in satisfactory agreement with each other. Whenever σ approaches a singular state, Q diverges whereas R remains finite by construction. An illustration is shown in Fig. 1 for $N=2$ with

$$\rho = \begin{pmatrix} x & 0 \\ 0 & 1-x \end{pmatrix}, \qquad \sigma = \begin{pmatrix} y & 0 \\ 0 & 1-y \end{pmatrix}, \qquad \begin{matrix} 0 \leqslant x \leqslant 1 \\ \frac{1}{2} \leqslant y < 1 \end{matrix} \tag{37}$$

where we use the abbreviated notation $Q(x, y)$ and $R(x, y)$.

Second, the lack of symmetry under exchange of arguments is not transferred from Q to R. The regularized functional is almost symmetric, and thus can be very well regarded as a distance in state space. The respective differences are shown in Fig. 2.

Apart from these most convenient extra properties of R, the important general properties of Q listed in Section 2.A are shared by R in the following sense:

(i) Relation (9) is literally true for $R(\rho, \sigma) = \alpha_N Q(\hat{\rho}, \hat{\sigma})$, since the proof for Q only assumes positive operators.[16, 17] Thus we have

$$R(\alpha\rho_1 + \beta\rho_2, \alpha\sigma_1 + \beta\sigma_2) \leqslant \alpha R(\rho_1, \sigma_1) + \beta R(\rho_2, \sigma_2), \qquad \alpha + \beta = 1 \tag{38}$$

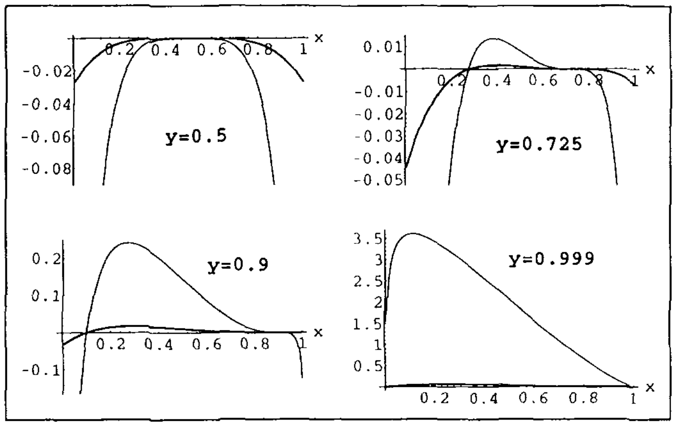

Fig. 2. Comparison of asymmetry under exchange of arguments for various final states. $Q(x, y) - Q(y, x)$ [thin line]; $R(x, y) - R(y, x)$ [thick line].

(ii) For $\hat{\Omega} = \hat{\rho} \otimes \hat{\tau}$ and $\hat{\Theta} = \hat{\sigma} \otimes \hat{\tau}$ where $\hat{\rho}, \hat{\sigma}$ are assumed to be $m \times m$ and $\hat{\tau}$ is $n \times n$, (10) translates into

$$R(\Omega, \Theta) = \frac{\alpha_{nm}}{\alpha_m} R(\rho, \sigma) \tag{39}$$

(iii) Contractivity in the sense of (11) cannot be proven in the form as it stands, due to the scaling factor α_N which depends on dimension. Nevertheless, numerical tests show that $R(\rho, \sigma) \leqslant R(\Omega, \Theta)$ is slightly violated in a few exceptional cases only. On the other hand, contractivity under completely positive maps remains true in analogy to (13). Again, we have

$$R(\rho_t, \sigma_t) \leqslant R(\rho, \sigma) \tag{40}$$

because Lindblad's proof[17] is valid for all normalized positive operators.

(iv) As has been proven already, the lower bound (16) is replaced by (34) whereas in analogy to (18) an upper bound is given by

$$R(\rho, \sigma) \leqslant \alpha_N [\mathrm{Tr}(\hat{\rho}^2 \hat{\sigma}^{-1}) - 1] \tag{41}$$

(v) Finally, one may inquire about the effect of the spectral shift on the von Neumann entropy itself. We denote by $\hat{S}(\rho)$ a suitable modification of $S(\hat{\rho})$ such that

$$\hat{S}(\xi) = S(\xi) = \ln N \tag{42}$$

Note that the central state $\hat{\xi} = \xi$ is a fixed point of the regularization procedure. The desired formula replacing (2) is then found to be given by

$$\hat{S}(\rho) = \alpha_N S(\hat{\rho}) - (\alpha_N - 1) \ln N \tag{43}$$

Again, $\hat{S}$ turns out to be zero on pure states and takes on its unique maximum equal to $\ln N$ for the central state in complete analogy to S.

III. APPLICATIONS TO MODELS OF IRREVERSIBLE DYNAMICS

In order to establish a generally meaningful notion of entropy production we recall that formula (14) is valid for completely positive maps and faithful states. For more general dynamics involving also arbitrary states we may assume that the time-derivative $\dot{R}$ of the regularized relative entropy plays a decisive role. Furthermore, we shall assume that the density operator of any dynamical evolution has a well-defined unique limit $\sigma = \lim_{t \to \infty} \rho_t$. Under these conditions the formula

$$P = \frac{1}{\tau} \int_0^\infty |\dot{R}(\rho_t, \sigma)| \, dt \tag{44}$$

provides a reasonable measure of entropy production. Variable τ denotes an average lifetime defined by

$$\tau = \frac{1}{\|\rho_o - \sigma\|} \int_0^\infty \|\rho_t - \sigma\| \, dt \tag{45}$$

This choice is compatible with (14) in the following sense. Under the assumption that the upper limit in the above integrals is replaced by a small time ε such that $\dot{R}(t) \equiv \dot{R}(\rho_t, \sigma)$ and ρ_t vary only slightly, one may write

$$|\dot{R}(\varepsilon)| \cong |\dot{R}(0)| + \alpha\varepsilon + O(\varepsilon^2)$$

$$\|\rho_\varepsilon - \sigma\| \cong \|\rho_o - \sigma\| + \beta\varepsilon + O(\varepsilon^2)$$

with α, β some constants. To first order in ε one obtains $P \cong |\dot{R}(0)|$, in agreement with (14). Note that in this case $\dot{R}(0)$ is always negative owing to (13). The following examples, however, refer to cases where (14) cannot be used.

A. Photon Emission

The emission of a quantum of energy ω by an open two-level system is described by the simple master equation[22]

$$\dot{\rho}_t = -\frac{1}{2} i\omega[s_3, \rho_t] + \frac{1}{2}\gamma\{2s_-\rho_t s_+ - s_+s_-\rho_t - \rho_t s_+ s_-\}, \qquad \rho_o = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \tag{46}$$

Vector $\mathbf{s} = (s_1, s_2, s_3)^T$ contains the Pauli matrices, $s_\pm = (1/2)(s_1 \pm is_2)$, and γ denotes the Einstein A-coefficient. The right-hand side of (46) generates a completely positive semigroup dynamics Λ_t with simple solution

$$\rho_t = \Lambda_t \rho_o = \begin{pmatrix} e^{-\gamma t} & 0 \\ 0 & 1 - e^{-\gamma t} \end{pmatrix}, \qquad t \geqslant 0 \tag{47}$$

such that the pure excited state deactivates into the pure ground state,

$$\rho_o = \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix} \xrightarrow[t \to \infty]{\Lambda_t} \sigma \equiv \rho_\infty = \begin{pmatrix} 0 & 0 \\ 0 & 1 \end{pmatrix} \tag{48}$$

The time-dependent relative entropy becomes

$$R(\rho_t, \sigma) = \frac{\alpha_2}{3}\left\{(1 + e^{-\gamma t}) \ln(1 + e^{-\gamma t}) + (2 - e^{-\gamma t}) \ln\left(1 - \frac{1}{2} e^{-\gamma t}\right)\right\} \tag{49}$$

where $\alpha_2 = (3 \ln 2)/(5 \ln 2 - 3 \ln 3)$. For integrations on a finite interval $[0, T]$ the corresponding quantities will be denoted by P_T and τ_T. This yields the results

$$\tau_T = (1 - z)/\gamma, \qquad z = e^{-\gamma T} \tag{50}$$

$$P_T = \frac{\gamma \alpha_2}{3(1 - z)}\{\ln 2 + (z - 2) \ln(1 - z/2) - (1 + z) \ln(1 + z)\} \tag{51}$$

The total entropy production is then obtained in the limit $T \to \infty$ or, alternatively, for $z = 0$

$$P = \gamma\left(\frac{\alpha_2 \ln 2}{3}\right) \tag{52}$$

As expected, the result is proportional to the inverse lifetime of the excited state. In contrast to any other available formula, (44) yields a finite value for a physically common irreversible process connecting two pure states via the central state. Representing ρ_t through a coherence vector $\mathbf{x}_t$ according to

$$\rho_t = \xi + (\mathbf{x}_t \cdot \mathbf{s}), \qquad \mathbf{x}_t = [u(t), v(t), w(t)]^T \tag{53}$$

one can characterize the model by

$$u(t) = v(t) = 0, \qquad w(t) = e^{-\gamma t} - \tfrac{1}{2}, \qquad \xi = \tfrac{1}{2}\mathbf{1}_2 \tag{54}$$

State space is made up by the full Bloch sphere with $\|\mathbf{x}_t\| \leqslant 1/2$ and the trajectory $w(t)$ passes from, say, the north pole straight through the origin to the south pole. The corresponding von Neumann entropy starts from zero, reaches its maximum of ln 2 after a time $\tau_T = (\ln 2)/\gamma$ and drops back to zero in the limit $t \to \infty$.

A more complicated situation will be presented in the next example.

B. General Relaxation in a 4-Level System

Consider the general Markovian master equation

$$\dot{\rho}_t = \mathscr{L}\rho_t \tag{55}$$

with infinitesimal generator $\mathscr{L}$ of Kossakowski-type[23]

$$\mathscr{L}\rho_t = -i[H, \rho_t] + \tfrac{1}{2} \sum_{i,k=1}^{15} a_{ik}\{[F_i, \rho_t F_k] + [F_i \rho_t, F_k]\} \tag{56}$$

guaranteeing complete positivity. For the complete and orthonormalized hermitian matrix set $\{F_i\}_1^{15}$ we use a standard representation of infinitesimal generators of $SU(4)$.[9] The basic requirement on the complex relaxation matrix A is that it be nonnegative,

$$A = \{a_{ik}\}_1^{15} \geqslant 0 \tag{57}$$

It therefore may be taken as hermitian. For a numerical solution of (55) with given initial condition ρ_o we choose a Hamiltonian H and a relaxation matrix A by some random procedure. Then, (56) is transformed into coherence vector representation[9] to obtain 15 coupled ordinary differential equations for real-valued functions $\mathbf{x}_t = [x_1(t), x_2(t),..., x_{15}(t)]^T$. They have the form

$$\dot{\mathbf{x}}_t = G\mathbf{x}_t + \mathbf{k} \tag{58}$$

where matrix G and vector $\mathbf{k}$ are determined by the elements of A. G is asymmetric with eigenvalues $\{g_i\} \in \mathscr{C}$. We always keep it regular with $\Re(g_i) < 0$ $(\forall i)$ so as to obtain genuine relaxation into a unique final state. In terms of functions $\{\mathbf{x}_i(t)\}$ the solution of (55) is given by

$$\rho_t = \xi + \sum_{i=1}^{15} x_i(t)\, F_i, \qquad \xi = \tfrac{1}{4}\mathbf{1}_4 \tag{59}$$

with the constraint

$$0 \leqslant \|\mathbf{x}_t\|^2 \leqslant \tfrac{3}{4} \tag{60}$$

The evolution matrix G in (58) decomposes into

$$G = Q_H + R_A \tag{61}$$

where Q_H is a skew-symmetric matrix related to H and R_A is a non-symmetric matrix which arises from the relaxation A.[9]

In the following numerical evaluations the first case refers to the dependence of P and τ on the strength of relaxation which is measured by means of the Frobenius norm $\|G\|$. This norm is used because for non-commuting Q_H and R_A there is also a Hamiltonian contribution to relaxation and entropy production, as has been derived in ref. 9. Figure 3(a) shows the results for fixed initial condition ρ_o (a pure state) and fixed H. Two regimes can be distinguished. If R_A dominates the Hamiltonian contribution Q_H, then the dependence of P and τ on $\|G\|$ is almost entirely determined by the dependence on $\|R_A\|$ and turns out to be linear. For smaller R_A the dynamics tends towards weakly damped quasi-periodic behavior

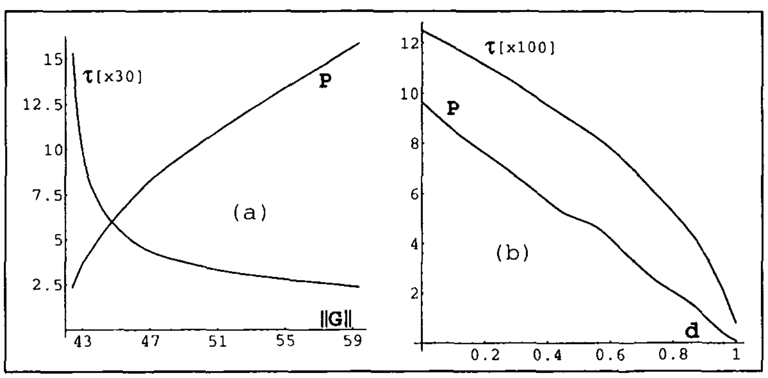

Fig. 3. Dependence of entropy production P and lifetime τ on the norm of the evolution matrix G in (a) and on the degree of mixture d in (b) [arbitrary units].

and P decreases more strongly whereas the lifetime τ increases strongly. Clearly, for $\|G\| = \|Q_H\|$ the entropy production must equal zero and the lifetime becomes infinite. This is the reason for bending over of the two curves for lower values of $\|G\|$. Qualitatively, this shows that definition (44) is reasonable, indeed. The relatively smooth behavior will always be characteristic of Markovian dynamics although single components $x_i(t)$ may show quite a strong time dependence.

In the second case H and A and, consequently, Q_H and R_A are kept fixed. Figure 3(b) then shows the variation of P and τ as a function of the initial state ρ_o. The degree of mixture d with $0 \leqslant d \leqslant 1$ is defined by means of the coherence vector representing ρ_o,

$$d = 1 - \tfrac{4}{3} \|\mathbf{x}_o\|^2 \tag{62}$$

Note that $d = 0$ refers to any pure state, whereas $d = 1$ is uniquely obtained for the central state ξ. Again, as qualitatively expected, the closer ρ_o to the final state σ (which is very close to ζ in this numerical example) the smaller are the values of P and τ. This conforms to the geometrical picture that the length of trajectories through the 15-dimensional state space decreases as the degree of mixture d increases.

C. Non-Markovian Dynamics

General dynamics of open quantum systems with factorizable initial state is described by an integro-differential equation of the Zwanzig-type,[25, 26]

$$\dot{\rho}_t = -i[H, \rho_t] + \int_0^t K(t, s)\, \rho_s\, ds \tag{63}$$

For a two-level system, for instance, the solution may give rise to complicated trajectories through the Bloch sphere and may provide an interesting test of the variation of P. For this purpose the solutions are chosen such that, as in Section III.A, the trajectories connect the two poles via the center but exhibit much more complicated behavior in the upper and lower hemispheres. A suitable choice is

$$u(t) = f(t) \cos \omega_1 t \tag{64}$$

$$v(t) = f(t) \sin \omega_2 t \tag{65}$$

$$w(t) = e^{-t} - \tfrac{1}{2} \tag{66}$$

where the time-function f is given by

$$f(t) = t(t - t_3)\{Ae^{-a(t-t_1)^2} + Be^{-a(t-t_2)^2}\} \tag{67}$$

One should remark that by known methods[26] the kernel $K(t, s)$ can be reconstructed from the above choice. Note also that a Markovian master equation can never possess solutions of the above type.[24]

Whereas ω_2 will be varied, the other parameters remain fixed with values

$$\begin{aligned} &t_1 = \ln\left(\frac{10}{7}\right), \qquad t_2 = \ln\left(\frac{10}{3}\right), \qquad t_3 = \ln 2 \\ &A = \sqrt{8}, \qquad B = \frac{\sqrt{2}}{10}, \qquad a = 3, \qquad \omega_1 = 80 \end{aligned} \tag{68}$$

(units are omitted throughout). The numerical evaluation of R is facilitated by using a formula in terms of the length $x_t \equiv \|\mathbf{x}_t\|$ of the coherence vector. One verifies[9] that

$$\begin{aligned} \frac{3}{\alpha_2} R(t) = {} & \frac{3}{2} \ln\left(\frac{9 - 4x_t^2}{9 - 4x_\infty^2}\right) + 2x_t \tanh^{-1}\left(\frac{2x_t}{3}\right) \\ & - \frac{2}{x_\infty} (\mathbf{x}_t \cdot \mathbf{x}_\infty) \tanh^{-1}\left(\frac{2x_\infty}{3}\right) \end{aligned} \tag{69}$$

with time-derivative given by

$$\frac{3}{\alpha_2} \dot{R}(t) = 2\dot{x}_t \tanh^{-1}\left(\frac{2x_t}{3}\right) - \frac{2}{x_\infty} (\dot{\mathbf{x}}_t \cdot \mathbf{x}_\infty) \tanh^{-1}\left(\frac{2x_\infty}{3}\right) \tag{70}$$

As an illustration, three cases are studied for zero, small and larger detuning between the two frequencies ω_1 and ω_2. As shown in Fig. 4, an increase of detuning leads to a stronger variation of the coherence vector length and, thus, to oscillations in the relative entropy. This can be ascribed to an increasing complexity of the trajectories. As a consequence, one expects that the entropy production as defined by (44) increases as well. In fact, the results are $P = 2.7$ for (a), $P = 11.5$ for (b), and $P = 16$ for (c). On the other hand, since the relaxation constants have been kept fixed, the lifetime should be the same in all three cases. In addition, one may define a measure of irreversibility κ in terms of relative entropy. The following formula is based on the idea that the partly reversible, oscillating

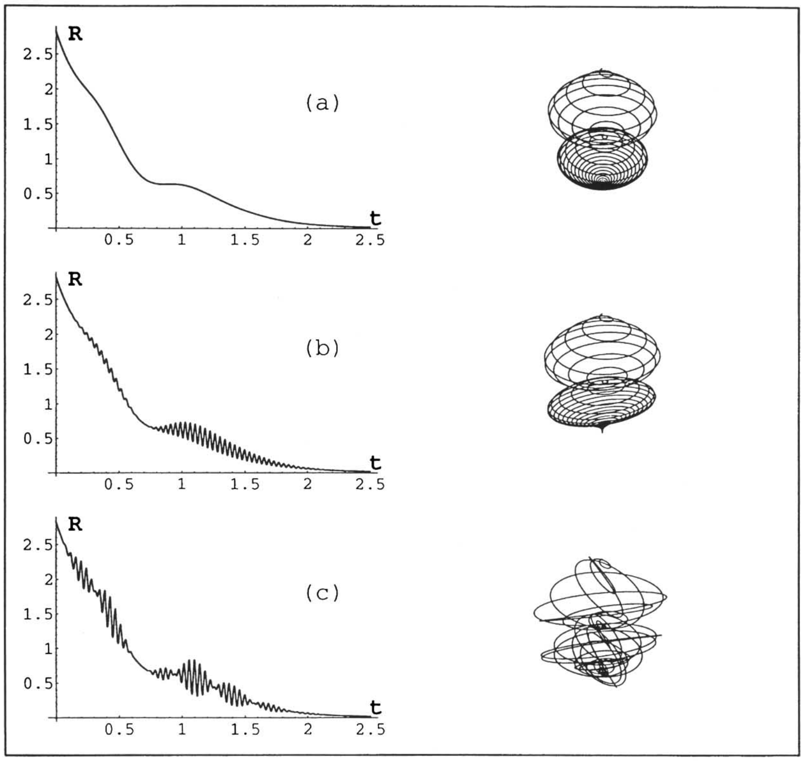

Fig. 4. Time dependence of relative entropy on the left with corresponding trajectories confined to the Bloch sphere on the right (the sphere itself is not shown). (a) $\omega_2 = 80$; (b) $\omega_2 = 80.5$; (c) $\omega_2 = 70$ [arbitrary units]. In all cases, $\omega_1 = 80$.

exchange of entropy between system and reservoir is averaged out by just integrating over R itself. Thus we propose

$$\kappa = \frac{1}{R(0)\,\tau} \int_0^{\infty} R(t)\,dt \tag{71}$$

and, again, κ is expected to adopt the same value in all three cases. All expectations are confirmed by a numerical evaluation of (45) and (71), which yields $\tau = 1.1$ and $\kappa = 0.54$ for all three processes (a), (b) and (c).

IV. CONCLUSION

The fundamental role of the concept of entropy production has been stressed since the early work on nonequilibrium thermodynamics for classical macroscopic systems (see refs. 27, 28 and references therein). However, the meaning of nonequilibrium implied states not too far from thermodynamic equilibrium where important stability properties of thermodynamic potentials should still hold. Under such assumptions the dynamics of irreversible processes may be expected to be relatively simple with time dependence of observables essentially given by some exponential decay laws from any nonequilibrium initial value back to its final equilibrium value. For small open systems the quantum analog of such dynamics will be given in terms of Markovian master equations and this explains why the first mathematically sound attempts towards a quantum formulation of entropy production were possible only after the theory of completely positive quantum dynamical semigroups had been established.[8, 10–12, 29]

The extension of the afore-mentioned notions to more general situations of nonequilibrium proved difficult. Nevertheless, very recent activities for classical systems must be mentioned where attempts are made to define entropy production for wide classes of dynamical systems (see refs. 30, 31 and references therein). In particular, Ruelle[30] gives definitions and proofs of positivity for iterated diffeomorfic dynamical maps under rather general assumptions.

For the quantum case we hope to propose also a reasonable extension as outlined in the preceding Sections. On the one hand, the weakly irreversible regime should be reproduced in the known way, and on the other hand, any process involving arbitrary pairs of initial and final states connected by either Markovian or non-Markovian dynamics should be allowed. This clearly leads to the task of generalizing relative entropy to an everywhere finite-valued functional. The details presented in Section II.B. set clear boundaries to the choice of a regularized functional such that by far not any regularization would be acceptable. Summarizing again, the physically important properties listed in Section II.A. for the ordinary functional should be shared by the regularized version, supplemented by symmetry and finiteness on the entire state space. After all, it turned out that a homogeneous spectral shift by unity is the distinguished choice. There remains a certain freedom in the determination of scaling α_N in (23) but the proposed constraint (24) is obviously the minimum which must be required. In any case it is surprising how closely related the new version is in comparison to the old one whenever the latter is well-defined.

Now, the problem of positivity needs some special remarks. The physical meaning of entropy production has been defined in the conventional nonequilibrium theory essentially by a continuity equation[27] and, as explained there, the values should be positive (nonnegative). Whenever an explicit mathematical definition for a class of processes has been chosen there is still need for a proof of positivity, in the classical[30] as well as in the quantum case.[10] Our proposal of P in (44) replaces the minus sign in (14) by the absolute value of the integrand and, as explained in the introduction to this section, adds nothing new to proven results in the weakly irreversible regime. For more general situations, particularly for those dynamical evolutions which are not described by Markovian master equations, the extension remains positive. Whenever there is a non-monotonous decrease of relative entropy, as for instance in our third example in Section III.C, there is a time-dependent exchange of entropy back and forth between open system and reservoir (the latter is not treated explicitly). The value of the production is then obviously determined by piling up all these oscillatory contributions with positive sign, too.

Finally, the three examples have been chosen in order to illustrate physically relevant dynamical cases not covered by conventional treatments. Whereas the first two obviously are of physical relevance, the third one might seem to be only of purely mathematical interest. On the contrary, we will show in a forthcoming paper[33] that a similarly complicated, highly non-Markovian dynamics can be derived and exactly solved for a real physical situation under experimentally accessible conditions. The analysis of entropy variation and of production rates will reveal the various problems encountered in a formulation for general dynamics.

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