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Hyperbolic Relaxation as a Sufficient Condition of a Fully Coherent Ergodic Field

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In three cases, one originating from a classical model, the second from the time-evolution operator, and the third from photocount statistics, it is shown that an initially excited coherent field which remains coherent in time development relaxes according to a hyperbolic rather than to an exponential law. This has particular relevance for the analysis of biological systems.

1. INTRODUCTION

It is well known that relaxing ergodic systems subject to a linear dependence of the intensities $\dot{n}$ upon the number of radiating atoms or molecules n display an exponential decay law $n \propto \exp(-\lambda t)$, where λ is the decay constant and t the time. If the coupling becomes nonlinear, e.g., $\dot{n} \propto n^2$, it is obvious that a hyperbolic law of the form $n \propto 1/t$ then represents the corresponding solution of the problem.

Many papers (Weisskopf and Wigner, 1931; Heitler and Ma, 1949; Ersak, 1969; Davies, 1975; Fonda *et al.*, 1978; Bunge and Kalnay, 1983; Li and Popp, 1983) have been devoted to the quantum description of exponential decay and its basic origins and problems. However, no investigation reveals the basis of the hyperbolic relaxation within the framework of quantum theory.

In addition to a semiclassical approach (Li and Popp, 1983), three striking cases are analyzed here, in order to show that a hyperbolic relaxation of an ergodic system is sufficient (but not necessary) for keeping its coherence, while an exponential one is necessary (but not sufficient) for a chaotic field.

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2. A CLASSICAL MODEL

Let us turn to the most simple case of an oscillator of amplitude $x(t)$. Instead of a constant damping λ, we introduce more generally a time-dependent damping factor $\lambda(t)$, requiring that the eigenfrequency ω_0 of the system shall remain exactly constant under relaxation.

Of course, the exponential relaxation is a consequence of a constant damping λ which changes the eigenfrequency ω_0 of the system to $\omega = (\omega_0^2 - \lambda^2)^{1/2}$. The damping energy is dissipated into heat. However, a constant eigenfrequency ω_0 provides at the same time a constant degree of coherence in terms of the visibility of the interference fringes. Systems with constant eigenfrequency ω_0 and combinations of them can communicate by means of frequency modulations independent on their amplitudes. Actually, by use of the equation

$$\ddot{x}(t) + 2\lambda(t)\,\dot{x}(t) + \omega_0^2 x(t) = 0 \tag{1}$$

where $2\lambda(t)$ describes the time-dependent damping of the system, $x(t)$ can be separated into an oscillating part $y(t)$ that keeps the frequency ω_0 stable and a decaying fraction $\exp[-\int \lambda(t)\,dt]$:

$$x(t) = \exp\left[-\int \lambda(t)\; y(t)\,dt\right] \tag{2}$$

After insertion of (2) into (1) we obtain

$$\ddot{y} + (\omega_0^2 - \lambda^2 - \dot{\lambda})\,y = 0 \tag{3}$$

Stability of ω_0 requires

$$\lambda^2 = -\dot{\lambda} \tag{4}$$

The requirement that the frequency ω_0 is independent of the changes of oscillator amplitude y is consistent with the classical coherence consideration by Schrödinger (1926).

The solution of equation (4) reads

$$\lambda(t) = \frac{\lambda_0}{1 + \lambda_0 t} \tag{5}$$

where λ_0 is a constant.

Consequently, the solution of (1) takes, under the constraint of (5), the final form

$$x(t) = \frac{y(t)}{1 + \lambda_0 t} \tag{6}$$

where $y(t)$ is periodic in time t with frequency ω_0. Equation (6) describes a hyperbolic relaxation behavior.

3. TIME-EVOLUTION

As Fonda *et al.* (1978) have shown, exponential decay is one of the possible consequences of the semigroup law

$$A(t_1)\,A(t_2) = A(t_1 + t_2) \tag{7}$$

of the time-evolution operator $A(t) = \exp[-(i/h)\int^t H(t')\,dt']$ for chaotic fields. Actually, the solution of (7) is exponential: $\langle A(t)\rangle \propto \exp(-\lambda t)$, where the real value of λ follows from the unitarity of $A(t)$ (Fonda *et al.*, 1978). Let us now start with the number of photons of a coherent field of amplitude $\kappa\alpha$, where κ represents a real parameter, for instance, a real function of time, while α accounts for a complex function of time.

Obviously we have

$$n = \kappa^2 |\alpha|^2 \tag{8}$$

In terms of the displacement operator $D(\kappa\alpha)$ (Glauber, 1963), equation (8) has to be rewritten as

$$n = \langle 0|\, D^*\,(\kappa\alpha)\, a^+ a D\,(\kappa\alpha)\,|0\rangle \tag{9}$$

where $|0\rangle$ is the vacuum state, and a^+, a are the creation and annihilation operators, respectively. They are subject to

$$[a, a^+] = 1 \tag{10}$$

and a^+a shall not explicitly depend on κ.

From (9) we obtain immediately

$$\frac{\partial n}{\partial \kappa} = \langle 0|\left(\frac{\partial}{\partial \kappa} D^*(\kappa\alpha)\right) a^+ a\, D(\kappa\alpha)\,|0\rangle + \langle 0|\, D^*(\kappa\alpha)\, a^+ a \left(\frac{\partial}{\partial \kappa} D(\kappa\alpha)\right)|0\rangle \tag{11}$$

Using Glauber's (1963) derivation

$$\frac{\partial}{\partial\kappa} D(\kappa\alpha) = (\alpha a^+ - \alpha^* a)\, D(\kappa\alpha)$$
$$\frac{\partial}{\partial\kappa} D^*(\kappa\alpha) = D^*(\kappa\alpha)(\alpha^* a - \alpha a^+) \tag{12}$$

we then get from (11)

$$\frac{\partial n}{\partial\kappa} = \langle 0|\, D^*(\kappa\alpha)[(\alpha^* a - \alpha a^+)\, a^+ a + a^+ a(\alpha a^+ - \alpha^* a)]\, D(\kappa\alpha)\, |0\rangle \tag{13}$$

For $(\alpha^* a - \alpha a^+)\, a^+ a + a^+ a(\alpha a^+ - \alpha^* a)$ we can write $\alpha a^+ + \alpha^* a$ on account of (10). Consequently

$$\frac{\partial n}{\partial\kappa} = \langle 0|\, D^*(\kappa\alpha)(\alpha a^+ + \alpha^* a)\, D(\kappa\alpha)\, |0\rangle \tag{14}$$

Mehta *et al.* (1967) have shown that a coherent state remains always coherent when the Hamiltonian $H_c(t)$ has the following form:

$$H_c(t) = \omega(t) a^+ a + [f(t) a^+ + f^*(t) a] + \beta(t) \tag{15}$$

where $\omega(t)$ and $\beta(t)$ are arbitrary real functions of t, while $f(t)$ is an arbitrary complex function of t. Any term on the r.h.s. of (15) accounts for the conservation of coherence.

Consequently, we are free in choosing $f(t)$ for $\omega \neq 0$ such that

$$f(t) a^+ + f^*(t) a = \omega(t)\, \mu(t)(\alpha a^+ + \alpha^* a) \tag{16}$$

After substitution of (15) into (14), by taking account of (16), we get

$$\frac{\partial n}{\partial\kappa} = \frac{1}{\omega(t)\, \mu(t)} \langle 0|\, D^*(\kappa\alpha)\{[H_c(t) - \beta(t)] - \omega(t) a^+ a\}\, D(\kappa\alpha)\, |0\rangle \tag{17}$$

where we have as well on the l.h.s. as on the r.h.s. of (17) only real functions. Since

$$\langle 0|\, D^*(\kappa\alpha)[H_c(t) - \beta(t)]\, D(\kappa\alpha)\, |0\rangle = \frac{\varepsilon_c(t) - \beta(t)}{n(t)}\, n(t)$$

where $\varepsilon_c(t) = \langle \kappa\alpha|\, H_c(t)\, |\kappa\alpha\rangle$, equation (17) can be written as

$$\frac{\partial n}{\partial\kappa} = -\frac{1}{\mu(t)\, \omega(t)} \left[\frac{\varepsilon_c(t) - \beta(t)}{n(t)} - \omega(t)\right] \langle 0|\, D^*(\kappa\alpha)\, a^+ a\, D(\kappa\alpha)\, |0\rangle$$
$$= -\frac{1}{\gamma(t)}\, n(t) \tag{18}$$

We are not completely free in the choice of $\gamma(t)$. Of course, the ergodicity condition requires that the ensemble average $\langle n(\bar{t})\rangle$ at any instant $\bar{t}$ equals the time average according to

$$\langle n(\bar{t})\rangle = \lim_{t_1, t_2 \to \infty} \frac{1}{t_1 + t_2} \int_{\bar{t}-t_1}^{\bar{t}+t_2} n(t)\, dt \tag{19}$$

(19) holds obviously for a stationary field $\langle n(\bar{t})\rangle = \text{const}$ for any t_1 and t_2. Let us provide that (19) shall be fulfilled also for the relaxing coherent field after excitation. n shall be a function of t only, which means that we can make the special choice $\kappa = t + t_1$ in (18), in order to evaluate the decay kinetics $\partial n/\partial\kappa \equiv \partial n/\partial t = dn/dt$ under the condition (19).

After insertion of (18) into (19) and taking $d\kappa = dt$, we then have

$$\langle n(\bar{t})\rangle = -\lim_{t_1, t_2 \to \infty} \frac{1}{t_1 + t_2} \int_{\bar{t}-t_1}^{\bar{t}+t_2} \left[\gamma(t)\frac{dn}{dt}\right] dt \tag{20}$$

After integration by parts we obtain

$$\begin{aligned}\langle n(\bar{t})\rangle = &-\lim_{t_1, t_2 \to \infty} \frac{1}{t_1 + t_2} [\gamma(t)\, n(t)]_{\bar{t}-t_1}^{\bar{t}+t_2} \\ &+ \lim_{t_1, t_2 \to \infty} \frac{1}{t_1 + t_2} \int_{\bar{t}-t_1}^{\bar{t}+t_2} \left(\frac{d\gamma}{dt}\right) n(t)\, dt\end{aligned} \tag{21}$$

Since $\langle n(\bar{t})\rangle$ shall not depend on the special choice of t_1, t_2 even if t_1, t_2 remain finite, the solution of $\gamma(t)$ has to satisfy the condition

$$[\gamma(t)\, n(t)]_{\bar{t}-t_1}^{\bar{t}+t_2} = \{\gamma(t)\, n(t)\}_{\bar{t}+t_2} - \{\gamma(t)\, n(t)\}_{\bar{t}-t_1} = 0 \tag{22}$$

The equality of (21) and (19) at any t_1 and t_2 requires then

$$\frac{d\gamma}{dt} = 1$$

or

$$\gamma(t) = t + t_0 = \kappa + (t_0 - t_1) \tag{23}$$

where t_0 and t_1 are arbitrary constant times.

Consequently, for an ergodic field, the relaxation of $n(t)$ after excitation satisfies condition (18) under the constraint of (19):

$$\frac{dn(t)}{dt} = -\frac{1}{t + t_0}\, n(t) \tag{24}$$

Equation (24) results in a hyperbolic relaxation

$$n(t) = \frac{n(0)\, t_0}{t + t_0} \tag{25}$$

where $n(0)$ is the photon number at time $t = 0$. The condition

$$[\gamma(t)\, n(t)]_{t-t_1}^{t+t_2} = 0$$

according to (22) is as well satisfied. Thus, the solution is self-consistent.

Equation (25) is sufficient for coherent relaxation under ergodic conditions, since the selection of a special case [defined by (19) and the choice $\kappa = t + t_0$] from a collection of possible cases which do not overlap with chaotic (exponential) relaxation involves a sufficient but certainly not a necessary condition. This means that if a hyperbolic relaxation under ergodic conditions has been registered, the decay products can originate only from a fully coherent field, while the measurement of an exponential decay function cannot necessarily be assigned to a chaotic field. On the other hand, a chaotic field which is an eigenstate of a^+a under ergodic conditions always decays according to an exponential function (Fonda *et al.*, 1978).

It should be mentioned that in contrast to the classical oscillator for this solution it is no longer necessary to keep the frequency constant.

In (24) the real time t can be assigned to the coherence time τ. As long as the relaxation of a fully coherent field takes place, there is no difference between τ and the real time t, while for a chaotic field $t + t_0$ on the r.h.s. of (24) has to be replaced by the reciprocal of the decay constant (Popp, 1986).

In order to show that (25) reflects a limiting case like that of the equality in case of the Schwartz inequality equation, let us briefly discuss the semigroup law (7). It can be formulated under the constraint of a fully coherent field in such a way that again a hyperbolic law is obtained.

Of course, if equation (7) is replaced by

$$A(t_1)\, A(t_2) = A\left(\frac{t_1 + t_2}{2}\right) \frac{1}{2} [A(t_1) + A(t_2)] \tag{26}$$

the exponential solution turns for $t_1 \neq t_2$ into the hyperbolic $A(t) = c/t$, where c is a constant:

$$\frac{c}{t_1} \frac{c}{t_2} = \left(\frac{2c}{t_1 + t_2}\right) \frac{1}{2} \left(\frac{c}{t_1} + \frac{c}{t_2}\right)$$

Because of

$$A(t_1)\,A(t_2)=A\left(\frac{t_1+t_2}{2}\right)A\left(\frac{t_1+t_2}{2}\right) \tag{27}$$

the change from (7) to (26) corresponds to the substitution of

$$A\left(\frac{t_1+t_2}{2}\right)=\frac{1}{2}\,[A(t_1)+A(t_2)] \tag{28}$$

By use of the identities

$$A\left(\frac{t_1+t_2}{2}\right)=[A(t_1+t_2)]^{1/2}=[A(t_1)\,A(t_2)]^{1/2}$$

we can see from (28) that the geometrical mean $[A(t_1)\,A(t_2)]^{1/2}$ has then to be substituted by its arithmetic mean $\frac{1}{2}[A(t_1)+A(t_2)]$ in order to turn from (7) to (26). This reminds us again of the ergodicity condition. Of course, the average value $\bar{t}=(t_1+t_2)/2$ can always be fixed, while t_1 and t_2 may take arbitrary values. Consequently, (28) can be interpreted as an adjustment in such a way that the ensemble average at a definite time $\bar{t}=(t_1+t_2)/2$ equals its timeaverage.

By the way, this includes also the case of "destructive interference." Of course, since the displacement operator $D(\gamma(t))$ represents $A(t)$ in the coherent-state representation, the replacement of

$$D(0)=[D(\gamma)\,D(-\gamma)]^{1/2}$$

by its arithmetic mean $\frac{1}{2}[D(\gamma)\,D(-\gamma)]$ provides that the vacuum state will be obtained if the field amplitudes γ and $-\gamma$ are superimposed within the coherence volume. This perfect coherence indicates again that the hyperbolic relaxation is sufficient for a fully coherent ergodic field.

4. PHOTOCOUNT STATISTICS (PCS)

The following proof of hyperbolic relaxation for a fully coherent ergodic field is based on the well-known fact that a fully coherent stationary field displays always a Poissonian distribution of the probability $p(n, \Delta t)$ of registering n photons in an arbitrarily small, but fixed time interval Δt. A chaotic field, on the other hand, follows a geometrical distribution of $p(n, \Delta t)$. If $p(n, \Delta t)$ remains Poissonian at any instant t, where the probability distribution shall generally account for an ensemble of identical samples, it is evident that this is then sufficient for a fully coherent ergodic field, even if it is not stationary.

Following Loudon (1983), we introduce the probability $p(t)\,dt$ of registering a particle of the decay product (for instance, a photon), originating from a relaxing field, within a small time interval between t and $t+dt$. The measurement of the number of particles $\langle \dot{n}T \rangle$ is always performed within the fixed time interval t and $t+T$, where $T > dt$ (see Fig. 1).

According to Loudon, we have generally the relation

$$P_0(t, T) = \exp\left[-\int_t^{t+T} p(t')\,dt'\right] \tag{29}$$

where $P_0(t, T)$ is the probability of registering no photon in the interval t and $t+T$. At any instant t, P_0 is normalized: $P_0(t, 0) = 1$. The probability of measuring no photon between t' and $t'+dt'$ is then [see Loudon (1983) or expand the r.h.s. of (29)]

$$P_0(t', dt') = 1 - p(t')\,dt' \tag{30}$$

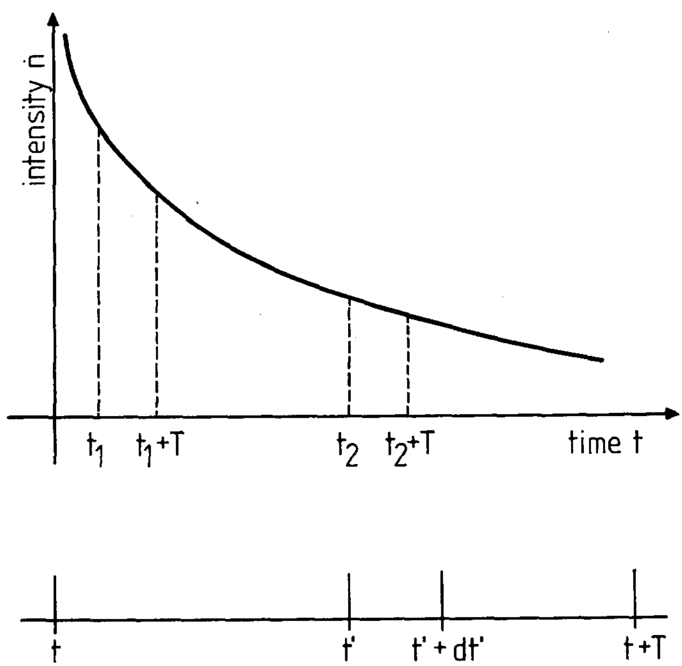

Fig. 1. A typical relaxation of an excited field, where the intensity $\dot{n}(t)$ decays. The number of particles of the decay product $(\dot{n}T)$ is always registered between time intervals t_i and t_i+T, $i=1, 2, \ldots$. These intervals can be divided into smaller ones t'_j and t'_j+dt', $j=1, 2, \ldots, N$, where either no particle or only one can be counted.

An ergodic coherent field displays (in contrast to chaotic fields) no bunching. This means that the probability of registering no photon within t and $t+T$ is, according to the probability theory of independent processes, the product of all the N single probabilities $[1-p(t')\,dt']$ of measuring no photon in the consecutive small time intervals dt' between t and $t+T$, where $N\,dt'=T$ (see Fig. 1):

$$P_0(t, T) = [1-p(t_1')\,dt'][1-p(t_2')\,dt'] \cdots [1-p(t_N')\,dt'] \tag{31}$$

Since the $[1-p(t_i')\,dt']$ for $i=1, 2, ..., N$ are positive real numbers, we then have

$$1-p(t_m')\,dt' \leqslant \left\{\prod_i [1-p(t_i')\,dt']\right\}^{1/N} \leqslant 1-\frac{1}{N}\sum_i p(t_i')\,dt' \tag{32}$$

where $p(t_m')$ represents the maximum of $p(t')$ within t and $t+T$. (32) reflects the fact that the geometrical mean is always lower than or at most equal to the arithmetic mean. According to the mean value theorem, we then can rewrite (31), taking account of (32), as

$$P_0(t, T) = \left[1-p(t+\mu T)\frac{T}{N}\right]^N \tag{33}$$

where $dt=T/N$, and μ with $0\leqslant\mu\leqslant 1$ is a time-independent quantity, since within the interval T the measurements do not allow the time resolution of $\dot{n}(t)$.

Consequently, from equations (29) and (33), we get

$$\left[1-p(t+\mu T)\frac{T}{N}\right]^N = \exp\left[-\int_t^{t+T} p(t')\,dt'\right] \tag{34}$$

The nonstationary solution for $\mu=1$ takes the form

$$p(t) = \frac{N}{t+t_0} \tag{35}$$

where t_0 is a constant. The special choice of $\mu=1$ cannot have any impact on the physical significance, since T is optional. (35) represents again a hyperbolic law.

We may conclude as follows: If a relaxing field keeps its Poissonian PCS-distribution at any instant, it decays according to a hyperbolic law. If, on the other hand, an ergodic field relaxes according to a hyperbolic function, one has to conclude that the field is a fully coherent one.

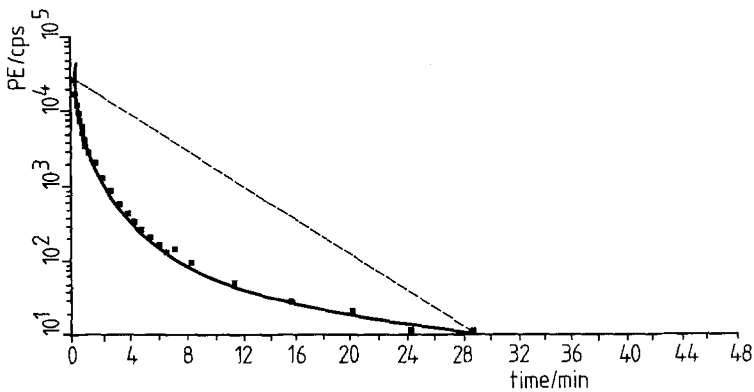

Fig. 2. "Delayed luminescense" of a *Bryophyllum daigremontanum* leaf at a wavelength of 676 ± 10 nm (Popp *et al.*, 1981). The measured count rate after exposure to a red-light illumination can be approximated by a hyperbolic decay law according to equation (20) (lower curve), while an exponential decay (dashed line), or even a superposition of three exponential functions with different decay constants cannot be adjusted.

This hyperbolic relaxation seems to be a rather common phenomenon in biological systems, e.g., the white-light-induced reemission of photons from cells (Scholz *et al.*, 1988) and living tissues (Popp *et al.*, 1981).

Figure 2 demonstrates this for the spectral reemission of a leaf of the plant *Bryophyllum daigremontanum* after illumination with red light of a wavelength of 676 ± 10 nm. The exponential function (dashed line) evidently cannot account for this decay, but the result clearly indicates coherent rescattering of light within living systems. This has also been indicated by "light-piping in plant tissues" (Mandoli and Briggs, 1982), which has been traced back to a high degree of coherence (Smith, 1982).

Hyperbolic relaxation may thus become a powerful tool for analyzing the living state in terms of coherence (Popp, 1986). This has been confirmed also by use of canonical coherent states in order to describe the long-range forces between human blood cells (Paul, 1983) and the long-range phase coherence in the bacteriorhodopsin macromolecules (Dunne *et al.*, 1983).

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