Exponential decay, recurrences, and quantum-mechanical spreading in a quasicontinuum model

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We consider a single quantum state coupled equally to each of a set of evenly spaced quasicontinuum (QC) states. We obtain a delay differential equation for the initial-state probability amplitude, and this equation is solved analytically. When the QC-level spacing goes to zero, the initial-state probability decays exactly exponentially. For finite QC-level spacings, however, there are recurrences of initial-state probability. We discuss Tolman's "quantum-mechanical spreading" of probability and also a classical analog of our model.

I. INTRODUCTION

One of the best-known general problems in physics is the decay of a single excited quantum state into a background of states. Examples may be found in spontaneous emission from an atom, decay of a radioactive nucleus, radiationless transitions in polyatomic molecules, autoionization, and, more generally, the problem of time asymmetry in physics.[1]

The problem of interest to us in this paper involves a discrete background or "quasicontinuum" (QC) of states. These background states are all coupled to a single, initially excited quantum state, but not directly to each other. Whereas the details of the distributions of coupling strengths and energy levels play a role in the dynamics of this system, some interesting general features may be obtained from specific models. In particular, we will consider the QC states to be equally spaced in energy and to have the same coupling to the initially excited state (Fig. 1).

Various researchers have found this model useful in their fields of interest. Davies,[1] for example, uses it to discuss the physics of time asymmetry. Bixon and Jortner[2] applied it to intramolecular radiationless transitions, while Stey and Gibberd[3] used it and other solvable models to discuss the decay of the initial state in the limit of a background continuum of states. Similar work was reported by Lefebvre and Savolainen.[4] More recently, Eberly *et al.*[5,6] have applied a somewhat more general model to a study of laser excitation of a molecular quasicontinuum. On the basis of numerical computations, they emphasized that the system has a characteristic "recurrence time" that is directly proportional to the QC density of states.

This problem in elementary quantum mechanics has a certain richness that deserves a simple and general treatment outside the specialized contexts in which it has appeared. It is our purpose here to give such a treatment, which we feel to be of considerable pedagogical value. The model illustrated in Fig. 1, while exactly solvable, is far from trivial. It can be used not only as a paradigm for the problem of dissipation and exponential decay in quantum mechanics, but also to elucidate Fermi's Golden Rule and the general phenomenon of "quantum-mechanical spreading."[7] We discuss these and other aspects of the model that are not evident in the cited research literature.[1-6]

II. THE SCHRÖDINGER EQUATION AND A DELAY EQUATION

The time-dependent Schrödinger equation for the QC model of Fig. 1 may be written in terms of the state amplitudes as follows:

$$\dot{a}(t)=-i\beta\sum_{n=-\infty}^{\infty}b_n(t)\ , \tag{2.1a}$$

$$\dot{b}_n(t)=-i(\Delta_0+n\rho^{-1})b_n(t)-i\beta a(t)\ , \tag{2.1b}$$

where a and b_n are the amplitudes for the initially excited state $[a(0)=1]$ and the background states, respectively.

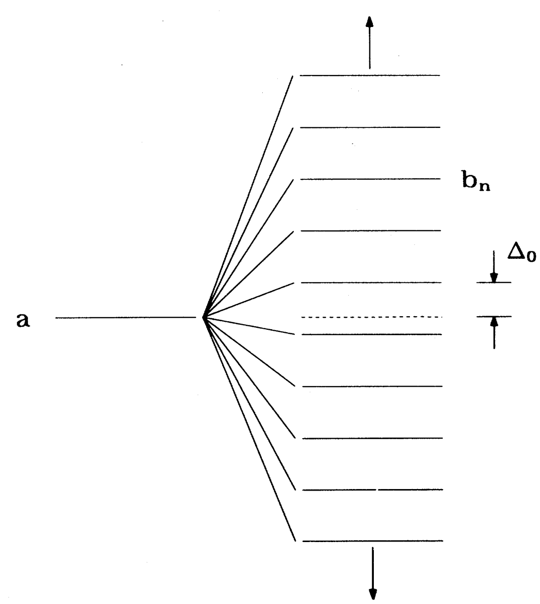

FIG. 1. Diagram of model shows coupling of initially excited state $|0\rangle$ with infinite number of equally spaced background states $|n\rangle$ with spacing ρ^{-1}.

The parameters β and Δ_0 are, respectively, the coupling strength and energy-level detuning of the initial state from the closest background state above the initial state (Fig. 1). ρ, the QC density of states, is the inverse of the QC-level spacing.

By formally integrating (2.1b) and using the solution for $b_n(t)$ in (2.1a), we obtain for $a(t)$ the integro-differential equation

$$\dot{a}(t) = -\beta^2 \sum_{n=-\infty}^{\infty} \int_0^t dt' e^{-i(\Delta_0 + n\rho^{-1})(t-t')} a(t')$$

$$= -\beta^2 \int_0^t dt' e^{-i\Delta_0(t-t')} a(t') \sum_{n=-\infty}^{\infty} e^{-in\rho^{-1}(t-t')} . \tag{2.2}$$

Now we invoke the Poisson summation formula[8]

$$\sum_{n=-\infty}^{\infty} e^{-2\pi i n x} = \sum_{n=-\infty}^{\infty} \delta(x-n) . \tag{2.3}$$

Thus we may write (2.2) in the form

$$\dot{a}(t) = -2\pi\beta^2\rho \sum_{n=-\infty}^{\infty} \int_0^t dt' e^{-i\Delta_0(t-t')} a(t') \times \delta(t-t'-2\pi n\rho) \tag{2.4}$$

or

$$\dot{a}(t) = -\frac{\gamma}{2} a(t) - \gamma \sum_{n=1}^{\infty} e^{-in\Delta_0\tau} a(t-n\tau)\Theta(t-n\tau) ,$$

where Θ is the unit step function,

$$\gamma = 2\pi\beta^2\rho \tag{2.5}$$

and

$$\tau = 2\pi\rho . \tag{2.6}$$

Equation (2.4) has the form of a delay differential equation.[9] When the QC-level spacing is taken to be zero, so that τ becomes very large, we have

$$\dot{a}(t) = -\frac{\gamma}{2} a(t) .$$

Therefore, the initial-state probability in this limit decays *exactly* exponentially:

$$|a(t)|^2 = e^{-\gamma t} .$$

The decay rate γ is precisely that given by Fermi's Golden Rule.

Delay differential equations have many interesting applications.[9] A typical application is to problems involving retarded interactions, in which case the origin of the delay time τ is physically clear.[10] In the present problem the delay equation arises from the additive phasing effect of the evenly spaced background states. Mathematically, this finds its expression in the Poisson sum rule (2.3). A similar equation arises in the *classical* problem of an oscillator coupled to a background of oscillators with evenly spaced frequencies. This classical analog is discussed in Sec. VIII.

III. SOLUTION OF THE DELAY EQUATION

The delay differential equation (2.4) with the initial condition $a(0)=1$ may be solved in a straightforward manner using the Laplace transform. We will instead give a more elegant solution by first assuming a solution of the form

$$a(t) = \sum_{n=0}^{\infty} e^{-\gamma t_n/2} e^{-in\Delta_0\tau} C_n(t_n)\Theta(t_n) , \tag{3.1}$$

where $t_n = t - n\tau$. Using this form in (2.4), we obtain an equation for the coefficients C_n:

$$\dot{C}_n(t) = -\gamma \sum_{m=0}^{n-1} C_m(t) \tag{3.2}$$

with $C_0(t)=1$ and $C_n(0)=0$ for $n \geq 1$. Now the Laguerre polynomials satisfy the relation

$$\dot{L}_n(t) = -\sum_{m=0}^{n-1} L_m(t), \quad L_n(0)=1 \tag{3.3}$$

for $n \geq 1$ and $L_0(t)=1$. The functions

$$\mathscr{L}_n(t) = \begin{cases} L_0(t), & n=0 \\ L_n(t) - L_{n-1}(t), & n \geq 1 \end{cases} \tag{3.4}$$

therefore satisfy the relation

$$\dot{\mathscr{L}}_n(t) = -\sum_{m=0}^{n-1} \mathscr{L}_m(t) \tag{3.5}$$

with $\mathscr{L}_0(t)=1$ and $\mathscr{L}_n(0)=0$ for $n \geq 1$. It follows from the uniqueness theorem for differential equations that

$$C_n(t) = \mathscr{L}_n(\gamma t) \tag{3.6}$$

so that the solution of Eq. (2.4) is

$$a(t) = \sum_{n=0}^{\infty} e^{-in\Delta_0\tau} e^{-\gamma(t-n\tau)/2} \times \mathscr{L}_n(\gamma(t-n\tau))\Theta(t-n\tau) . \tag{3.7}$$

Solutions equivalent to this have been reported by Stey and Gibberd[3] and Lefebvre and Savolainen.[4]

IV. INITIAL-STATE PROBABILITY

Writing out the first few terms of (3.7), we have

$$a(t) = e^{-\gamma t/2} - \gamma(t-\tau)e^{-\gamma(t-\tau)/2}\Theta(t-\tau) + [\tfrac{1}{2}\gamma^2(t-2\tau)^2 - \gamma(t-2\tau)] \times e^{-\gamma(t-2\tau)/2}\Theta(t-2\tau) - \cdots , \tag{4.1}$$

where for simplicity we have assumed $\Delta_0=0$, i.e., one of the QC levels is exactly resonant with $|0\rangle$. At each integral multiple of τ, a new contribution arises. The variation of $a(t)$ with t can therefore be quite complicated, especially when contributions from earlier "intervals" $(n-1)\tau \leq t \leq \tau$ overlap appreciably. Stey and Gibberd[3] have aptly described this variation as "bizarre." We will therefore show a few graphs of $|a(t)|^2$ for various values of γ and τ. In each case we have evaluated (3.7) up to $t=5\tau$.

Figure 2 shows the initial-state probability for $\gamma=2$ and $\tau=6$. The exponential decay in the first interval $(0 \leq t \leq 6)$ is followed by a recurrence of probability beginning at $t=\tau$. The variation in successive intervals of time τ is more complicated owing to the polynomials multiplying the factors $\exp[-\gamma(t-n\tau)/2]$ in the expression for $a(t)$.

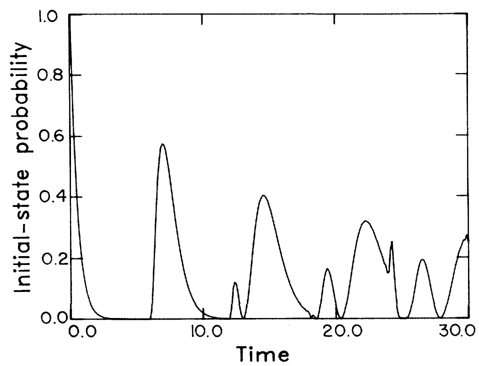

FIG. 2. Initial-state probability vs time with $\gamma=2$, $\tau=6$, and $\Delta_0=0$.

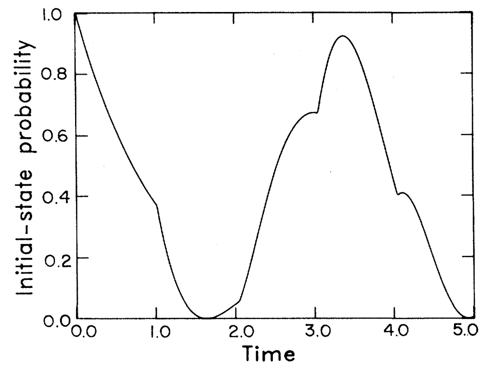

FIG. 4. Same as Fig. 2, but with $\gamma=\tau=1$.

Figure 3 is for $\gamma=\tau=2$. Because the basic "recurrence" time is now shorter than in Fig. 2, there is more of a contribution in a given interval from previous intervals, and this is reflected in the fact that the probability comes up higher for $t>\tau$ than it does in Fig. 2. Figure 4 is for $\gamma=\tau=1$, where this overlap effect is more pronounced.

Figures 2–4 are obtained assuming $\Delta_0=0$. In Figs. 5 and 6 we show $|a(t)|^2$ for two cases in which $\exp(-i\Delta_0\tau)=-1$. Destructive interference in the contribution to $a(t)$ from previous time intervals is now incomplete at all times, unlike the case in which $\Delta_0=0$.

V. CONSERVATION LAWS

In general the variation of $a(t)$ with t is complicated. However, some simplification is possible when $\gamma\tau \gg 1$. In this case the "lifetime" γ^{-1} associated with the exponential decay factor on each time interval is short compared with the duration τ of each interval, and so the contributions from different intervals do not appreciably overlap. From (3.7) we can write

$$a^{(n)}(t) \cong e^{-\gamma t_n} e^{-in\Delta_0\tau} \mathscr{L}_n(\gamma t_n) \tag{5.1}$$

in this case, where $a^{(n)}(t)$ denotes $a(t)$ for $n\tau \le t \le (n+1)\tau$, i.e., in the nth time interval.

For $\gamma\tau \gg 1$ there are two area-conservation theorems that follow from (5.1) and the properties of the Laguerre polynomials. These are[6]

$$\left| \int_{n\tau}^{(n+1)\tau} dt\, a^{(n)}(t) \right| \cong \left| \int_{n\tau}^{\infty} dt\, a^{(n)}(t) \right| = \begin{cases} 2/\gamma, & n=0 \\ 4/\gamma, & n \ge 1 \end{cases} \tag{5.2}$$

and

$$\int_{n\tau}^{(n+1)\tau} dt\, |a^{(n)}(t)|^2 \cong \int_{n\tau}^{\infty} dt\, |a^{(n)}(t)|^2 = \begin{cases} 1/\gamma, & n=0 \\ 2/\gamma, & n \ge 1\,. \end{cases} \tag{5.3}$$

In both cases the value of the integral on the first interval is half its value on all subsequent intervals. Equation (5.3) says that the integrated probability for the initial state in each interval $n \ge 1$ is conserved, and (5.2) is a similar result for the integrated probability amplitude.

VI. FINITE NUMBER OF BACKGROUND STATES

Our analytical solution for the initial-state probability amplitude assumes an infinite number of QC states. What

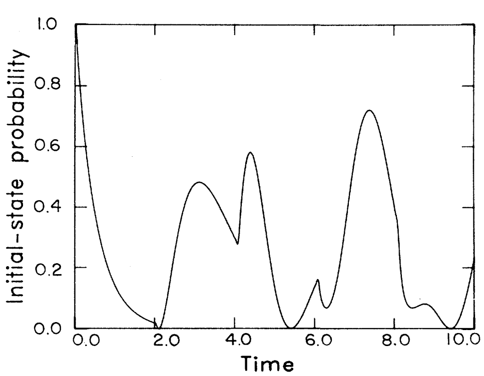

FIG. 3. Same as Fig. 2, but with $\tau=2$.

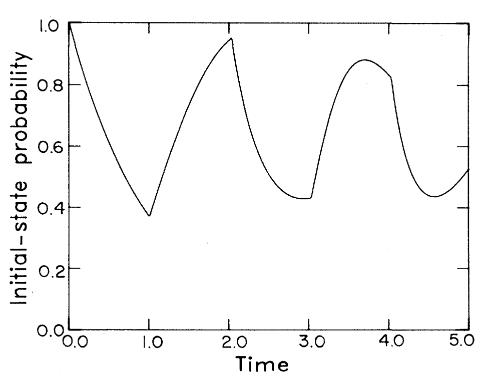

FIG. 5. Same as Fig. 2, but with $\gamma=\tau=1$ and $\Delta_0\tau=\pi$.

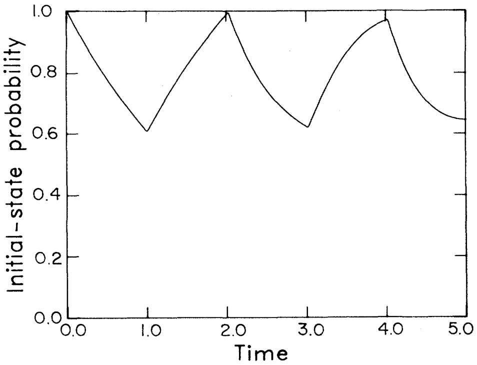

FIG. 6. Same as Fig. 5, but with $\gamma=\frac{1}{2}$.

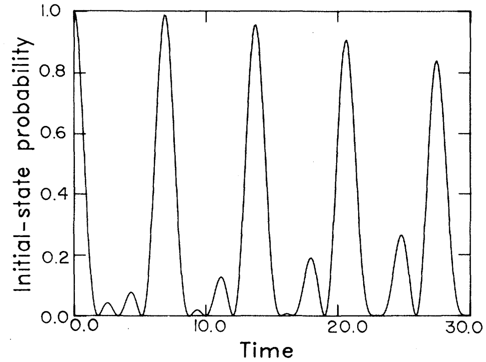

FIG. 8. Same as Fig. 2, but with $N=3$.

happens if we have a finite number of equally spaced background states, as in Fig. 7?

For the case shown in Fig. 7, the infinite summation in the integrand of Eq. (2.2) is replaced by a finite summation, in which case the integration is not so easily performed. Figures 8—10 show the results of numerical integration of (2.1) for the case $\gamma=2$, $\tau=6$, and for $N=3$, 9, and 15, respectively. These should be compared with Fig. 2, the result for the case of an infinite number of background states. As N increases, $|a(t)|^2$ approaches the analytical result shown in Fig. 2 for an infinite number of background states. The case $N=15$, in Fig. 10, is already in excellent agreement with Fig. 2. The reason for this is discussed in the Appendix.

VII. QUANTUM-MECHANICAL SPREADING

In his treatise on statistical mechanics, Tolman[7] identified a distinctly quantum-mechanical source of the decrease with time of the H function. This quantum-mechanical contribution stems from the Klein relation

$$\sum_n \rho_{nn}(0)\ln\rho_{nn}(0) \geq \sum_n \rho_{nn}(t)\ln\rho_{nn}(t) \,, \tag{7.1}$$

where ρ is the density matrix and is assumed to be diagonal at $t=0$. This "quantum-mechanical spreading"[7] is the spreading of probability over the possible states of the system. One way to appreciate the purely quantum-mechanical nature of this spreading of (fine-grained) probability is to note that

$$\sum_n \rho_{nn}\ln\rho_{nn}$$

is not a trace of any quantum-mechanical operator.[11]

In our problem the density matrix is simply

$$\rho_{mn}(t)=c_m^*(t)c_n(t) \,, \tag{7.2}$$

where $c_1(t)=a(t)$ and $c_m(t)=b_{m-1}(t)$ otherwise. The system starts out in a pure state and therefore remains in a pure state:

$$\mathrm{Tr}\,\rho(t)=\mathrm{Tr}\,\rho^2(t)=1 \,. \tag{7.3}$$

It follows furthermore that the entropy

$$S=-k\,\mathrm{Tr}\,(\rho\ln\rho) \tag{7.4}$$

is zero throughout the evolution of our system.

We have computed the function

$$F(t)=\sum_n \rho_{nn}(t)\ln\rho_{nn}(t)=\sum_n |c_n(t)|^2\ln|c_n(t)|^2 \tag{7.5}$$

for our system in order to check (7.1) and to elucidate the nature of the quantum-mechanical spreading of probability. Figure 11 shows $F(t)$ for the parameters used in obtaining Fig. 2. Clearly the Klein relation $F(0)\geq F(t)$ is satisfied. Note, however, that if $t_2>t_1$ it does not follow that $F(t_1)\geq F(t_2)$, for off-diagonal terms appear in the density matrix as the system evolves in time, whereas the

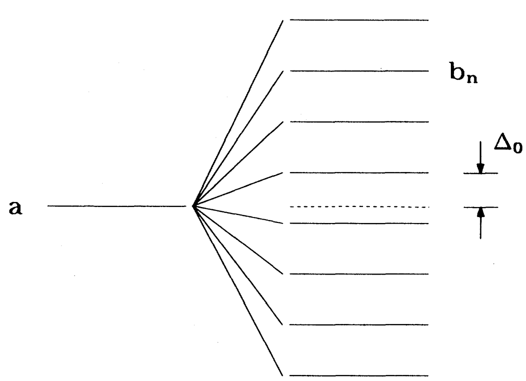

FIG. 7. Same as Fig. 1, but with only N background states.

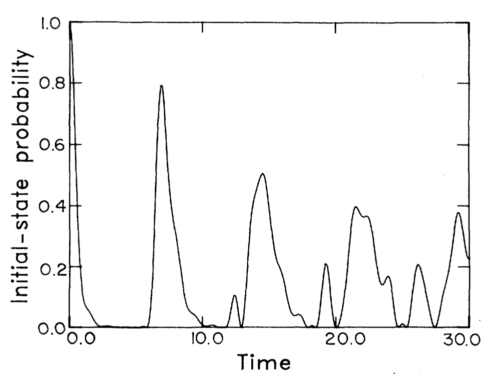

FIG. 9. Same as Fig. 8, but with $N=9$.

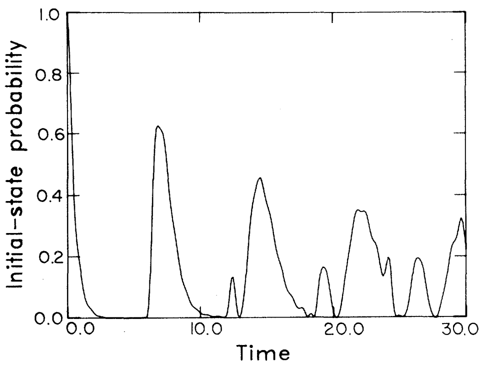

FIG. 10. Same as Fig. 8, but with $N=15$.

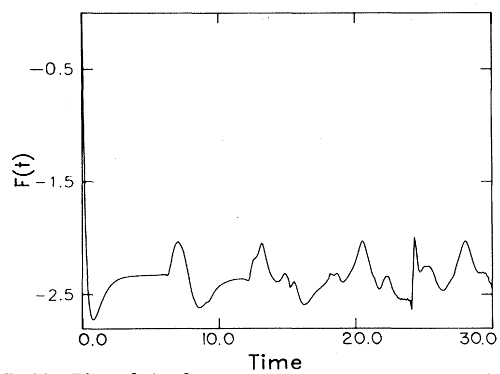

FIG. 11. Plot of the function $F(t)$ vs time using the parameters of Fig. 2. Note: $F(0)\geq F(t)$.

derivation of the Klein relation assumes that there is no off-diagonal coherence.

If the system starts out at $t=0$ with some nonvanishing off-diagonal density matrix elements, the Klein relation is invalid as a general statement. It fails, for instance, if we assume initial conditions corresponding to the state of the system at $t=10$ in Fig. 11.

The fact that the entropy (7.4) of a system initially in a pure state remains zero has led in the past to an alternative definition of entropy, one that gives an entropy increase due to quantum-mechanical spreading even for a system initially in a pure state. In particular, Born[12] preferred the definition

$$\text{“}S\text{”}=-kF(t)\ . \tag{7.6}$$

Jaynes[13] has noted that this definition does not give the same entropy for all pure states, whereas von Neumann demonstrated that any pure state may be reversibly and adiabatically transformed into any other pure state of the system. Furthermore, $F(0)\geq F(t)$ in general only if there is no off-diagonal coherence at $t=0$.

In Fig. 12 we show the distribution of probabilities $|c_n|^2$ at different times for the same parameters as in Fig. 2. The spreading of probability is largely confined to the background states closest in energy to the initial state. That is, QC states that are far off resonance from the initial state $|0\rangle$ are not visited with high probability during the evolution of the system.

VIII. A CLASSICAL ANALOG

Consider a harmonic oscillator of (circular) frequency ω_0 coupled equally to a large set of "background" oscillators with frequencies ω_n. If the background oscillators are not coupled among themselves we have the equations of motion

$$\ddot{x}(t)+\omega_0^2 x(t)=-A\sum_n q_n(t)\ , \tag{8.1a}$$

$$\ddot{q}_n(t)+\omega_n^2 q_n(t)=-Ax(t)\ , \tag{8.1b}$$

where A is the coupling constant. Write

$$x(t)=\text{Re}[a(t)e^{-i\omega_0 t}]\ ,$$
$$q_n(t)=\text{Re}[b_n(t)e^{-i\omega_0 t}] \tag{8.2}$$

and assume that

$$|\ddot{a}|\ll\omega_0^2|a|\ ,\quad |\ddot{b}(t)|\ll\omega_0^2|b(t)|\ , \tag{8.3}$$

an assumption that can be checked once a solution has been obtained. Then we may replace (8.1) by

$$\dot{a}(t)=-i\beta\sum_n b_n(t)\ , \tag{8.4a}$$

$$\dot{b}_n(t)=-i(\omega_n-\omega_0)b_n(t)-i\beta a(t)\ , \tag{8.4b}$$

where $\beta=A/2\omega_0$.

Suppose that

$$\omega_n=\omega_0+\Delta_0+n\rho^{-1},\quad n=0,\pm1,\pm2,\ldots,\pm N \tag{8.5}$$

with

$$1\ll N<\rho(\omega_0+\Delta_0)\ . \tag{8.6}$$

This ensures that the oscillator frequencies $\omega_n>0$, while the number of background oscillators is large enough so that we can take $N\to\infty$ as an approximation in the solution of (8.4) for $a(t)$ (see the Appendix). Then $a(t)$ is given approximately by (3.7), and we have a classical analog of our quantum-mechanical QC model.

IX. DISCUSSION

An important feature of the QC model we have discussed is that it is exactly solvable for the initial-state probability. In the limit of a continuum of QC states, this probability exhibits *exact* exponential decay.

The problem of the decay of an unstable state in quantum mechanics has a large literature.[14] A necessary condition for pure exponential decay is that the energy spectrum have no lower bound; this condition may be inferred from the Paley-Wiener theorem.[14] Thus in realistic physical models exponential decay is only an approximation. Fonda and Ghirardi[14] have associated the deviations from exponential decay with the regeneration of the initial state from the "background" states. Robiscoe and Hermanson[14] have considered a model of "exponential decay with a memory," which gives pure exponential decay in the limit in which the memory time goes to zero. A similar model was discussed by Pietenpol,[14] who obtains exponential decay in the limit of weak coupling to the background continuum, corresponding to the case of a short memory time in the model of Robiscoe and Hermanson.

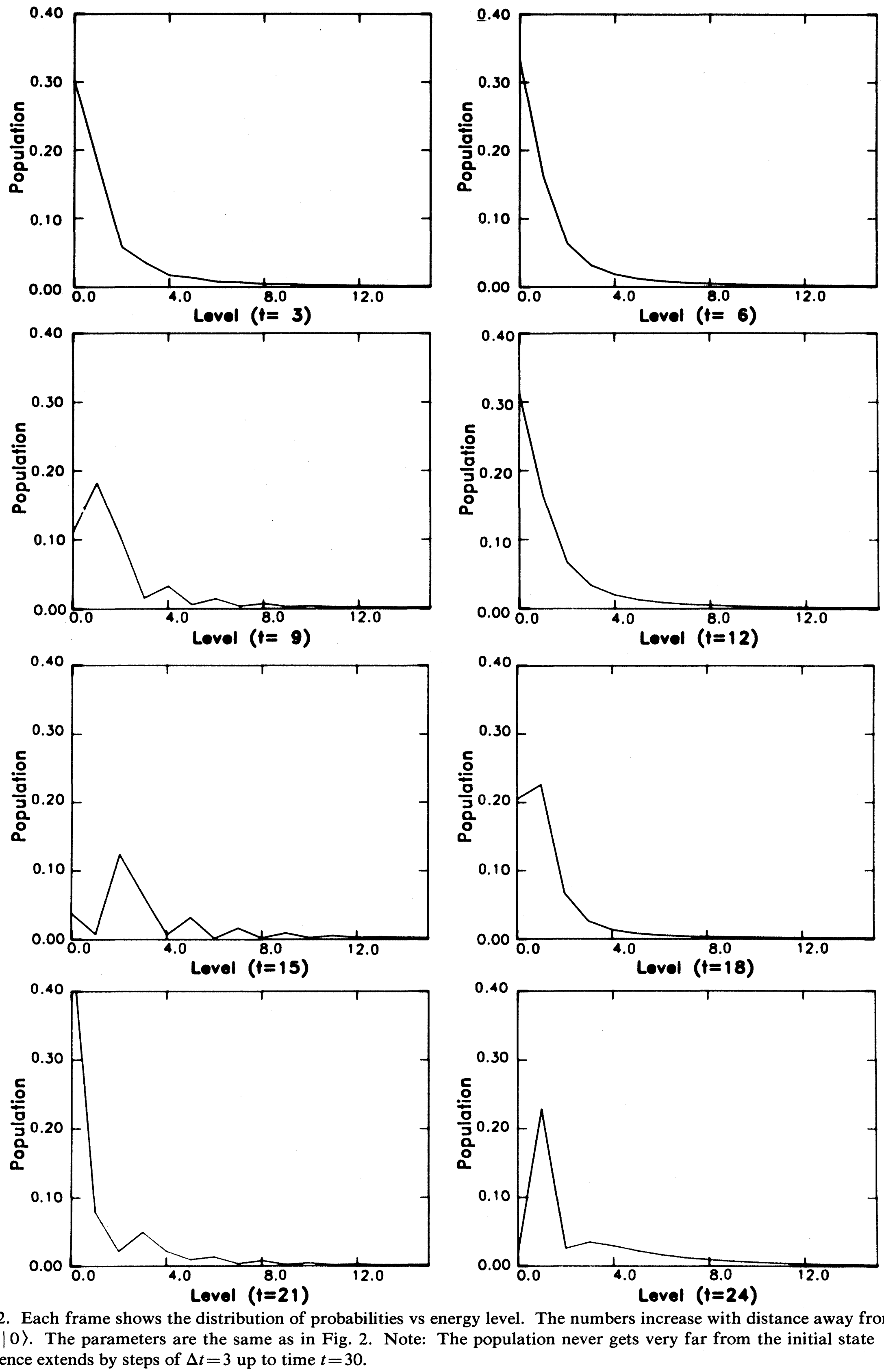

FIG. 12. Each frame shows the distribution of probabilities vs energy level. The numbers increase with distance away from the initial level $|0\rangle$. The parameters are the same as in Fig. 2. Note: The population never gets very far from the initial state $|0\rangle$. The time sequence extends by steps of $\Delta t = 3$ up to time $t = 30$.

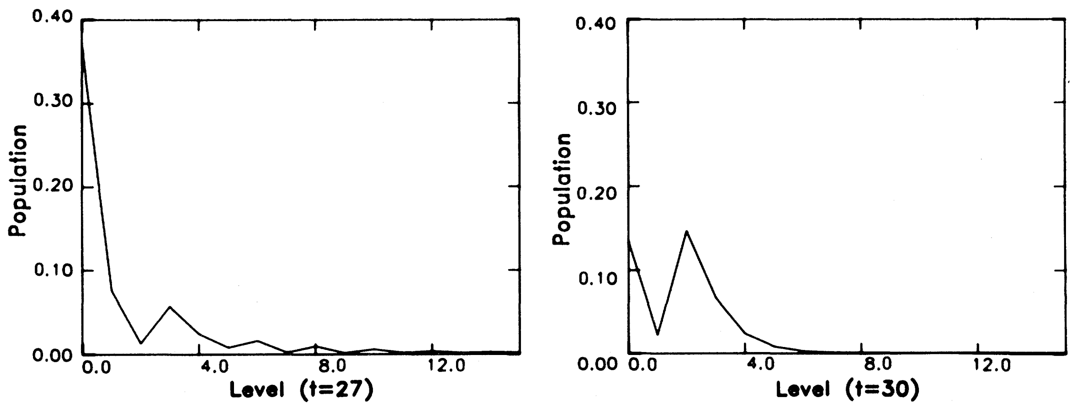

FIG. 12. *(Continued).*

The QC model we have considered gives exact exponential decay in the continuum limit. If we retain only a finite number of QC levels, the energy spectrum has upper and lower bounds, and the Paley-Wiener theorem precludes exponential decay.[15] However, exponential decay is an excellent approximation, even for a relatively small number of QC levels, when the density of background states is large.

Exponential decay of an unstable state is, of course, very often an excellent approximation. Nevertheless, there are situations in which nonexponential decay may be inferred from the observation of a non-Lorentzian line shape. Robiscoe and Hermanson[14] have suggested that non-Lorentzian line shapes might be associated with finite memory times. We are currently investigating the QC model from this point of view.[16]

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We wish to thank Professor J. H. Eberly for informing us that our analytical results were obtained independently

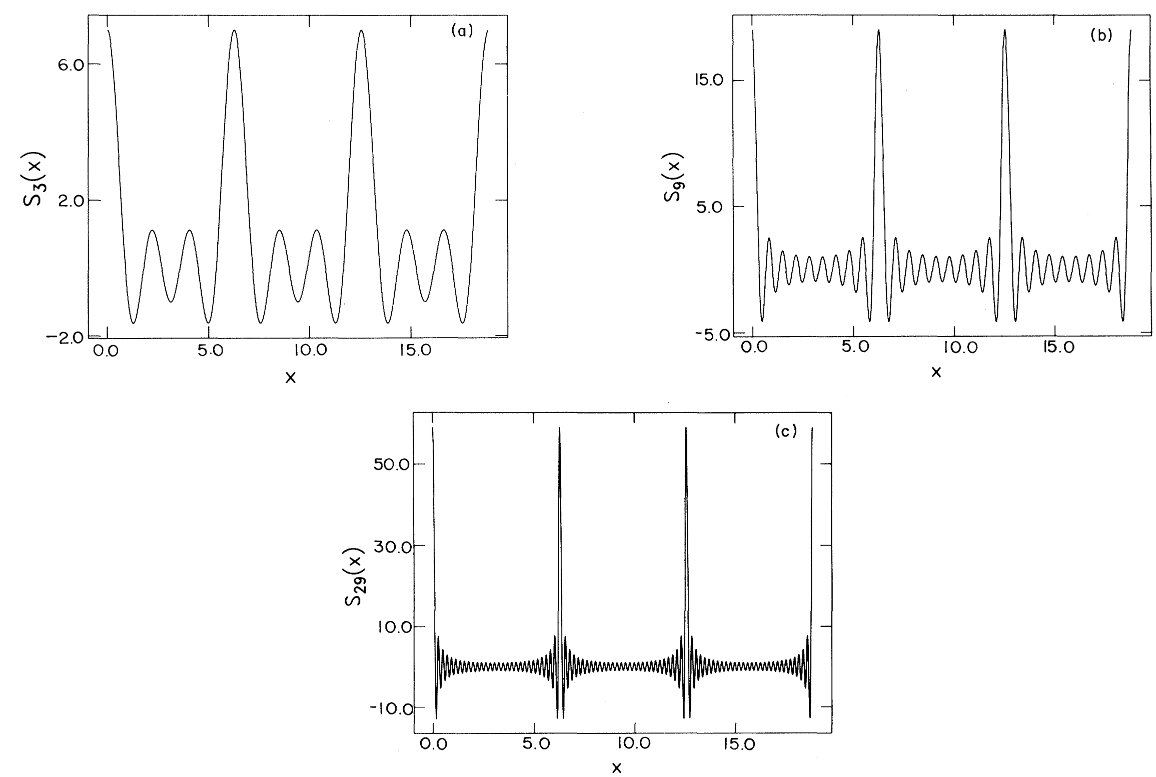

FIG. 13. Three graphs showing the function $S_n(x)$ vs x, (a) $N=3$, (b) $N=9$, (c) $N=29$. For $N\rightarrow\infty$ we would obtain a sequence of delta functions at the locations indicated in (a), (b), or (c).

in Refs. 3 and 4. We also thank Professor O. H. Zinke for encouraging us to consider the aspects of the problem described in Sec. VII. Dr. R. T. Pack originally suggested that our analytical results could be cast into a form involving Laguerre polynomials. Research at the University of Arkansas was supported in part by the National Science Foundation under Grant No. EPSOR ISP-80-11447 and the state of Arkansas.

APPENDIX

If we assume a finite number N of background states as in Fig. 7, then

$$\sum_{n=-\infty}^{\infty} e^{-in\rho^{-1}(t-t')} \to \sum_{n=-N}^{N} e^{-in\rho^{-1}(t-t')}$$

in Eq. (2.2). The sum

$$S_N(x) \equiv \sum_{n=-N}^{N} e^{-inx} = \cos(N_x) + \frac{\sin(N_x)\cos(x/2)}{\sin(x/2)} \tag{A1}$$

has maxima at $x=2\pi n$, $n=0,\pm1,\pm2,\ldots$. In Fig. 13 we plot $S_N(x)$ versus x for $0 \le x \le 20$. As N increases we approach the limit

$$\lim_{N\to\infty} \sum_{n=-N}^{N} e^{-inx} = \sum_{n=-\infty}^{\infty} \delta\left[\frac{x}{2\pi} - n\right], \tag{A2}$$

i.e., the Poisson sum rule. Even for N as small as 3 or 9 we can see from Fig. 13 that values of $a(t')$ with $t' \cong t - 2\pi\rho n = t - n\tau$ are weighted strongly in the integrand of (2.2). For $N=29$ we are in fact very close to the limit (A2), i.e., the limit of an infinite number of background states.

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[14]We list here only some representative papers with fairly extensive references: J. L. Pietenpol, Phys. Rev. 162, 1301 (1967); L. Fonda and G. C. Ghirardi, Nuovo Cimento 7A, 180 (1972); 10A, 850 (1972); R. T. Robiscoe and J. C. Hermanson, Am. J. Phys. 40, 1443 (1972); 41, 414 (1973). Exponential decay of an atomic state due to spontaneous emission was obtained approximately by L. D. Landau [Z. Phys. 45, 430 (1927)] in a remarkable paper that seems to have been forgotten. Credit for this approximation usually goes to V. F. Weisskopf and E. Wigner [Z. Phys. 63, 54 (1930)], who used the approximation of exponential decay as an ansatz. Deviations from exponential decay in spontaneous emission are discussed, for instance, by P. L. Knight and P. W. Milonni, Phys. Lett. 56A, 275 (1976). These deviations appear to be too small to have been observed in atomic spontaneous emission.

[15]The energy spectrum of the coupled system of Fig. 1 has been discussed by several authors. See, for example, Ref. 3.

[16]The earliest reference to a model of this type, equally spaced background with equal coupling strengths, which we have found, is the work of Weisskopf and Wigner, see Ref. 14.