# Interaction between a Nonlinear Oscillator and a Radiation Field

I. R. Senitzky\*

U. S. Army Electronics Technology and Devices Laboratory, Fort Monmouth, New Jersey 07703 (Received 13 March 1972)

The behavior of a nonlinear oscillator (NO) coupled to a radiation field is investigated. The NO considered is an angular momentum oscillator, of energy  $\hbar\omega L_3$ , that describes the collective effect of a number of identical two-level (or spin- $\frac{1}{2}$ ) systems under given idealized conditions, a large total angular momentum quantum number  $L_0$  corresponding to a large number of two-level systems. The field is described by a set of modes. Free decay-with the NO initially excited and the radiation field in the ground state-and forced oscillation-with the NO subject to a prescribed resonant driving field—are studied. The analysis is performed both classically and quantum mechanically, using the classical and Heisenberg equations of motion, respectively, so as to display explicitly the difference in the results. Interest in the comparison between the two formalisms is motivated by the expectation that for large  $L_0$ , the NO should behave essentially classically, except near its highest-energy state in the absence of a driving field. The general equations of motion are reduced to equations for the NO variables only. In the classical analysis of free decay, expressions for the energy and oscillating coordinates are derived. The decay time is shown to approach infinity as  $L_3(0)$ approaches  $L_0$  (the limiting condition being that of unstable equilibrium) and the radiative frequency shift is shown to be approximately proportional to  $L_3(t)$ . It is also shown that use of the rotating-wave approximation alters qualitatively the expression for the frequency shift. In the classical analysis of forced oscillation, approximate results are obtained for a weak driving field and a strong driving field, the NO being initially in the ground state. The weakfield results exhibit a monotonic approach of  $L_3(t)$  to a constant (negative) value-or steady state—at which the power absorbed equals the power radiated; in the strong-field case,  $L_3(t)$ oscillates periodically between the limits  $\pm L_0$ , the coupling to the radiation field having negligible effect on the frequency of this oscillation. In the quantum-mechanical analysis, the equations of motion become simplified for  $L_0 = \frac{1}{2}$ , and this case is treated first. The free-decay results are essentially similar to those of the Weisskopf-Wigner theory, exhibiting an exponential decay of  $\langle L_3(t) \rangle$  and a radiative frequency shift in the oscillating coordinates. Under forced oscillation, with  $\langle L_3(0) \rangle = -L_0$ ,  $\langle L_3(t) \rangle$  approaches a constant value either monotonically, if the driving field is sufficiently weak, or by means of a damped oscillation, if the driving field is strong. For  $L_0 > \frac{1}{2}$ , the free decay is treated by a method that involves the derivation of a set of expressions for the kth derivative of  $\langle L_3(t) \rangle$  as an expectation value of a polynomial in  $L_3$  of order k+1. This set, together with the eigenvalue equation for  $L_3$ , is shown to lead to a solution for all the moments of  $L_3$ . The method is used to obtain complete solutions for several low values of  $L_0$ , and also to obtain initial derivatives of  $\langle L_3(t) \rangle$  as polynomials in  $L_0$  for  $\langle L_3(0) \rangle = L_0$ . For large  $L_0$ , a comparison of classical and quantum-mechanical equations shows that only the condition  $\langle L_3(0) \rangle \approx L_0$  requires quantum-mechanical treatment. and that for a short time only. The free-decay problem with initial condition  $\langle L_3(0) \rangle = L_0$  is solved quantum mechanically up to such a time  $t_1$  by means of the initial derivatives previously derived, and then this solution is used to provide initial conditions determining a classical solution for  $t \ge t_1$ . The statistical aspects introduced by the quantum mechanics are preserved in the classical solution, and their significance is discussed, with several examples; comparison is also made with a nonstatistical approximation. In the case of forced oscillation, the behavior of  $\langle L_3(t) \rangle$  -for arbitrary  $L_0$ -is examined in detail for a strong field, and turns out to be described by a damped oscillation, the existence of the damping being independent of  $L_0$ . The apparent inconsistency of this result with the expectation that a large system subject to strong forces should behave classically is discussed, and  $\langle L_3^2(t) \rangle$  is examined. It is concluded that in a single experiment, the NO energy oscillates approximately like the classical energy, without damping; quantum mechanics introduces, however, a slight randomness (or unpredictability) in the frequency of this oscillation, because of the coupling with the radiation field. Since  $\langle \mathcal{L}_3(t) \rangle$  describes the average over an ensemble of which each member consists of a NO coupled to a radiation field, the random frequency variation among the members accounts for the damping of the average. The usefulness of the combination of classical and quantum-mechanical analyses in achieving and interpreting theoretical results for a class of phenomena involving the collective interaction between a number of atoms and a radiation field is pointed out.

### I. INTRODUCTION

In a previous paper,<sup>1</sup> hereafter referred to as I, the resonant interaction between a linear and a nonlinear oscillator was analyzed in a nonperturbative manner. The type of nonlinear oscillator (NO) considered was that which can be described by an

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angular momentum system in an external field, the energy being proportional to  $l_3$  and the oscillating coordinates being proportional to  $l_1$  and  $l_2$ . As pointed out in I, such an oscillator can describe not only a spin system of arbitrary l, but also a two-level (electric-dipole) system or, under certain ideal conditions, an arbitrary number of identical two-level systems. The linear oscillator, on the other hand, describes a single field mode or a lumped constant circuit, so that the coupled system represents, in an idealized dissipationless manner, a number of identical atomic systems interacting with a single resonator. The main interest was related to the long-time behavior of the coupled system and the absence of a steady state for general initial conditions.

In the present paper, we consider the same NO coupled to an entire radiation field, that is, to an infinite number of modes, or linear oscillators, covering a range of frequencies. We thus have an idealized model of a number of atoms coupled to the entire electromagnetic field or acoustic field.<sup>2</sup> In view of the fact that the entire field has properties of a large reservoir to which the NO loses energy, we will allow for the presence of a prescribed resonant driving force acting on the NO so that both the driven and the freely decaying NO may be studied.

An important feature of the present paper, as in I, is the fact that the analysis will be carried out in both a classical and a guantum-mechanical manner, with the differences explicitly displayed. This procedure is motivated by the viewpoint that the radiation field interacting with a large number of atoms acting collectively may be considered in many respects to be a classical system. In I, some seemingly puzzling differences between the quantum-mechanical and classical descriptions were elucidated. In the present paper, a comparison between the two descriptions will likewise be enlightening. We will obtain further evidence for the idea, expounded previously,<sup>1</sup> that in phenomena where a large number of atoms behave collectively-such as those of quantum electronics, for instance-the quantum-mechanical expectation value does not necessarily give even a qualitative description of the result of a macroscopic experiment, and that a classical analysis is useful in the interpretation of some of the quantum-mechanical results.

Certain special aspects of the present problem are related to one treated in considerable detail some years ago, that of a two-level system (driven as well as freely decaying) coupled to a general dissipation mechanism.<sup>3</sup> The features not present in the earlier treatment consist of a specialization of the dissipation mechanism to a radiation field, the generalization of the two-level system to an

angular momentum oscillator with an arbitrary number of levels, and the parallel classical analysis. A very early analysis of the average free decay of a two-level system coupled to the electromagnetic field is found in the classic paper of Weisskopf and Wigner.<sup>4</sup> The average behavior of a large number of microscopic spin systems coupled to a dissipation mechanism was investigated by Wangsness and Bloch.<sup>5</sup> Spontaneous emission-in its usual sense as a perturbation-theory description of free decay-from a number of atoms behaving collectively was analyzed by Dicke.<sup>6</sup> He introduced the term "superradiance" and a formalism similar to the angular momentum formalism to describe cooperative spontaneous emission. The meaning of "superradiance" in terms of-and its relation to-oneatom states (recently referred to by some authors as "Bloch states") was explained subsequently.<sup>7</sup> The behavior of a driven two-level system and the free cooperative decay of a number of two-level systems have been discussed more recently by Dillard and Robl<sup>8</sup> while the free decay itself has been the subject of other recent studies, 9-16 in some of which<sup>14-16</sup> the model is different (the twolevel systems being coupled to a highly damped single mode rather than to the entire field) but the mathematical problem is essentially similar. There will be occasion later to compare the present work with-and comment on-some of the above references.

#### **II. EQUATIONS OF MOTION**

The notation to be used will be similar to that of I, except that the consideration of a large number of modes will require that the variables of each mode be labeled with an index. A denumerable set of modes will be considered, such as is obtained by the imposition of appropriate boundary conditions on the field in a finite volume. The coupling between the NO and the field will be assumed to be sufficiently weak so as not to alter the qualitative characteristics of either system, but aside from this requirement, the strength of the coupling constant between the NO and the kth mode and the density of modes per unit frequency interval will be left arbitrary in the present discussion for the following reasons: (i) These matters raise questions concerning frequency dependence. divergencies, and cutoff, which require separate discussion for electric, magnetic, and acoustic coupling; (ii) there exists disagreement in the literature with reference to some of these questions which merits critical examination<sup>17</sup>; (iii) the purpose of the present analysis is accomplished by incorporating expressions for coupling strengths and mode densities into constants that determine the decay rate and frequency shift of the NO.

The kth mode of (angular) frequency  $\omega_k$  is de-

scribed by the Hamiltonian

$$H_k = \frac{1}{2} \hbar \omega_k (q_k^2 + p_k^2) , \qquad (1)$$

where

$$[q_k, p_k]_P = i , \qquad (2)$$

the bracket  $[, ]_P$  standing for the commutator in the quantum-mechanical description and *i* times the Poisson bracket in the classical description. The NO, of natural frequency  $\omega$ , is described by the variables  $l_1$ ,  $l_2$ , and  $l_3$ , the Hamiltonian being given by

$$H_N = \hbar \omega l_3 , \qquad (3)$$

and the variables satisfying the relationship

$$[l_r, l_s] = il_t , \qquad (4)$$

with r, s, and t standing for the cyclic permutation of 1, 2, and 3. The (dipole-type) coupling between a mode and the NO is specified quite generally by the interaction Hamiltonian

$$H_{kN} = \hbar q_k (\gamma_{1k} l_1 + \gamma_{2k} l_2 + \gamma_{3k} l_3) , \qquad (5)$$

where the  $\gamma$ 's are the coupling constants between the *k*th mode and the NO. It is immaterial whether  $q_k$  or  $p_k$  is used in the interaction Hamiltonian, and the coupling can be changed from "q" type to "p" type by the transformation  $q_k + p_k$ ,  $p_k + -q_k$ . A detailed discussion of the coupling constants and certain assumptions inherent in combining a number of systems into a single NO, as well as comments on the model used in Refs. 14–16, is given in Appendix A.

As in I, it is convenient to use the familiar non-Hermitian operators

$$a_{k} = 2^{-1/2} (q_{k} + ip_{k}) , \quad a_{k}^{\dagger} = 2^{-1/2} (q_{k} - ip_{k}) ,$$
  
$$l_{+} = 2^{-1/2} (l_{1} + il_{2}) , \quad l_{-} = 2^{-1/2} (l_{1} - il_{2}) , \qquad (6)$$

and then introduce the associated reduced variables  $A_k$ ,  $A_k^{\dagger}$ ,  $L_+$ ,  $L_-$ ,  $L_3$ , specified by

$$a_{k} = A_{k} e^{-i\omega_{k}t} , \quad a_{k}^{\dagger} = A^{\dagger} e^{i\omega_{k}t} ,$$
  
$$l_{+} = L_{*} e^{i\omega t} , \quad l_{-} = L_{*} e^{-i\omega t} , \quad l_{3} = L_{3} ,$$
 (7)

or, in general,

$$R = e^{-(i/\hbar)H_0 t} \gamma e^{(i/\hbar)H_0 t} , \qquad (8)$$

where r is a nonreduced variable, R is a reduced variable, and

$$H_0 = H_N + \sum_k H_k . \tag{9}$$

In the absence of coupling, the reduced variables are constants, and their (slow) time variation is due to the coupling only.

The interaction between the NO and the field is now given by

$$H' = \sum_{k} H_{kN}$$

$$= \frac{1}{2} \hbar \sum_{k} \left[ \gamma_{k} A_{k}^{\dagger} L_{-} e^{i\nu_{k}t} + \gamma_{k}^{*} A_{k} L_{+} e^{-i\nu_{k}t} \right. \\ \left. + \gamma_{k} A_{k} L_{-} e^{-i\tilde{\nu}_{k}t} + \gamma_{k}^{*} A_{k}^{\dagger} L_{+} e^{i\tilde{\nu}_{k}t} \right. \\ \left. + \Gamma_{b} L_{3} \left( A_{k} e^{-i\omega_{k}t} + A_{k}^{\dagger} e^{i\omega_{k}t} \right) \right], \quad (10a)$$

where

$$\nu_{k} \equiv \omega_{k} - \omega , \qquad \nu_{k} \equiv \omega_{k} + \omega ,$$
  

$$\gamma_{k} \equiv \gamma_{1k} + i\gamma_{2k} , \qquad \Gamma_{k} \equiv 2^{1/2}\gamma_{3k} .$$
(10b)

The effect of a prescribed resonant driving field (or force) acting on the NO may be described by

$$H'' = \frac{1}{2}\hbar [\hat{\gamma}(\hat{A}^{+}L_{-} + \hat{A}L_{-}e^{-2i\omega t}) + \hat{\gamma}^{*}(\hat{A}L_{+} + \hat{A}^{\dagger}L_{+}e^{2i\omega t})], \quad (11)$$

where  $\hat{A}$  is a (*c*-number) constant.<sup>18</sup> We may approximate H'' by dropping the rapidly oscillating terms, or, what amounts to the same thing, describe H'' in the rotating-wave approximation:

$$H^{\prime\prime} \approx \frac{1}{2} \hbar [\hat{\gamma} \hat{A}^{\dagger} L_{\perp} + \hat{\gamma}^{*} \hat{A} L_{\perp}] .$$
(12)

Although one frequently finds in the literature an analogous approximation to H', where the  $e^{\pm i\tilde{\nu}_k t}$  and the  $e^{\pm i\omega_k t}$  terms are dropped, it will be seen later that these terms play an essential role in determining radiative frequency shifts, and indeed, convergence of certain integrals. The complete Hamiltonian is now given by

$$H = H_0 + H_1$$
, (13a)

with

$$H_1 = H' + H''$$
 . (13b)

We will assume that the interaction between the NO and the field is "turned on" at t=0, so that initially both systems are described by their free states. Furthermore, in accordance with a previous statement, the interaction will be assumed to be sufficiently weak so that a significant change in the NO occurs only after a large number of cycles of its natural frequency. Analytically, this implies that the reduced variables change slowly compared to  $e^{i\omega t}$ .

The equations of motion, in terms of the reduced variables, are obtained from the relationship

$$i\hbar \dot{R} = [R, H_1]_P$$
, (14)

where R stands for any reduced variable. Noting that the only nonvanishing (equal-time) brackets are

$$[A_k, A_k^{\mathsf{T}}]_P = 1 , \qquad [L_+, \ L_-]_P = L_3 ,$$

$$[L_3, \ L_+]_P = L_+ , \qquad [L_3, \ L_-]_P = -L_- ,$$
(15)

we obtain

$$\dot{A}_{k} = -\frac{1}{2}i(\gamma_{k}L_{-}e^{i\nu_{k}t} + \gamma_{k}^{*}L_{+}e^{i\tilde{\nu}_{k}t} + \Gamma_{k}L_{3}e^{i\omega_{k}t}), \quad (16a)$$

$$\dot{A}_{k}^{\dagger} = \frac{1}{2}i(\gamma_{k}^{*}L_{*}e^{-i\nu_{k}t} + \gamma_{k}L_{-}e^{-i\tilde{\nu}_{k}t} + \Gamma_{k}L_{3}e^{-i\omega_{k}t}),$$
(16b)

$$\dot{L}_{+} = A^{*}L_{3} + \alpha^{\dagger}L_{3} - L_{3} \alpha + L_{+} \alpha - \alpha^{\dagger}L_{+},$$
 (16c)

$$\dot{L}_{2} = AL_{3} + L_{3} @ - @^{\dagger}L_{3} + @^{\dagger}L_{2} - L_{2} @ ,$$
 (16d)

$$\dot{L}_{3} = -(AL_{+} + A^{\dagger}L_{-}) - (L_{+}\alpha + \alpha^{\dagger}L_{-})$$

$$+ \oplus^{\dagger}L_{+} + L_{-} \oplus$$
, (16e)

where, for simplicity, we have introduced the notation  $% \left( {{{\left( {{{{{\bf{n}}}} \right)}_{{{\bf{n}}}}}}} \right)$ 

$$A \equiv \frac{1}{2}i\hat{\gamma}^*\hat{A} ,$$

$$a \equiv \frac{1}{2}i\sum_{k}\gamma_k^*A_k e^{-i\nu_k t} ,$$

$$a \equiv \frac{1}{2}i\sum_{k}\gamma_k^*A_k e^{-i\overline{\nu}_k t} ,$$

$$a \equiv \frac{1}{2}i\sum_{k}\gamma_k A_k e^{-i\overline{\omega}_k t} ,$$

$$a \equiv \frac{1}{2}i\sum_{k}\Gamma_k A_k e^{-i\omega_k t} .$$
(17)

We also have

$$L^{2} \equiv L_{3}^{2} + L_{+}L_{-} + L_{-}L_{+}$$

$$= \begin{cases} L_{0}^{2} & \text{classically} \\ L_{0}(L_{0}+1) & \text{quantum mechanically,} \end{cases}$$
(18)

where  $L_0$  is the constant that specifies the total angular momentum, classically, and the corresponding quantum number, quantum mechanically. These are the fundamental equations of motion, and are valid both classically and quantum mechanically. It should be noted that if the rotating-wave approximation had been used in H' (which describes the coupling of the NO to the radiation field), the  $e^{\pm i\tilde{\nu}_{k}t}$  and the  $e^{\pm i\omega_{k}t}$  terms would be missing in Eqs. (16a) and (16b), while the B and the C terms would be missing in Eqs. (16c)-(16e). It should also be noted that the order of the factors in the products of dynamical variables occurring on the right-hand sides of Eqs. (16c)-(16e) is immaterial, since the two (equal-time) variables in each product commute. However, for later purposes (of substitution, where variables at time t are expanded in terms of variables at other times), we write the products so that  $\alpha$  is on the right-hand side and  $a^{\dagger}$  is on the left-hand side of the NO variable, and do likewise for B and C.

Our main interest, in the present paper, will be concentrated on the behavior of the NO. Of course, if the NO variables are known, the field variables can be obtained from Eqs. (16a) and (16b). We proceed, therefore, to eliminate the field variables (except for their initial values) from Eqs. (16c)-(16e). From Eq. (16a), we have

$$\begin{aligned} \alpha(t) &= \alpha_0(t) + \frac{\tau}{4} \sum_k \int_0^t dt_1 \left[ \left| \gamma_k \right|^2 L_{-}(t_1) e^{-i\nu_k (t-t_1)} \right. \\ &+ \gamma_k^{*2} L_{+}(t_1) e^{-i(\nu_k t - \tilde{\nu}_k t_1)} + \gamma_k^{*} \Gamma_k L_3(t_1) e^{-i(\nu_k t - \omega_k t_1)} \right], \end{aligned}$$

$$(19a)$$

where

$$\alpha_0 = \frac{1}{2} i \sum_k \gamma_k^* A_k(0) e^{-i\nu_k t} \quad . \tag{19b}$$

For later purposes, we also define  $\mathfrak{B}_0$  and  $\mathfrak{C}_0$  analogously to  $\mathfrak{A}_0$ . [These quantities, although func-

tions of time, contain only the initial values of the (reduced) field variables, and may be regarded as the free-or uncoupled-values of  $\alpha$ ,  $\alpha$ , and  $\alpha$ . The only term in the integrand of Eq. (19a) which, when summed over k, can be expected to give a significant contribution is the first, for the following reasons: (i) Every  $|\gamma_k|^2$  is positive, while  $\gamma_k^{*2}$ and  $\gamma_k^* \Gamma_k$  fluctuate in sign as well as in magnitude, more or less independently of the exponent: (ii) the first exponent is the only one that contains terms that vary slowly with respect to  $t_1$  (for  $\nu_k$  sufficiently small); (iii) as  $t_1$  approaches  $t_1$ , the number of terms in the k summation which contribute to the integral becomes very large for the first term but not for the others. We, therefore, retain only the first term. Furthermore, because of the last reason, the main contribution to the  $t_1$  integration comes from  $t_1$  close to t. Since  $L_{-}(t_1)$  is a slowly varying function of  $t_1$ , we approximate  $L_{-}(t_1)$  by  $L_{(t)}$  and take it outside of the integral sign, obtaining

$$\alpha \approx \alpha_0 + \beta L_{(t)}, \qquad (20a)$$

where

$$\beta \equiv \frac{1}{4} \sum_{k} \int_{0}^{t} dt_{1} |\gamma_{k}|^{2} e^{-i\nu_{k}(t-t_{1})} .$$
 (20b)

We proceed to evaluate  $\beta$ . Integration yields

$$\beta = \frac{1}{4} \sum_{k} |\gamma_{k}|^{2} \frac{1 - \cos(\omega_{k} - \omega)t + i\sin(\omega_{k} - \omega)t}{i(\omega_{k} - \omega)} .$$
(21)

Two approximations will be made in this evaluation. The first consists of a conversion of the summation to an integration (since the normalization volume can be made arbitrarily large):

$$\beta \approx \frac{1}{4} \int_{0}^{\infty} d\omega_{k} |\gamma(\omega_{k})|^{2} \rho(\omega_{k})$$
$$\times \frac{1 - \cos(\omega_{k} - \omega)t + i\sin(\omega_{k} - \omega)t}{i(\omega_{k} - \omega)} , \quad (22)$$

where  $|\gamma(\omega_k)|^2$  is the *average* of  $|\gamma_k|^2$  over all k's corresponding to a small neighborhood about  $\omega_k$ , and  $\rho(\omega_k) \Delta \omega_k$  is the number of modes in the interval  $\Delta \omega_k$  at  $\omega_k$ . The second approximation is based on the fact that the NO behavior changes slowly compared to a natural cycle of oscillation. The quotient in the integrand of Eq. (22) can be approximated quite easily for t sufficiently large. For t much larger than a natural cycle [but small compared to a significant change in the NO, so that  $\beta$ , in Eq. (20a), may be considered essentially independent of the time; strictly speaking  $\beta - 0$  as  $t \to 0$ , from Eq. (20b)], we can write

$$\beta \approx \frac{1}{4} \int_0^\infty d\omega_k \left| \gamma(\omega_k) \right|^2 \rho(\omega_k) \left( \pi \delta(\omega_k - \omega) - i \frac{\vartheta}{\omega_k - \omega} \right),$$
(23)

or, for notational convenience,

(24)

 $\beta \equiv \alpha_1 - i\alpha_2 ,$ 

where  $\alpha_1$  and  $\alpha_2$  are real constants given by

$$\alpha_1 \equiv \frac{1}{4}\pi |\gamma(\omega)|^2 \rho(\omega)$$
(25a)

and

$$\alpha_2 \equiv \frac{1}{4} \int_0^\infty d\omega_k \left| \gamma(\omega_k) \right|^2 \rho(\omega_k) \frac{\mathcal{O}}{\omega_k - \omega} \quad . \tag{25b}$$

The expression for  $\alpha$  can now be written as<sup>19</sup>

$$\alpha = \alpha_0 + (\alpha_1 - i\alpha_2)L_{-}$$

The Hermitian conjugate relation yields

$$\alpha^{\dagger} = \alpha_0^{\dagger} + (\alpha_1 + i\alpha_2)L_+ . \tag{27}$$

Using similar reasoning with an obvious slight modification, we obtain

$$\mathfrak{B} = \mathfrak{B}_0 - i\alpha_3 L_+ , \qquad (28a)$$

$$\mathscr{B}^{\dagger} = \mathscr{B}_{0}^{\dagger} + i\alpha_{3}L_{-} , \qquad (28b)$$

$$\mathbf{C} = \mathbf{C}_0 - i\alpha_4 L_3 , \qquad (29a)$$

$$\mathbf{C}^{\dagger} = \mathbf{C}_{\mathbf{0}}^{\dagger} + i\alpha_{\mathbf{4}}L_{\mathbf{3}} , \qquad (29b)$$

where

$$\alpha_{3} \equiv \frac{1}{4} \int_{0}^{\infty} d\omega_{k} \left| \gamma(\omega_{k}) \right|^{2} \rho(\omega_{k}) \frac{1}{\omega_{k} + \omega} , \qquad (30a)$$

$$\alpha_4 \equiv \frac{1}{4} \int_0^\infty d\omega_k \, \Gamma^2(\omega_k) \rho(\omega_k) \frac{1}{\omega_k} \, , \qquad (30b)$$

the calculation of  $\alpha_4$  being based on the assumption  $\Gamma^2(0)\rho(0)=0$ .

With the above expressions for the field variables, the equations of motion for the NO become

$$\dot{L}_{+} = A^{*}L_{3} + [\alpha_{1} + i(\alpha_{2} - \alpha_{4})]L_{+}L_{3} + i(\alpha_{3} - \alpha_{4})L_{3}L_{+} + F_{0},$$
(31a)
$$\dot{L}_{-} = AL_{3} + [\alpha_{1} - i(\alpha_{2} - \alpha_{4})]L_{3}L_{-} - i(\alpha_{3} - \alpha_{4})L_{-}L_{3} + F_{0}^{\dagger},$$
(31b)

$$\dot{L}_3 = -(AL_+ + A^*L_-) - 2\alpha_1L_+L_- + G_0$$
, (31c)

where

$$F_{0} \equiv \alpha_{0}^{\dagger} L_{3} - L_{3} \mathcal{B}_{0} + L_{+} \mathcal{C}_{0} - \mathcal{C}_{0}^{\dagger} L_{+}, \qquad (31d)$$

and

$$G_{0} \equiv - (L_{+} \alpha_{0} + \alpha_{0}^{\dagger} L_{-}) + L_{-} \alpha_{0} + \alpha_{0}^{\dagger} L_{+}.$$
 (31e)

These equations contain only the NO variables, the driving field, and the initial (or free) radiation field. It should be observed that the field variables and the NO variables in  $F_0$  and  $G_0$  no longer commute, and their order is significant. As mentioned previously, the field variables can be obtained from the NO variables, if the latter are known. Of particular interest is the rate of change of field energy, given by

$$\sum_{k} \dot{H}_{k} = \sum_{k} \hbar \omega_{k} (\dot{A}_{k}^{\dagger} A_{k} + A_{k}^{\dagger} \dot{A}_{k}) .$$
(32)

Using approximation methods employed above, we obtain from Eqs. (16a) and (16b)

$$\sum_{k} H_{k} = 2\hbar\omega \alpha_{1} L_{+} L_{-} + \hbar\omega J_{0} , \qquad (33)$$

where

- .

$$J_0 \equiv L_+ \tilde{\alpha}_0 + L_- \tilde{\alpha}_0 + L_3 \tilde{\mathcal{C}}_0 + \text{H.c.}$$

the tilde indicating that the factor  $(\omega_k/\omega)$  is inserted under the summation sign in the definition of  $\alpha_0$ ,  $\alpha_0$ , and  $\alpha_0$ ; that is,

$$\tilde{\alpha}_{0} \equiv \frac{1}{2} \sum_{k} (\omega_{k}/\omega) \gamma_{k}^{*} A_{k}(0) e^{-i\nu_{k}t} , \text{ etc}$$

No operations have vet been performed that invalidate the equations for either a guantum-mechanical or a classical analysis. Before specialization to one of the two analyses, we specify that the initial conditions for the radiation field are those of the ground state. Although we will consider various initial conditions for the NO and various amplitudes for the driving field (which is a specified c number), the radiation field will be assumed to be initially unexcited throughout the present paper. The reason for the choice of order of the field and NO variables in the terms of the original equations of motion is now apparent. The initial field variables in the final equations of motion are now ordered so that we can take advantage of the relationships

$$|\alpha_0|\rangle = |\alpha_0|\rangle = |\alpha_0|\rangle = \langle |\alpha_0^{\dagger} = \langle |\alpha_0^{\dagger} = \langle |\alpha_0^{\dagger} = 0, \quad (34)$$

where  $|\rangle$  represents the (initial) state used in Heisenberg-picture calculations.

### III. CLASSICAL ANALYSIS

The system will be analyzed classically, first. For this purpose all variables in the equations of motion are to be considered c numbers; the initial field conditions imply

$$F_0 = G_0 = J_0 = 0 ; (35)$$

we have the relation

$$L^2 = L_0^2 = 2L_+ L_- + L_3^2 , (36)$$

where  $L_0$  is the (constant) total angular momentum, and the equations of motion for the NO [Eqs. (31)] become

$$\dot{L}_{+} = A^{*}L_{3} + (\alpha_{1} + i\tilde{\alpha})L_{+}L_{3}$$
, (37a)

$$\dot{L}_{2} = AL_{3} + (\alpha_{1} - i\tilde{\alpha})L_{2}L_{3},$$
 (37b)

$$\dot{L}_3 = -(AL_+ + A^*L_-) - \alpha_1(L_0^2 - L_3^2), \qquad (37c)$$

where

$$\tilde{\alpha} \equiv \alpha_2 + \alpha_3 - 2\alpha_4 \quad . \tag{37d}$$

Equation (37c) shows that the energy (in units of  $\hbar\omega$ ) absorbed from the driving field by the NO is  $-(AL_++A^*L_-)$ , and Eq. (37c) together with Eq. (33) shows that the energy radiated by the NO into

the field is 
$$\alpha_1(L_0^2 - L_3^2)$$
. Since  $L_* = L_*^*$ , we can write

$$L_{\pm} = \left[\frac{1}{2}(L_0^2 - L_3^2)\right]^{1/2} e^{\pm i\theta}, \qquad (38)$$

and, using the notation

$$\Omega^2 \equiv 2 \left| A \right|^2 , \tag{39}$$

we can likewise express A in terms of an amplitude and phase (both of which are constant),

$$A = 2^{-1/2} \Omega e^{-i\varphi} . (40)$$

The power absorbed by the NO from the driving field is

$$-\Omega(L_0^2-L_3^2)^{1/2}\cos(\theta-\varphi)$$
,

and Eq. (37c) reads

$$\dot{L}_3 = -\Omega (L_0^2 - L_3^2)^{1/2} \cos(\theta - \varphi) - \alpha_1 (L_0^2 - L_3^2) , \quad (41)$$

a form that will be useful later. Both the free decay of an initially excited NO and the behavior of a driven NO are of interest.

#### A. Free Decay

If A = 0, Eq. (37c) can be solved immediately:

$$L_{3} = -L_{0} \tanh \alpha_{1} L_{0} (t - t_{0}) , \qquad (42a)$$

where

$$t_0 = (1/\alpha_1 L_0) \tanh^{-1} [L_3(0)/L_0] .$$
 (42b)

Except for the initial condition  $L_3(0) = L_0$ ,  $L_3(t)$ decays monotonically to  $-L_0$ . If  $L_3(0) = L_0$ , then  $L_3(t)$  remains constant at  $L_0$ . One may refer to this as the "unstable equilibrium" condition. The rate of decay is given by

$$-L_3 = \alpha_1 L_0^2 \operatorname{sech}^2 \alpha_1 L_0 (t - t_0) , \qquad (43)$$

and assumes its maximum value  $\alpha_1 L_0^2$  when  $L_3 = 0$ . As  $L_3(0)$  approaches  $L_0$ , one can regard  $t_0$  as a measure of the decay time, which approaches infinity. Specifically, if

$$L_3(0) = L_0(1 - \epsilon), \quad \epsilon \ll 1 \tag{44}$$

then

$$t_0 = (1/\alpha_1 L_0) \tanh^{-1}(1 - \epsilon)$$
  
 
$$\approx (1/\alpha_1 L_0) \frac{1}{2} \ln(2/\epsilon) , \qquad (45)$$

so that, approximately,

$$L_{3} = -L_{0} \tanh[L_{0}\alpha_{1}t - \frac{1}{2}\ln(2/\epsilon)] .$$
 (46)

In addition to the energy of the NO, which is described by  $L_3$ , the oscillating coordinates, which are described by  $L_{\pm}(t)$ , are also of interest. From Eq. (37a), in the case of free decay, we obtain

. .

$$L_{+}(t) = L_{+}(0) \exp[(\alpha_{1} + i\tilde{\alpha}) \int_{0}^{t} dt_{1} L_{3}(t_{1})]$$
  
=  $L_{+}(0) \exp[\alpha_{1} \int_{0}^{t} dt_{1} L_{3}(t_{1})] \exp[i\tilde{\alpha} \int_{0}^{t} dt_{1} L_{3}(t_{1})].$   
(47)

Setting

$$L_{+}(0) = |L_{+}(0)| e^{i\theta_{0}}, \qquad (48)$$

and noting that

$$L_{\pm}(t) = \left\{ \frac{1}{2} \left[ L_0^2 - L_3^2(t) \right] \right\}^{1/2}, \qquad (49)$$

we can write the expression for  $L_{+}(t)$  in the alternate form

$$L_{*}(t) = \left\{ \frac{1}{2} \left[ L_{0}^{2} - L_{3}^{2}(t) \right] \right\}^{1/2} \exp i \left[ \theta_{0} + \tilde{\alpha} \int_{0}^{t} dt_{1} L_{3}(t_{1}) \right] .$$
(50)

Let us consider  $L_3(0)$  only slightly less than  $L_0$ . Our explicit expressions for  $L_3(t)$  show that  $L_3$  remains close to  $L_0$  for a long time [of the order of  $t_0$ , as given by Eq. (45)], then passes from 0.76 $L_0$  $(= L_0 \tanh 1)$  to  $-0.76L_0$  in the time interval  $2(\alpha_1 L_0)^{-1}$ , and finally approaches  $-L_0$  asymptotically. The magnitude of  $L_{+}(t)$ —or the amplitude of oscillation of the NO-thus increases from a small quantity to a maximum (when  $L_3 = 0$ ) and then decreases again. The frequency of oscillation, which is  $\omega$  for the free NO [see Eqs. (7)], is affected by coupling to the field through the term containing the factor  $\tilde{\alpha}$  in the imaginary exponent of  $L_{\star}(t)$ above. When the relative change in  $L_3$  during the time  $\omega^{-1}$  is small, we can regard  $\tilde{\alpha}L_3$  as a frequency shift, the effective frequency of the NO being  $\omega + \tilde{\alpha} L_3$ .

It may be remarked, at this point, that  $\tilde{\alpha}$ , and therefore the frequency shift, is determined, in part, by terms that would have been neglected in the rotating-wave approximation, since  $\alpha_3$  and  $\alpha_4$ come from such terms. If we consider the NO to be coupled to the field by a dipole moment that is proportional to the angular momentum, then

$$\gamma_{1k} = \gamma_{2k} = \gamma_{3k} , \qquad (51)$$

so that, from Eqs. (10b),

$$\left|\gamma_{k}\right|^{2}=\Gamma_{k}^{2}, \qquad (52)$$

and

$$\tilde{\alpha} = \alpha_2 + \alpha_3 - 2\alpha_4$$

$$= \int_0^\infty d\omega_k |\gamma(\omega_k)|^2 \rho(\omega_k) \left( \frac{\varphi}{\omega_k - \omega} + \frac{1}{\omega_k + \omega} - \frac{2}{\omega_k} \right)$$

$$= \int_0^\infty d\omega_k |\gamma(\omega_k)|^2 \rho(\omega_k) \frac{2\omega^2 \varphi}{\omega_k (\omega_k^2 - \omega^2)} \quad .$$
(53)

It is seen that  $\alpha_3$  and  $\alpha_4$  not only affect the value of  $\tilde{\alpha}$ , but may also affect the convergence properties of the integral (if no cutoff is used), producing a  $\omega_k^{-3}$  variation rather than a  $\omega_k^{-1}$  variation in the last factor of the integrand as  $\omega_k \rightarrow \infty$ . The actual value of the integral depends, of course, on the functions  $|\gamma(\omega_k)|^2$  and  $\rho(\omega_k)$ , specification of which requires specialization that will not be made in the present article.

### **B.** Forced Oscillation

The effect of the driving field will now be considered. We approximate by neglecting the frequency shift in the NO produced by the coupling to the radiation field; this approximation is accomplished by the assumption

$$\tilde{\alpha} = 0 . \tag{54}$$

Setting  $\alpha_1 \equiv \alpha$  for notational simplicity, we obtain from Eqs. (37)

$$\dot{L}_{+} = A^* L_3 + \alpha L_{+} L_3 , \qquad (55a)$$

$$\dot{L}_{2} = AL_{3} + \alpha L_{2}L_{3} , \qquad (55b)$$

$$\dot{L}_3 = -(AL_+ + A^*L_-) - \alpha(L_0^2 - L_3^2) .$$
 (55c)

Differentiating Eq. (55c), substituting for  $L_{\star}$  and  $L_{\pm}$  from the two preceding equations, and using Eq. (55c) again, one obtains a second-order differential equation for  $L_{3}$  only,

$$\ddot{L}_{3} - 3\alpha L_{3}\dot{L}_{3} + [\Omega^{2} - \alpha^{2}(L_{0}^{2} - L_{3}^{2})]L_{3} = 0 , \qquad (56)$$

where, it is recalled,  $\Omega^2 = 2|A|^2$ . In the case of the undriven NO, our interest lay in the free decay of the system, and was therefore related to an initial excited state. In the present instance the effect of the driving field is illustrated best by considering initial conditions in which the NO is in the ground state. We consider, therefore, the initial condition

$$L_3(0) = -L_0 , (57)$$

which immediately implies, in the classical description,

$$L_{+}(0) = L_{-}(0) = 0$$
,  $L_{3}(0) = 0$ . (58)

These initial conditions determine uniquely a solution of the above second-order differential equation for  $L_3$ . This equation is, however, a nonlinear differential equation which does not appear to have an exact solution in closed form. We will, therefore, study the properties of approximate solutions for special, but interesting, relationships between the coefficients, and then interpolate, qualitatively, for some of the remaining relationships.

Consider, first, the case

$$\Omega \ll \alpha L_0 , \qquad (59)$$

that is, the case of a weak driving field. In the absence of a driving field,  $\Omega^2 = 0$ , and for the above initial conditions the solution is, of course, the stable equilibrium solution  $L_3 = -L_0$ . For a weak driving field,  $L_3$  will increase only slightly above  $-L_0$ . [This can be seen by noting that the power absorbed from the driving field is of the order of  $\Omega(L_0^2 - L_3^2)^{1/2}$ , while the power radiated by the NO is  $\alpha(L_0^2 - L_3^2)$ , so that when the two are of the same order of magnitude,  $1 - (L_3/L_0)^2 \sim \Omega^2/\alpha^2 L_0^2$ .] We

therefore set

$$L_3 = -L_0(1-x) , (60)$$

treat  $(\Omega^2/\alpha^2 L_0^2)$  and x as small quantities of first order, and retain only lowest-order quantities in Eq. (56). This equation then becomes

$$\ddot{x} + 3\alpha L_0 \dot{x} + 2\alpha^2 L_0^2 x - \Omega^2 = 0 , \qquad (61)$$

with the initial conditions  $x(0) = \dot{x}(0) = 0$ . The solution is easily seen to be

$$x = \frac{1}{2} \frac{\Omega^2}{\alpha^2 L_0^2} \left( 1 - 2e^{-\alpha L_0 t} + e^{-2\alpha L_0 t} \right), \qquad (62)$$

which indicates a monotonic increase of  $L_3$  from  $-L_0$  to the steady-state (or constant) value  $-L_0$  $+\frac{1}{2}(\Omega^2/\alpha^2 L_0^2).$ 

In the steady state, we have  $\dot{L}_3 = 0$ , and Eq. (41) yields (for arbitrary driving field)

$$-\Omega(L_0^2 - L_3^2)^{1/2}\cos(\theta - \varphi) = \alpha(L_0^2 - L_3^2), \qquad (63)$$

where the left-hand side is power absorbed, and the right-hand side is power radiated. Substitution of the above steady-state value of  $L_3$  (to lowest order in x) shows that  $-\cos(\theta - \varphi)$  must be equal to unity. In other words, in the steady state the induced oscillation of the NO has a phase such as to maximize the energy absorption from the driving field. The last statement can be seen to be true for all driving fields such that

$$\Omega < \alpha L_0 . \tag{64}$$

Equation (63), together with this inequality, implies  $L_3 \neq 0$ . Equation (56) then implies that in the steady state we must have

$$\Omega^2 = \alpha^2 (L_0^2 - L_3^2) . \tag{65}$$

Taking the value of  $L_0^2 - L_3^2$  thus obtained and substituting in Eq. (63), we obtain

$$-\cos(\theta - \varphi) = 1 . \tag{66}$$

Although we have explicitly solved the equations of motion [for the initial condition  $L_3(0) = -L_0$ ] only for the range of driving-field strength given by inequality (59) rather than for the wider range given by inequality (64), we may reasonably extrapolate qualitatively by assuming that, in this wider range,  $L_3$ increases monotonically and approaches a constant (negative) value given by Eq. (65), at which the power absorbed equals the power radiated. One may regard the equation  $\Omega = \alpha L_0$  as the specification of the saturation driving-field strength at which the steady-state value  $L_3 = 0$  can be maintained for optimum phase relationship between driving field and NO. In this steady state, the NO radiates the maximum power, as mentioned previously.

We consider now the other extreme relationship,

$$\Omega \gg \alpha L_0 . \tag{67}$$

Under this condition, the driving field is sufficiently strong so that the maximum power that can be absorbed by the NO is much greater than the maximum power that the NO can radiate. Equation (56) becomes, approximately,

$$\ddot{L}_3 - 3\alpha L_3 \dot{L}_3 + \Omega^2 L_3 = 0 . (68)$$

This equation may be regarded as being derived from Eq. (56) by retaining the first-order term in  $\alpha L_0/\Omega$  but dropping the second-order term. If we drop the first-order term also, we obtain the equation which describes the well-known sinusoidal energy oscillation of an undamped angular momentum oscillator being driven by a resonant force. This zeroth-order solution, for the present initial conditions, is

$$L_3^{(0)} = -L_0 \cos\Omega t \; . \tag{69}$$

An explicit closed-form solution of Eq. (68) has not been found. An analysis of the equation in the  $(L_3, \dot{L}_3)$  plane, carried out in Appendix B, does yield considerable information, however. The most important result, from the present point of view, is the fact that the exact solution of Eq. (68) is *periodic*, and oscillates between  $-L_0$  and  $L_0$ . The period, to first order in  $\alpha L_0/\Omega$ , is unaffected by coupling to the radiation field (although the oscillation is no longer sinusoidal), and is given by

$$T = 2\pi/\Omega \quad . \tag{70}$$

The above discussion of forced oscillation can be summarized qualitatively by the statement that for a weak driving field, the NO, initially in the ground state, approaches asymptotically a constant (negative) energy, while for a strong driving field, the NO energy oscillates periodically between  $-L_0$ and  $L_0$ .

#### IV. QUANTUM-MECHANICAL ANALYSIS

We consider, now, the quantum-mechanical description of the NO coupled to the radiation field. In the classical results,  $L_0$  appears as a parameter and the expressions are applicable to arbitrary  $L_0$ . In the quantum-mechanical analysis, systems corresponding to different values of  $L_0$ , especially low values, require individual consideration. We begin with  $L_0 = \frac{1}{2}$ , that is, with the case in which the NO is a two-level system.

### A. Two-Level System

Equations (31) become simplified for a two-level system. In this case, we have

$$L_3 L_4 = -L_4 L_3 = \frac{1}{2} L_4 , \qquad (71a)$$

$$L_3 L_{-} = -L_{-} L_3 = -\frac{1}{2} L_{-}, \qquad (71b)$$

$$L_{+}L_{-} = \frac{1}{2}(L_{3} + \frac{1}{2}); \qquad (71c)$$

the terms containing  $\alpha_4$  cancel, and we obtain (re-

calling that we have set  $\alpha_1 \equiv \alpha$ )

$$\dot{L}_{+} = A^{*} L_{3} - \frac{1}{2} \left[ \alpha + i(\alpha_{2} - \alpha_{3}) \right] L_{+} + F_{0} , \qquad (72a)$$

$$\dot{L}_{2} = A L_{3} - \frac{1}{2} \left[ \alpha - i(\alpha_{2} - \alpha_{3}) \right] L_{2} + F_{0}^{+},$$
 (72b)

$$\dot{L}_3 = -(AL_+ + A^*L_-) - \alpha(L_3 + \frac{1}{2}) + G_0$$
 (72c)

We calculate first the expectation values of  $L_{*}$ ,  $L_{-}$ , and  $L_{3}$ . The initial conditions for the radiation field imply [see Eqs. (34)]

$$\langle F_0 \rangle = \langle G_0 \rangle = 0 , \qquad (73)$$

and the expectation-value equations of motion become

$$\langle \dot{L}_{*} \rangle = A^{*} \langle L_{3} \rangle - \frac{1}{2} (\alpha + i\alpha') \langle L_{*} \rangle,$$
 (74a)

$$\langle \dot{L}_{-} \rangle = A \langle L_{3} \rangle - \frac{1}{2} (\alpha - i\alpha') \langle L_{-} \rangle$$
, (74b)

$$\langle \dot{L}_3 \rangle = - (A \langle L_* \rangle + A^* \langle L_- \rangle) - \alpha (\langle L_3 \rangle + \frac{1}{2}), \quad (74c)$$

where

$$\alpha' \equiv \alpha_2 - \alpha_3 . \tag{74d}$$

### 1. Free Decay

As in the case of the classical analysis, the free decay of the NO will be analyzed first. For A = 0, the equations of motion yield the solution

$$\langle L_{+} \rangle = \langle L_{+}(0) \rangle e^{-(\alpha + i\alpha')t/2} , \qquad (75a)$$

$$\langle L_{-}\rangle = \langle L_{-}(0)\rangle e^{-(\alpha-i\alpha')t/2}$$
, (75b)

$$\langle L_3 \rangle = -\frac{1}{2} + (\langle L_3(0) \rangle + \frac{1}{2}) e^{-\alpha t}$$
 (75c)

From Eqs. (7), which relate the reduced variables to the original (oscillating) variables, one sees that  $\alpha'$  accounts for an effective frequency shift, and produces a replacement of  $\omega$  by  $\omega - \frac{1}{2}\alpha'$  in the oscillating factors of  $\langle l_+ \rangle$  and  $\langle l_- \rangle$ . Looking at this frequency shift in greater detail, we have, from Eqs. (25b) and (30a).

$$\alpha' = \alpha_2 - \alpha_3$$

$$= \int_0^\infty d\omega_k |\gamma(\omega_k)|^2 \rho(\omega_k) \left(\frac{\varphi}{\omega_k - \omega} - \frac{1}{\omega_k + \omega}\right)$$

$$= \int_0^\infty d\omega_k |\gamma(\omega_k)|^2 \rho(\omega_k) \frac{2\omega\varphi}{\omega_k^2 - \omega^2} \quad . \tag{76}$$

It is interesting to note again that had we used the rotating-wave approximation for the coupling between NO and radiation field, the frequency shift would have been  $\alpha_2$ , which is not only different from  $\alpha'$ , but, in its integral form, has weaker convergence properties.

If  $\langle L_3(0) \rangle = L_0 = \frac{1}{2}$ ,  $\langle L_3(t) \rangle$  yields, essentially, the Weisskopf-Wigner result<sup>4</sup> for the exponential decay of the upper-level occupation probability. That which appears in the Weisskopf-Wigner theory as a level shift appears in the present theory as a shift in the frequency of oscillation of the two-level oscillator. This frequency shift is also exhibited

$$\left\langle \pm \frac{1}{2} \left| L_{\pm}(t) \right| \mp \frac{1}{2} \right\rangle \approx \left\langle \pm \frac{1}{2} \left| L_{\pm}(0) \right| \mp \frac{1}{2} \right\rangle e^{\mp i \, \alpha' t / 2} \,. \tag{77}$$

The corresponding elements for  $l_{\star}$  are obtained by multiplication with the factor  $e^{\star i \omega t}$ .

#### 2. Forced Oscillation

Let us consider, next, the case of a driven twolevel NO. We will approximate (as in the case of the classical forced oscillator), by neglecting the radiative frequency shift. The equations of motion for the expectation values become

$$\langle \dot{L}_{+} \rangle = A^{*} \langle L_{3} \rangle - \frac{1}{2} \alpha \langle L_{+} \rangle ,$$
 (78a)

$$\langle \dot{L}_{-} \rangle = A \langle L_{3} \rangle - \frac{1}{2} \alpha \langle L_{-} \rangle$$
, (78b)

$$\langle \dot{L}_3 \rangle = - (A \langle L_* \rangle + A^* \langle L_- \rangle) - \alpha (\langle L_3 \rangle + \frac{1}{2}) .$$
 (78c)

Differentiating Eq. (78c), substituting from Eqs. (78a) and (78b), and then substituting for  $(A \langle L_{+} \rangle + A^* \langle L_{-} \rangle)$  from Eq. (78c) in the result, we obtain

$$\langle \ddot{L}_3 \rangle + \frac{3}{2} \alpha \langle \dot{L}_3 \rangle + (\Omega^2 + \frac{1}{2} \alpha^2) \langle L_3 \rangle + \frac{1}{4} \alpha^2 = 0$$
. (79)

The initial condition is chosen, analogously to the classical treatment, to be the ground state. We, therefore, have

$$\langle L_3(0) \rangle = -\frac{1}{2}, \ \langle \dot{L}_3(0) \rangle = 0.$$
 (80)

The solution is then given by

$$\langle L_3 \rangle = -\frac{1}{2} \left( 1 + \frac{2\Omega^2}{\alpha^2} \right)^{-1} + K_* e^{k_* t} + K_* e^{k_* t} , \qquad (81)$$

where

$$k_{\pm} = -\frac{3}{4}\alpha \pm \frac{1}{4}\alpha \left(1 - \frac{16\Omega^2}{\alpha^2}\right)^{1/2},$$
 (82a)

$$K_{\pm} = \left(1 + \frac{\alpha^2}{2\Omega^2}\right)^{-1} \left[ -\frac{1}{4} \mp \frac{3}{4} \left(1 - \frac{16\Omega^2}{\alpha^2}\right)^{-1/2} \right]$$
(82b)

for  $\alpha > 4\Omega$ , and

$$k_{\pm} = -\frac{3}{4}\alpha \pm i\Omega \left(1 - \frac{\alpha^2}{16\Omega^2}\right)^{1/2},$$
 (83a)

$$K_{\pm} = \left(1 + \frac{\alpha^2}{2\Omega^2}\right)^{-1} \left[ -\frac{1}{4} \pm \frac{3}{16} \frac{i\alpha}{\Omega} \left(1 - \frac{\alpha^2}{16\Omega^2}\right)^{-1/2} \right]$$
(83b)

for  $\alpha \leq 4\Omega$ .

This result shows that for a weak driving field  $(\Omega \leq \frac{1}{4}\alpha), \langle L_3 \rangle$  approaches asymptotically a steadystate value, which increases with increasing field from  $-\frac{1}{2}$  until it reaches  $-\frac{4}{9}$ . For a sufficiently strong field  $(\Omega > \frac{1}{4}\alpha), \langle L_3 \rangle$  oscillates initially, the frequency of oscillation being  $\Omega(1 - \alpha^2/16\Omega^2)^{1/2}$ , but these oscillations are damped by the factor  $e^{(-3\alpha t)/4}$ . After a time larger than  $\alpha^{-1}$ ,  $\langle L_3 \rangle$  approaches a constant negative value which is greater than  $-\frac{4}{9}$  and increases asymptotically to zero as the field increases. Qualitatively, the weak-driving-field behavior of  $\langle L_3 \rangle$  is similar to that of the classical  $L_3$ , but the strong-driving-field behavior of these two quantities is different. While the latter oscillates periodically, for a strong driving field, the oscillations of the former are exponentially damped. We shall see that this qualitative difference persists as the number of levels of the NO increases.

Once  $\langle L_3(t) \rangle$  is known,  $\langle L_{\pm}(t) \rangle$  can be obtained immediately (for arbitrary initial conditions) from Eqs. (78a) and (78b):

$$\langle L_{\star}(t) \rangle = \langle L_{\star}(0) \rangle e^{-\alpha t/2} + A^{*} \int_{0}^{t} dt_{1} \langle L_{3}(t_{1}) \rangle e^{-\alpha (t-t_{1})/2}; \quad (84)$$

the complex-conjugate equation yields  $\langle L_{-}(t) \rangle$ . For a two-level system, all products of the (equaltime) operators  $L_3$ ,  $L_{\pm}$  can be expressed in linear form by means of Eqs. (71) and the additional relationships  $L_3^2 = \frac{1}{4}$ ,  $L_{+}^2 = L_{-}^2 = 0$ . Thus, from a knowledge of  $\langle L_3 \rangle$  and  $\langle L_{\pm} \rangle$ , one obtains immediately the expectation values of all products of the fundamental operators.

#### **B.** Multilevel Systems

We proceed to an analysis of the behavior of NO's with  $L_0 \ge \frac{1}{2}$  coupled to the radiation field. As in the previous cases, the free decay will be studied first.

#### 1. Free Decay

A study of the problem in a nonperturbative manner is greatly facilitated by dropping the terms whose main effect is the production of a frequency shift. Before making this approximation, however, we examine the frequency shift by means of a perturbation-theory argument. Using the notation

$$L_3(0) \mid m \rangle = m \mid m \rangle , \qquad (85)$$

one obtains from Eq. (31a), for a time small compared to  $\alpha^{-1}$ ,

$$\langle m | L_{\star}(t) | m - 1 \rangle$$
  

$$\approx \langle m | L_{\star}(0) | m - 1 \rangle \exp \{ [im(\alpha_{2} + \alpha_{3} - 2\alpha_{4}) - i(\alpha_{2} - \alpha_{4})]t \}, \quad (86)$$

and the complex-conjugate relationship for  $\langle m-1 \times | L_{-}(t) | m \rangle$ . It is easily seen that for  $m = \frac{1}{2}$ , this equation reduces to Eq. (77), and for *m* large, the frequency shift indicated here is approximately the same as the classical frequency shift obtained from Eq. (50).<sup>20</sup>

Resuming the nonperturbative approach, we find it necessary—for reasons that will be apparent later—to return to the equations of motion in which the NO variables commute with the field variables, Eqs. (16). The approximation of dropping the frequency-shift terms involves the neglect of the terms containing the factor  $\mathfrak{B}$  or  $\mathfrak{C}$  in these equations. This approximation is then equivalent to the rotating-wave approximation, or the retention of only resonant terms in the equations of motion. (Generally speaking, resonant terms produce mainly an energy transfer, while nonresonant terms produce mainly a frequency shift.) To be consistent, we also neglect the frequency-shift constant  $\alpha_2$  in Eq. (26). The equations of motion, in the absence of a driving field, are now

$$\dot{L}_{+} = \mathbf{a}^{\dagger} L_{3} , \qquad (87a)$$

$$\dot{L}_{-} = L_{3} \alpha \quad , \tag{87b}$$

$$\dot{L}_3 = -L_* \alpha - \alpha^{\dagger} L_-, \qquad (87c)$$

with

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$$\boldsymbol{\alpha} = \boldsymbol{\alpha}_0 + \alpha \boldsymbol{L}_-, \quad \boldsymbol{\alpha}^{\dagger} = \boldsymbol{\alpha}_0^{\dagger} + \alpha \boldsymbol{L}_+.$$
 (88)

Our interest will be directed chiefly at  $L_3$ , the energy of the NO (in units of  $\hbar\omega$ ).

It is convenient to introduce the dimensionless time

$$\tau \equiv \alpha t \tag{89}$$

and to indicate the *n*th derivative of  $L_3$  with respect to  $\tau$  by a parenthetical superscript (*n*). Equations (87c) and (88), together with the relationship

$$2L_{+}L_{-}=L^{2}-L_{3}^{2}+L_{3}$$
, (90)  
yield

$$\langle L_3^{(1)} \rangle = \langle L_3^2 - L_3 - L^2 \rangle , \qquad (91)$$

which expresses the first derivative of  $\langle L_3 \rangle$  as the expectation value of a quadratic polynomial in  $L_3$ . Equation (91) is not, of course, a differential equation for  $\langle L_3 \rangle$ , since  $\langle L_3^2 \rangle \neq \langle L_3 \rangle^2$ , in general. The following procedure is aimed at deriving such an equation; it will also provide information about the higher moments of  $L_3$ .

We set up a hierarchy of equations for successively higher-order derivatives of  $\langle L_3 \rangle$ , in which the derivative is expressed as the expectation value of a polynomial in  $L_3$ . Consider the second derivative, which, from Eq. (91), is given by

$$\langle L_3^{(2)} \rangle = \langle L_3^2 \rangle^{(1)} - \langle L_3^{(1)} \rangle .$$

$$(92)$$

The equations of motion yield

$$\langle L_3^2 \rangle^{(1)} = \langle L_3 L_3^{(1)} + L_3^{(1)} L_3 \rangle$$
  
=  $-\alpha^{-1} \langle L_3 L_+ \alpha + L_3 \alpha^{\dagger} L_-$   
 $+ L_+ \alpha L_3 + \alpha^{\dagger} L_- L_3 \rangle .$ (93)

Since a and  $a^{\dagger}$  commute with  $L_3$ ,  $L_*$ , and  $L_-$ , we can move a to the extreme right of the term in which it appears, move  $a^{\dagger}$  to the extreme left of the term in which it appears, and then substitute for a and  $a^{\dagger}$  from Eq. (88). The expectation value of each term removes the terms containing  $a_0$  and  $a_0^{\dagger}$ , according to Eqs. (34), and we have

$$\langle L_3^2 \rangle^{(1)} = - \langle L_3 L_+ L_- + 2L_+ L_3 L_- + L_+ L_- L_3 \rangle.$$
 (94)

Using Eqs. (15) and (90), one obtains

$$\langle L_3^2 \rangle^{(1)} = \langle (L_3^2 - L_3 - L^2) (2L_3 - 1) \rangle .$$
 (95)

Thus, the second derivative can be expressed as

$$\langle L_3^{(2)} \rangle = 2 \langle L_3^3 - 2L_3^2 + (1 - L^2) L_3 + L^2 \rangle ,$$
 (96)

the expectation value of a cubic polynomial in  $L_3$ . The third derivative of  $\langle L_3 \rangle$  is obtained by differentiating Eq. (96), which means that we need the (first) derivative of  $\langle L_3^3 \rangle$  as the expectation value of a polynomial in  $L_3$ . It is shown in Appendix C, using a generalization of the method just applied, that we can write

$$\langle L_3^{p} \rangle^{(1)} = \langle (L_3^2 - L_3 - L^2) [L_3^{p} - (L_3 - 1)^{p}] \rangle .$$
 (97)

[It is seen that Eqs. (91) and (95) are special cases of this relationship.] It is clear that we can write, generally,

$$\langle L_3^{(n)} \rangle = \langle P_{n+1}(L_3) \rangle , \qquad (98)$$

where  $P_{n+1}$  is a polynomial of the order n+1, and is obtained by the application of Eqs. (97) to each term of  $\langle P_n(L_3) \rangle$ . For the third derivative, we obtain, in this manner,

$$\langle L_3^{(3)} \rangle = 2 \langle 3L_3^4 - 10L_3^3 + (11 - 4L^2) L_3^2 + (8L^2 - 4) L_3 + L^2(L^2 - 4) \rangle .$$
 (99)

An iterative procedure is set up in Appendix C for obtaining the coefficients of  $P_{n+1}$  from those of  $P_n$ .

Of particular interest, for later purposes, is the initial condition in which the NO is in its highestenergy state. Since classically, this is the unstable equilibrium condition, we will refer to this initial condition also in the quantum-mechanical treatment as the "unstable equilibrium" (UE) initial condition. We list here, for later use, the first six initial derivatives for UE initial conditions obtained by the methods of Appendix C:

$$\langle L_{3}^{(1)}(0) \rangle = -2L_{0} ,$$

$$\langle L_{3}^{(2)}(0) \rangle = -2^{2}(L_{0}^{2} - L_{0}) ,$$

$$\langle L_{3}^{(3)}(0) \rangle = -2^{3}(L_{0}^{3} - 4L_{0}^{2} + 2L_{0}) ,$$

$$\langle L_{3}^{(4)}(0) \rangle = -2^{4}(L_{0}^{4} - 11L_{0}^{3} + 19L_{0}^{2} - 7L_{0}) ,$$

$$\langle L_{3}^{(5)}(0) \rangle = -2^{5}(L_{0}^{5} - 26L_{0}^{4} + 107L_{0}^{3} - 123L_{0}^{2} + 38L_{0}) ,$$

$$\langle L_{3}^{(6)}(0) \rangle = -2^{6}(L_{0}^{6} - 57L_{0}^{5} + 474L_{0}^{4} - 1195L_{0}^{3} + 1076L_{0}^{2} - 295L_{0}) .$$

Although generalized expressions for the numerical coefficients in these derivatives are not easily obtained, we assume the following generalizations: (i) The term with the highest power of  $L_0$  in  $\langle L_3^{(n)}(0) \rangle$  is  $-(2L_0)^n$ . (ii) The term with the next-highest power of  $L_0$  in  $\langle L_3^{(n)}(0) \rangle$  is  $(2^n - n - 1) \times 2^n L_0^{n-1}$ .

Returning to the quest for a differential equation

for  $\langle L_3 \rangle$ , we utilize a well-known identity for powers of  $L_3$ , namely, the expression of the fact that  $L_3$  satisfies its own eigenvalue equation. Thus,

$$L_3^2 - \frac{1}{4} = 0$$
 for  $L_0 = \frac{1}{2}$ , (101a)

$$(L_3^2 - 1) L_3 = 0$$
 for  $L_0 = 1$ , (101b)

$$(L_3^2 - \frac{9}{4}) (L_3^2 - \frac{1}{4}) = 0$$
 for  $L_0 = \frac{3}{2}$ , (101c)

and so on. For an n-level system, the eigenvalue equation reads

$$Q_n(L_3) = 0 , (102)$$

where  $Q_n(L_3)$  is the *n*th-order polynomial in  $L_3$ , the roots of which are the *n* eigenvalues of  $L_3$ .

Let us consider now an n-level system. The set of equations

$$\langle L_3^{(1)} \rangle = \langle P_2(L_3) \rangle ,$$

$$\langle L_3^{(2)} \rangle = \langle P_3(L_3) \rangle ,$$

$$\vdots$$

$$\langle L_3^{(n-1)} \rangle = \langle P_n(L_3) \rangle ,$$

$$(103)$$

and

 $\langle Q_n(L_3) \rangle = 0$ 

form a set of *n* equations from which we can eliminate the n-1 quantities  $\langle L_3^n \rangle$ ,  $\langle L_3^{n-1} \rangle$ , ...,  $\langle L_3^2 \rangle$ , the result being a linear differential equation (with constant coefficients) of order n-1 for the quantity  $\langle L_3 \rangle$ . One can then solve this equation by standard methods, subject to initial conditions which specify  $\langle L_3 \rangle$  and the first n-2 derivatives at  $\tau=0$ . The initial derivatives may be obtained from the initial moments by the first n-2 equations of the above set. (Any initial moment can be obtained from a knowledge of the initial state, in principle.)

It is worth noting that an explicit expression for  $\langle L_3(\tau) \rangle$  yields information about all moments of  $L_3$ , through Eqs. (103). Thus,  $\langle L_3^2(\tau) \rangle$  is obtained from the first equation if  $\langle L_3(\tau) \rangle$  and  $\langle L_3^{(1)}(\tau) \rangle$  are known,  $\langle L_3^3(\tau) \rangle$  is obtained from the second equation if  $\langle L_3(\tau) \rangle$ ,  $\langle L_3^2(\tau) \rangle$ , and  $\langle L_3^{(2)}(\tau) \rangle$  are known, and so on. Higher moments than the *n*th for an *n*-level system are reduced to lower moments by means of Eq. (102).

a. Small  $L_0$ . We illustrate the above method for several systems. For n=2  $(L_0=\frac{1}{2})$ , we have

$$\langle L_3^{(1)} \rangle = \langle L_3^2 - L_3 - \frac{3}{4} \rangle$$
, (104a)

$$\langle L_3^2 \rangle - \frac{1}{4} = 0 . \qquad (104b)$$

Eliminating  $\langle L_3^2 \rangle$ , we obtain

$$\langle L_3^{(1)} \rangle + \langle L_3 \rangle + \frac{1}{2} = 0 , \qquad (105)$$

the solution of which is

$$\langle L_3 \rangle = -\frac{1}{2} + (\langle L_3(0) \rangle + \frac{1}{2}) e^{-\tau}$$
 (106)

If the NO is initially in the upper state, then

$$\langle L_3(\tau) \rangle = -\frac{1}{2} + e^{-\tau} . \qquad (107)$$

The expression for  $\langle L_3^2 \rangle$  is given trivially by Eqs. (104b). Equation (106) is identical to Eq. (75c), which is to be expected.

For n=3 ( $L_0=1$ ), the set of Eqs. (103) becomes

$$\langle L_3^{(1)} \rangle = \langle L_3^2 - L_3 - 2 \rangle,$$
 (108a)

$$\langle L_3^{(2)} \rangle = 2 \langle L_3^3 - 2L_3^2 - L_3 + 2 \rangle$$
, (108b)

$$\langle L_3^3 - L_3 \rangle = 0 \quad . \tag{108c}$$

Elimination of  $\langle L_3^3 \rangle$  and  $\langle L_3^2 \rangle$  leads to

$$\langle L_3^{(2)} \rangle + 4 \langle L_3^{(1)} \rangle + 4 \langle L_3 \rangle + 4 = 0$$
 (109)

For the UE initial conditions,

$$\langle L_3(0) \rangle = \langle L_3^2(0) \rangle = 1 , \qquad (110)$$

so that, from Eq. (108a),

$$\langle L_3^{(1)}(0) \rangle = -2$$
, (111)

and the solution of Eq. (109) is

$$\langle L_3 \rangle = -1 + 2 e^{-2\tau} (\tau + 1) .$$
 (112)

We also obtain,

$$\langle L_3^2 \rangle = 1 - 2\tau \, e^{-2\tau} \, .$$
 (113)

Our last illustration is that for a four-level NO  $(n = 4, L_0 = \frac{3}{2})$ , for which we have the set of four equations

$$\langle L_3^{(1)} \rangle = \langle L_3^2 - L_3 - \frac{15}{4} \rangle$$
, (114a)

$$\langle L_3^{(2)} \rangle = 2 \langle L_3^3 - 2L_3^2 - \frac{11}{4}L_3 + \frac{15}{4} \rangle$$
, (114b)

$$\langle L_3^{(3)} \rangle = 2 \langle 3L_3^4 - 10L_3^3 - 4L_3^2 + 26L_3 - \frac{15}{4} \rangle$$
, (114c)

$$\langle L_3^4 - \frac{5}{2}L_3^2 + \frac{9}{16} \rangle = 0$$
 (114d)

Elimination of  $\langle L_3^4 \rangle$ ,  $\langle L_3^3 \rangle$ , and  $\langle L_3^2 \rangle$  leads to

$$\langle L_3^{(3)} \rangle + 10 \langle L_3^{(2)} \rangle + 33 \langle L_3^{(1)} \rangle + 36 \langle L_3 \rangle + 54 = 0$$
. (115)

Assuming the UE initial condition again, we have

$$\langle L_3^r(0) \rangle = \left(\frac{3}{2}\right)^r , \qquad (116)$$

which yields

$$\langle L_3^{(1)}(0) \rangle = \langle L_3^{(2)}(0) \rangle = -3$$
, (117)

and the solution of Eq. 
$$(115)$$
 is

$$\langle L_3 \rangle = -\frac{3}{2} + 3 e^{-3\tau} (4\tau - 1 + 2 e^{-\tau}).$$
 (118)

The application of the above method to cases of increasing  $L_0$  becomes tedious, of course, and if the NO is of macroscopic proportions, the application becomes prohibitive.<sup>21</sup> It is natural, therefore, to seek approximation methods that may be applicable to the case of large  $L_0$ .

b. Large  $L_0$ ; semi-quantum-mechanical approximation. We return to Eq. (91). Since  $\langle L_3^2 - L_3 \rangle \leq L^2$ , the slope of  $\langle L_3(\tau) \rangle$  is negative (which is consistent with the fact that the physical phenomenon is one of radiative decay). Also, since

$$\langle L_3^2 \rangle \ge \langle L_3 \rangle^2 ,$$
 (119)

the slope of  $\langle L_3 \rangle$  is less negative (less steep) than the slope of  $\hat{L}_3$ , where  $\hat{L}_3$  is a *c* number satisfying the differential equation

$$\hat{L}_{3}^{(1)} = \hat{L}_{3}^{2} - \hat{L}_{3} - L^{2} , \qquad (120)$$

and the same initial condition as  $\langle \, L_3 \, \rangle$  . We therefore have

$$\langle L_3 \rangle \ge \hat{L}_3$$
 (121)

The first-order nonlinear differential equation (120) can be solved simply in closed form, the result being

$$\hat{L}_3 = \frac{1}{2} - (L_0 + \frac{1}{2}) \tanh\left[ (L_0 + \frac{1}{2}) (\tau - \hat{\tau}_0) \right], \quad (122a)$$
 where

$$\hat{\tau}_0 = \frac{1}{L_0 + \frac{1}{2}} \tanh^{-1} \left( \frac{\hat{L}_3(0) - \frac{1}{2}}{L_0 + \frac{1}{2}} \right)$$
 (122b)

 $\hat{L}_3$  may be regarded as an approximation for  $\langle L_3 \rangle$  in which the difference between  $\langle L_3^2 \rangle$  and  $\langle L_3 \rangle^2$  is neglected. We shall refer to this approximation as the "semi-quantum-mechanical" (SQM) approximation. For initial conditions in which the NO is in an energy state,  $\hat{L}_3$  and  $\langle L_3 \rangle$  have the same initial derivative, and  $\hat{L}_3$  exhibits the correct (initial) spontaneous emission. In particular, setting  $L_3(0) = L_0$ , we have

$$\hat{\tau}_0 = \frac{1}{L_0 + \frac{1}{2}} \tanh^{-1} \left( \frac{1 - (2L_0)^{-1}}{1 + (2L_0)^{-1}} \right) \,. \tag{123}$$

As  $L_0$  becomes large, one can write, to lowest significant order in  $L_0^{-1}$ ,

$$\hat{\tau}_0 \approx L_0^{-1} \tanh^{-1} (1 - L_0^{-1}) \approx (2L_0)^{-1} \ln 2L_0$$
, (124)

and

$$\hat{L}_3 \approx -L_0 \tanh(L_0 \tau - \frac{1}{2} \ln 2L_0)$$
 . (125)

Comparison with Eq. (46) shows that  $L_3$ , for UE initial conditions, behaves like the classical  $L_3$  for slightly-off-UE initial conditions, with the initial energy given by  $L_3(0) = L_0 - 1$ .

It is clear that an improvement over the SQM approximation requires cognizance of the fact that  $\langle L_3^2 \rangle \neq \langle L_3 \rangle^2$ , in general, and must involve statistical considerations. We seek such an improvement by investigating the physical aspects of the difference between the quantum-mechanical and the classical problem.

c. Large  $L_0$ ; statistical approximation. For large  $L_0$ , the NO may be regarded as an essentially classical system except under two limiting conditions, when  $L_3$  is near  $L_0$  or  $-L_0$ . It is then that the uncertainty principle is significant, because, classically,  $l_1$  and  $l_2$ —or  $L_4$  and  $L_4$ —vanish for  $L_3 = \pm L_0$ , while according to the uncertainty principle,  $l_1$  and  $l_2$  cannot vanish, since these variables do not commute with  $L_3$ . If we consider an ensemble of identical NO's, with  $L_3 = \pm L_0$ , then  $l_1$ and  $l_2$ , according to the uncertainty principle, must be considered random variables (random with respect to members of the ensemble) for which the ensemble average of the sum of their squares is given by the corresponding quantum-mechanical expectation value,

$$\langle l_1^2 + l_2^2 \rangle = L^2 - \langle L_3^2 \rangle = L_0$$
 (126)

These "quantum-mechanical" coordinates of oscillation—which may be considered as remaining approximately constant in the neighborhood of the two energy extremes—are significant only when they are not much smaller than the classical coordinates of oscillation, given by

$$l_1^2 + l_2^2 = L_0^2 - L_3^2 . (127)$$

It is, therefore, reasonable to regard the problem as essentially quantum mechanical (in the sense of nonclassical) only when the "quantum-mechanical" amplitudes (or "uncertainty-principle" amplitudes) are of the order of, or greater than, the classical amplitudes, or when the condition

$$L_0 \gtrsim L_0^2 - L_3^2 \tag{128}$$

holds. For large  $L_0$ , this condition can be written, approximately,

$$L_0 - \left| L_3 \right| \lesssim 1 . \tag{129}$$

(The symbol " $\leq$ " is to be read "less than, or of the order of.") The quantum-mechanical aspects of the field should also be taken into consideration, of course. It can be shown, in fact, that the zero-point oscillation of the field (or, more precisely, a quantum-mechanical treatment of the field when it is in the lowest-energy state) doubles the radia-tion effect of the "highest-point" oscillation of the NO, and cancels the radiation effect of the zero-point oscillation of the NO.<sup>22</sup> We can therefore drop the absolute-value sign in Eq. (129). In conclusion, it can be said that a quantum-mechanical treatment is necessary if, and only if, the NO is near UE, or

$$L_0 - L_3 \stackrel{<}{\scriptstyle\sim} 1 \ . \tag{130}$$

The above reasoning is consistent with the classical and quantum-mechanical expressions for the expectation value of the power radiated into the field by the NO. For the free decay of the NO, we can write, from Eqs. (31c) and (37c),

$$\langle \dot{L}_3 \rangle = -2\alpha_1 \langle L_* L_- \rangle$$
  
=  $-\alpha_1 \langle (L_0 + L_3) (L_0 - L_3) + \lambda (L_0 + L_3) \rangle , \qquad (131)$   
where

$$\lambda = \begin{cases} 0 & \text{classically} \\ 1 & \text{quantum mechanically} \end{cases}$$
(132)

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Consider a classical *statistical* description of the decay (necessary, for instance, if the initial conditions are specified statistically, with respect to an ensemble). The expectation-value brackets are then entirely appropriate in the classical description, and indicate an ensemble average. Furthermore, classically, also, we have  $\langle L_3^2 \rangle \neq \langle L_3 \rangle^2$ , in general. On the basis of Eqs. (131) and (132), it is reasonable to regard the behavior of the system as describable classically (but statistically, if necessary) when the  $\lambda$  term (with  $\lambda = 1$ ) is insignificant compared to the first term, and requiring a quantum-mechanical description otherwise. We thus have

$$L_0 - L_3 \gg 1$$
, (133)

as the condition which allows a classical description. This inequality is consistent with that of (130). It is worth noting that, although Eq. (131) contains explicitly only NO variables, the quantummechanical effects of the field are already built into it, since the order of the NO variables  $L_*$  and  $L_$ was determined, in part, by the quantum-mechanical properties of the field.

Obviously, the "most quantum-mechanical" problem-and, therefore, the most interesting problem, for present purposes-is that for UE initial conditions. We proceed to consider this problem for large  $L_0$ . In accordance with the above discussion, the quantum-mechanical solution need be carried only up to a time when the energy radiated is large compared to unity (in units of  $\hbar \omega$ ), beyond which the behavior of the system is describable classically. Let that time be given by  $\tau = \tau_1$ . More precisely, since the quantum-mechanical solution offers only a statistical description, we take  $\tau_1$ to be the time when the average energy radiated is large compared to unity. Our method of approximation will be the following: (a) For  $0 \le \tau \le \tau_1$ (which will be referred to as the "quantum-mechanical region"), the problem will be treated quantum mechanically, with UE initial conditions; (b) for  $\tau_1 \leq \tau$  (which will be referred to as the "classical region"), the problem will be treated classically; the results of the quantum-mechanical solution will be considered initial conditions, and the statistical properties introduced by the quantum-mechanical solution will be preserved. We note that this method of approximation explicitly avoids the assumption  $\langle L_3^2 \rangle = \langle L_3 \rangle^2$ . We also note that a small statistical spread in initial conditions near unstable equilibrium will have a significant effect on the classical solution, as can be seen from Eqs. (44)-(46). The result of this method will be referred to as the "statistical approximation."

Let  $\langle L_3 \rangle$  be expanded in a Taylor series at  $\tau = 0$ ,

$$\langle L_3(\tau) \rangle = L_0 + \sum_{n=1}^{\infty} \langle L_3^{(n)}(0) \rangle \frac{\tau^n}{n!}$$
 (134)

Taking note of Eq. (100) and the two following generalizations, we can write

$$\langle L_3(\tau) \rangle = L_0 + f_0 (L_0 \tau) + L_0^{-1} f_1 (L_0 \tau) + \cdots$$
, (135a)

where

$$f_0(L_0\tau) = -\sum_{n=1}^{\infty} \frac{(2L_0\tau)^n}{n!} = 1 - e^{2L_0\tau}$$
(135b)

and

<

$$f_1(L_0\tau) = \sum_{n=1}^{\infty} (2^n - n - 1)(2L_0\tau)^n$$
$$= e^{2L_0\tau} (e^{2L_0\tau} - 2L_0\tau - 1) . \qquad (135c)$$

For  $L_0$  sufficiently large and  $\tau$  sufficiently small, or more precisely, for the condition

$$L_0 \gg e^{2L_0 \tau}$$
, (136)

 $\langle L_3(\tau) \rangle$  can be approximated by the relationship

$$L_{3}(\tau) \rangle \approx L_{0} + (1 - e^{2L_{0}\tau}) + L_{0}^{-1} e^{2L_{0}\tau} (e^{2L_{0}\tau} - 2L_{0}\tau - 1) . \quad (137)$$

In accordance with the previous discussion, we need to treat the problem quantum mechanically only up to  $\tau = \tau_1$ , where  $|\langle L_3(\tau_1) \rangle - L_0|$  is significantly larger than unity. The requirement on  $\tau_1$  is thus

$$L_0 \gg e^{2L_0 \tau_1} \gg 1. \tag{138}$$

It is convenient to consider

$$\tau_1 \sim L_0^{-1}$$
 (139)

We will now use Eq. (137) to determine the initial conditions for the problem in the classical region. As mentioned previously, the statistical properties of  $L_3(\tau)$ , expressed by the moments of  $L_3(\tau)$ , can be determined from the explicit expression of  $\langle L_3(\tau) \rangle$  as a function of  $\tau$ . [The moments can be determined if the derivatives are known, as is illustrated by Eqs. (91), (96), and (99). ] In order to facilitate the following discussion, we will label orders of magnitude in terms of powers of  $L_0$ , with  $e^{2L_0\tau}$  being considered of the order of  $L_0^0$ , that is, unity. Thus, the three terms in Eq. (137) are of the orders of  $L_0$ , unity, and  $L_0^{-1}$ , respectively. In the classical region, the problem will be treated with the approximation in which only terms down to order unity are considered. In other words, terms of the order of  $L_0^{-1}$  or smaller will be neglected. Furthermore, the statistical properties will be specified only to the extent of an explicit expression for the first and second moments and the sign of the third moment. It will become obvious that these approximations are dictated by the fact that  $\langle L_3(\tau) \rangle$  is given explicitly in Eq. (137) only to order  $L_0^{-1}$ .

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The initial conditions for the classical problem can now be written as

$$L_3(\tau_1) = L_0 + (1 - e^{2L_0 \tau_1}) + D , \qquad (140)$$

where D is a random variable. Some of its statistical properties are given by

$$\langle D \rangle = 0 \tag{141}$$

and

$$\langle D^2 \rangle = \langle L_3^2(\tau_1) \rangle - \langle L_3(\tau_1) \rangle^2 , \qquad (142)$$

with the right-hand side to be obtained from the quantum-mechanical solution. Using the quantummechanical relationship

$$\langle L_3^2 \rangle = \langle L_3^{(1)} \rangle + \langle L_3 \rangle + L_0(L_0 + 1)$$
(143)

and Eq. (137), one obtains

$$\langle D^2 \rangle = e^{2L_0 \tau_1} (e^{2L_0 \tau_1} - 1) .$$
 (144)

It is seen that  $\langle D^2 \rangle$  is of order unity. One can show, from Eqs. (100), that  $\langle D^3 \rangle$  is also of order unity and is negative. The derivation of a closedform expression for  $\langle D^3 \rangle$  by the above method, however, would require  $\langle L_3^{(2)} \rangle$  to order unity, and therefore  $\langle L_3 \rangle$  to order  $L_0^{(2)}$ , which we do not have in closed form. The rms deviation of D (and of the initial  $L_3$  in the classical region) is given by

$$\langle D^2 \rangle^{1/2} = e^{2L_0 \tau_1} (1 - e^{-2L_0 \tau_1})^{1/2} .$$
 (145)

Now, the maximum (positive) value that D may assume, since the initial  $L_3$  in the classical region cannot be greated than  $L_0$ , is

$$D_{\max} = e^{2L_0 \tau_1} (1 - e^{-2L_0 \tau_1}) . \tag{146}$$

This quantity is slightly smaller than the rms deviation; hence the negative value of  $\langle D^3 \rangle$ .

The classical solution for  $\tau \ge \tau_1$  is, from Eqs. (42),

$$L_{3}(\tau) = -L_{0} \tanh L_{0}(\tau - \tau_{0}) , \qquad (147a)$$

where

$$\tau_0 = \frac{1}{L_0} \tanh^{-1} \left( \frac{L_3(\tau_1)}{L_0} \right) + \tau_1 .$$
 (147b)

Noting that

we have, approximately,

$$\tanh^{-1}\left(\frac{L_{3}(\tau_{1})}{L_{0}}\right) \approx \frac{1}{2}\ln\left(\frac{2L_{0}}{e^{2L_{0}\tau_{1}}-1-D}\right)$$
$$= \frac{1}{2}\ln\left(\frac{2L_{0}e^{-2L_{0}\tau_{1}}}{1-(1+D)e^{-2L_{0}\tau_{1}}}\right). \quad (149)$$

The strong formal dependence on  $\tau_1$  cancels, and Eqs. (147) yield

$$L_{3}(\tau) = -L_{0} \tanh\{L_{0}\tau - \frac{1}{2}\ln 2L_{0} + \ln[1 - (1+D)e^{-2L_{0}\tau_{1}}]\}$$

for  $\tau \ge \tau_1$ . (150)

Equation (150) is the "statistical approximation"; it differs from the SQM approximation by the presence of the last term (the statistical term) in the argument of the hyperbolic tangent. The statistical properties of D that have been derived are contained in Eqs. (141), (144) or (145), and (146). The fact that  $\langle D^3 \rangle$  is of order unity and is negative has also been mentioned. In principle, the statistical information about D can be increased to any required amount by expanding  $\langle L_3(\tau) \rangle$  in Eq. (134) to a sufficient degree of accuracy.

The dependence of  $L_3(\tau)$ , as given by Eq. (150), on the exact value of  $\tau_1$  is less significant than may appear at first glance, since the  $\tau_1$  dependence of D mainly cancels the effect of the exponential containing  $\tau_1$ . We illustrate this point by making an approximate calculation of the average time of decay of the NO energy to zero, that is, to half its initial value. For any particular member of the ensemble, this decay time  $\tau_d$  is given by setting the argument of the hyperbolic tangent in Eq. (150) equal to zero. This yields

$$L_0 \tau_d = \frac{1}{2} \ln 2L_0 - \ln [1 - (1+D)e^{-2L_0 \tau_1}].$$
 (151)

The average of  $\tau_d$  requires an average of the logarithmic term. Expanding formally the logarithm about zero, and retaining only the first two terms, we have

$$\langle \ln[1+(1+D)e^{-2L_0\tau_1}]\rangle$$
  
  $\approx -\langle (1+D)e^{-2L_0\tau_1}+\frac{1}{2}(1+D)^2e^{-4L_0\tau_1}\rangle.$  (152)

It is possible to utilize, now, the expressions for the first and second moments of D, to obtain, for the average of the logarithm,

$$-\frac{1}{2}(1+e^{-2L_0\tau_1}+e^{-4L_0\tau_1})\approx -\frac{1}{2}, \qquad (153)$$

the approximation being based on the inequality (138). We thus obtain

$$\langle \tau_d \rangle = (2L_0)^{-1} (\ln 2L_0 + 1)$$
 (154)

It is not the purpose of the present treatment to investigate the statistical properties of  $L_3$  in detail, but only to illustrate the above method. Some qualitative observations can be made easily, however, and are instructive. Since the second logarithm in Eq. (150) approaches  $-\infty$  as D approaches its maximum value  $e^{2L_0\tau_1} - 1$ , while this logarithm is finite for all other values of D (and the spread in D is of the order of magnitude of unity), it is apparent that the average effect of this logarithm on the argument of the hyperbolic tangent will be negative, and its effect on  $L_3(\tau)$  will therefore be positive. Comparing Eq. (150) with Eq. (125), that is, the statistical approximation with the SQM approximation, we see explicitly that the former gives a larger value of  $\langle L_3(\tau) \rangle$  than the latter, a result consistent with the previous discussion of the error contained in the SQM approximation. For instance, for  $\tau = (1/2L_0) \ln 2L_0$ , which yields  $\hat{L}_3 = 0$ , the average of  $L_3$  over the three values D = 0,  $\pm \frac{1}{2} \langle D^2 \rangle^{1/2}$  is approximately 0.07 $L_0$ .

It is also interesting to comment on a disagreement in the literature (involving numerical calculations) concerning the maximum power radiated by the NO.  $^{13-15}$  From Eq. (150), we obtain

$$-L_{3}^{(1)}(\tau) = L_{0}^{2} \operatorname{sech}^{2} \left\{ L_{0} \tau - \frac{1}{2} \ln 2L_{0} + \ln \left[ 1 - (1+D) e^{-2L_{0} \tau_{1}} \right] \right\}, \quad (155)$$

which is the radiated power, in units of  $\alpha \hbar \omega$ . We see that for any one member of the ensemble under consideration (any specific value of the random variable D), the maximum power radiated is  $L_0^2$ . Therefore, the average of the maximum power radiated is  $L_0^2$ . However, the maximum of the average power radiated is another matter. At any given time  $\tau$ , an average over  $|L_3^{(1)}(\tau)|$  yields a value less than  $L_0^2$ , because the argument of the hyperbolic secant has a statistical spread that reaches out to  $-\infty$  (thus giving a range of  $|L_3^{(1)}|$ from  $L_0^2$  down to zero). One must, therefore, carefully define the meaning, in statistical terms, of "the maximum power radiated." For instance, the average of  $|L_3^{(1)}|$  obtained analogously to the average of  $L_3$  in the last statement of the preceding paragraph is  $0.83L_0^2$ .

### 2. Forced Oscillation

In the analysis of the driven multilevel NO, the same approximation will be made in the equations of motion as that which was made in the analysis of the free decay, namely, the neglect of the frequency-shift terms. We merely have to supplement Eqs. (87) and (88) by the driving terms contained in Eqs. (16), to obtain

 $\dot{L}_{+} = A * L_{3} + \alpha^{\dagger} L_{3}$ , (156a)

$$\dot{L}_{-} = A L_{3} + L_{3} \alpha$$
, (156b)

$$L_{3} = -(AL_{+} + A^{*}L_{-}) - (L_{+}\alpha + \alpha^{\dagger}L_{-}), \qquad (156c)$$

$$\alpha = \alpha_0 + \alpha L_{\perp}, \quad \alpha^{\dagger} = \alpha_0^{\dagger} + \alpha L_{\perp}. \quad (156d)$$

Since the classical analysis was carried out on the basis of the same approximation, it is instructive to write the following equations (unless otherwise indicated) in a manner that displays explicitly the corresponding classical equations. This will facilitate a physically meaningful comparison between the classical and quantum-mechanical results. We make use again of the factor  $\lambda$ , which, as indicated in Eq. (132), is zero in the classical interpretation and unity in the quantum-mechanical interpretation of the equations.

The present equations of motion yield

$$\langle \dot{L}_{3} \rangle = - \langle A \langle L_{*} \rangle + A^{*} \langle L_{-} \rangle) - 2\alpha \langle L_{*} L_{-} \rangle , \qquad (157)$$

$$\frac{1}{\langle L_3 \rangle} = -\Omega^2 \langle L_3 \rangle - 3\alpha \langle AL_*L_3 + A^*L_3L_- \rangle$$

$$-4\alpha^2 \langle L_{\star}L_3L_{\star}\rangle$$
, (158)

where, it is recalled,  $\Omega^2 = 2|A|^2$ . Utilizing the relationships

$$L_{+}L_{3} = \frac{1}{2}(\{L_{+}, L_{3}\} - \lambda L_{+}), \qquad (159a)$$

$$L_{3}L_{-} = \frac{1}{2}(\{L_{3}, L_{-}\} - \lambda L_{-}), \qquad (159b)$$

$$L_{+}L_{-} = \frac{1}{2} \left[ L_{0}^{2} - L_{3}^{2} + \lambda (L_{0} + L_{3}) \right], \qquad (159c)$$

where the notation  $\{X, Y\} \equiv XY + YX$  is used, we obtain

$$\langle \dot{L}_3 \rangle + \frac{3}{2} \alpha \lambda \langle \dot{L}_3 \rangle + \Omega^2 \langle L_3 \rangle - \frac{3}{2} \alpha \langle \{ \dot{L}_3, L_3 \} \rangle$$

$$= \alpha^2 \langle [L_0^2 - L_3^2 + \lambda (L_0 + L_3)] (L_3 - \lambda) \rangle .$$
(160)

It is seen immediately that, for  $\lambda = 0$ , this equation (without the expectation-value brackets) reduces to Eq. (56), the classical equation. It may also be noted that if the NO is a two-level system, this equation reduces to the corresponding Eq. (79) by use of the applicable relationships

$$\{L_{+}, L_{3}\} = \{L_{-}, L_{3}\} = \{\dot{L}_{3}, L_{3}\} = 0, \quad L_{3}^{2} = \frac{1}{4}, \quad \lambda = 1.$$

As in the classical (and two-level ) analysis of the driven oscillator, we consider the problem for which the NO is initially in the ground state:

$$\langle L_3(0)\rangle = -L_0 . \tag{161}$$

The striking difference between the classical equation (56) and the quantum-mechanical equation (160) is the presence of the  $\langle \dot{L}_3 \rangle$  term in the latter. We have seen in the classical analysis that  $L_3$  will oscillate periodically between  $-L_0$  and  $L_0$  for a sufficiently strong driving field. In the present instance, however, such an oscillation of  $\langle L_3 \rangle$  appears impossible, since-as is well known-the  $\langle L_3 \rangle$  term is a damping term. The interest of the qualitative difference in the equations of motion is enhanced by the consideration that, from a physical viewpoint, a strongly driven NO of large  $L_0$  may be expected to behave, essentially, as a classical system, since the amplitudes of oscillation of the NO and of the force acting on the NO are large compared to the (quantum-mechanical) uncertainties in these amplitudes.<sup>23</sup> In the case of a weak driving field, for which, classically,  $L_3$  approaches a constant value, we may expect the effect of the  $\langle L_3 \rangle$  term to be less significant. We will therefore consider, in the present analysis, only the case of a strong driving field, which, as in the classical analysis, satisfies the inequality

$$\Omega \gg \alpha L_0 \quad . \tag{162}$$

Approximating Eq. (160) by dropping the  $\alpha^2$  term,

we obtain

$$\langle L_3 \rangle + \frac{3}{2} \alpha \lambda \langle \dot{L}_3 \rangle + \Omega^2 \langle L_3 \rangle = \frac{3}{2} \alpha \langle \{ \dot{L}_3, L_3 \} \rangle . \quad (163)$$

This equation (with  $\lambda = 1$ ) differs from the corresponding classical equation for a strongly driven NO, Eq. (68), in two respects essentially: the presence of the damping term, and the fact that  $\langle \{L_3, L_3\} \rangle$  is not equal to  $\{\langle L_3 \rangle, \langle L_3 \rangle\}$ , in general. If we approximate by ignoring the latter difference, Eq. (163) becomes a nonlinear differential equation for  $\langle L_3 \rangle$  that differs from the corresponding classical equation only by the presence of the damping term. An analysis of this equation by means of phase-plane trajectories, somewhat similar to the analysis of the corresponding classical equation,<sup>24</sup> shows that  $\langle L_3 \rangle$  executes a damped oscillation in time, and approaches zero. For the twolevel NO, the right-hand side of Eq. (163) vanishes, and the solution is an exponentially damped sinusoidal oscillation, a result consistent with that previously obtained.

We can also analyze Eq. (163) by a perturbationtheory approach in which it is not necessary to approximate explicitly the right-hand side by a product of expectation values. In this analysis, one considers the coupling to the radiation field (but not to the driving field) to be a small perturbation. The zeroth-order *operator* solution of Eqs. (156) is then obtained immediately (by setting  $\alpha = 0$ ) as

$$L_{3}^{(0)} = L_{3}(0) \cos\Omega t + \Omega^{-1} [AL_{*}(0) + A^{*}L_{-}(0)] \sin\Omega t .$$
(164)

Equation (163) is formally satisfied by

$$\langle L_3(t) \rangle = \langle L_3^{(0)}(t) \rangle + \frac{3}{2} \frac{\alpha}{\Omega} \int_0^t dt_1 \sin\Omega(t-t_1) \\ \times \frac{d}{dt_1} \langle L_3^2(t_1) - \lambda L_3(t_1) \rangle .$$
 (165)

In order to obtain  $\langle L_3(t) \rangle$  to order  $\alpha$ , we need only the lowest- (zeroth-) order contribution of the integrand. For t such that

$$L_0 \Omega^{-1} \ll t \ll \alpha^{-1} , \qquad (166)$$

we obtain, thus, to order  $\alpha$ ,

$$\langle L_{\mathbf{3}}(t) \rangle \approx \left(1 - \frac{3}{4} \lambda \alpha t\right) \langle L_{\mathbf{3}}^{(0)}(t) \rangle . \tag{167}$$

This perturbation-theory expression is consistent with a long-time quantum-mechanical solution containing a damping factor  $e^{-3\alpha t/4}$ , the same damping factor that is found in the solution for the two-level NO. (Classically, this expression shows that there exists no damping at least to order  $\alpha$ .) Thus, although we have not obtained an exact solution for  $\langle L_3(t) \rangle$  in the case of a NO with an arbitrary number of levels, we can reasonably conclude that the behavior of  $\langle L_3(t) \rangle$  is described quantum mechanically by a damped oscillation, and differs qualitatively from that of the classical  $L_3(t)$ , for which the oscillations are undamped.

The explanation of this difference-which seems paradoxical, in view of the fact that it exists even under conditions for which the system may be regarded as essentially classical-must be sought (as in I) in the statistical aspects of the quantum-mechanical description. While an individual member of a quantum-mechanical ensemble may behave in a manner only slightly different from that of a classical system, the differences among the members (these differences are the "quantum fluctuations") may add in such a way as to make the average behavior appear greatly different from the classical behavior. In the present instance, the most obvious aspect of the behavior in which the differences can account for such an effect is the frequency of oscillations of  $L_3$ . Either a slight spread in (constant) frequencies among members of the ensemble, or a slight random variation of frequency with respect to time for each member of the ensemble, can result in a damped oscillation of the average, even though the oscillation of each member is undamped.

Of course, one could argue that classical considerations are of little relevance, and that each member of the ensemble-or, at least, a *typical* member-being a large system, behaves in a manner described by the expectation value, the energy being a damped oscillation specified by  $\langle L_3 \rangle$ . It should be possible to distinguish between the two types of ensembles, which are associated, respectively, with the two conflicting arguments, by an examination of  $\langle L_3^2 \rangle$ . If the individual members of the ensemble are damped (in the sense that  $L_3$  approaches zero), then  $\langle L_3^2 \rangle$  will likewise be damped, while a spread in frequencies of an ensemble of undamped oscillations will not lead to a damping of  $\langle L_3^2 \rangle$ .

We use perturbation theory to examine  $\langle L_3^2 \rangle$ . The coupling constant between the NO and the radiation field will be considered a quantity of first order, with  $\alpha$ , therefore, being a quantity of second order. The perturbation-theory orders will be indicated by a parenthetical superscript (which should not be confused with previous similar notation indicating differentiation with respect to  $\tau$ ; no such differentiation is used in the present section). We seek results up to second order. (Note, however, that the driving field is taken into account to all orders.) Equations (156) lead, by suitable substitution in the expression for  $L_3$ , to the operator equation

$$\dot{L}_{3} = - \left[ A L_{*}(0) + A^{*}L_{-}(0) \right] - \Omega^{2} \int_{0}^{t} dt_{1} L_{3}(t_{1}) \\ - \int_{0}^{t} dt_{1} \left[ A \alpha_{0}^{\dagger}(t_{1}) L_{3}(t_{1}) + A^{*}L_{3}(t_{1}) \alpha_{0}(t_{1}) \right] \\ \cdot - \alpha \int_{0}^{t} dt_{1} \left[ A L_{*}(t_{1}) L_{3}(t_{1}) + A^{*}L_{3}(t_{1}) L_{-}(t_{1}) \right]$$

This equation is formally satisfied by the relationship

$$L_{3} = L_{3}^{(0)} + \Omega^{-1} \int_{0}^{t} dt_{1} \sin \Omega (t - t_{1}) Z(t_{1}) + Kt , \quad (169a)$$

where

$$Z = - \left[ A \alpha_0^{\dagger} L_3 + A^* L_3 \alpha_0 \right] - \alpha \left[ A L_+ L_3 + A^* L_3 L_- \right] \\ - \frac{d}{dt} \left( L_+ \alpha_0 + \alpha_0^{\dagger} L_- \right) - 2\alpha \frac{d}{dt} \left( L_+ L_- \right) \quad (169b)$$

and

$$K = - \left[ L_{+}(0) \alpha_{0}(0) + \alpha_{0}^{\dagger}(0) L_{-}(0) + 2\alpha L_{+}(0) L_{-}(0) \right].$$
(169c)

[The simplest way to see that Eqs. (169) satisfy Eq. (168) is to note that both equations yield the same  $\ddot{L}_3(t)$  and  $\dot{L}_3(0)$ . Incidentially, these equations are valid both classically and quantum mechanically.]

Writing

$$L_{3} = L_{3}^{(0)} + L_{3}^{(1)} + L_{3}^{(2)} , \qquad (170)$$

we proceed to calculate the expression

$$\langle L_3^2 \rangle = \langle L_3^{(0)2} \rangle + \langle \{L_3^{(0)}, \ L_3^{(1)} \} \rangle + \langle L_3^{(1)2} \rangle + \langle \{L_3^{(0)}, \ L_3^{(2)} \} \rangle .$$
 (171)

The details of the calculation, utilizing previously used approximations, with t satisfying inequality (166), are outlined in Appendix D. The results are (in the quantum-mechanical analysis only)

$$\langle L_3^{(0)2} \rangle = L_0^2 \cos^2 \Omega t + \frac{1}{2} L_0 \sin^2 \Omega t , \qquad (172a)$$

$$\langle \{L_3^{(0)}, L_3^{(1)}\} \rangle = 0$$
, (172b)

$$\langle L_{3}^{(1)2} \rangle = \frac{1}{4} \alpha t L_{0} \left[ (4L_{0} + 1) \sin^{2}\Omega t + 3 \cos^{2}\Omega t \right],$$
(172c)

$$\langle \{L_{3, \cdot}^{(0)}, L_{3, \cdot}^{(2)}\} \rangle = -\frac{3}{2} \alpha t L_0 [L_0 \cos^2 \Omega t + \frac{1}{2} \sin^2 \Omega t],$$
(172d)

so that, up to second order,

$$\langle L_3^2 \rangle = \left[ L_0^2 - \frac{3}{2} \alpha t L_0 \left( L_0 - \frac{1}{2} \right) \right] \cos^2 \Omega t$$
  
+  $\left[ \frac{1}{2} L_0 + \alpha t L_0 \left( L_0 - \frac{1}{2} \right) \right] \sin^2 \Omega t .$  (173)

To the same accuracy we have, from Eq. (167),

$$\langle L_3 \rangle^2 = L_0^2 \left( 1 - \frac{3}{2} \alpha t \right) \cos^2 \Omega t.$$
 (174)

As a check, we note that for  $L_0 = \frac{1}{2}$ , Eq. (173) yields  $\langle L_3^2 \rangle = \frac{1}{4}$ , the exact result.

Equation (173) serves as a qualitative corroboration of our physical interpretation of the quantummechanical damping. If we were averaging over an ensemble of (say, exponentially) damped oscillations, all of frequency  $\Omega$ , then there could not exist in the expression for  $\langle L_3^2 \rangle$  a  $\sin^2 \Omega t$  component that increases with time, the increase being of the same order of magnitude as the decrease in the  $\cos^2 \Omega t$  component. However, such a component is entirely consistent with the assumption that the ensemble consists of undamped oscillations that have a random frequency spread.

It should be possible to explain, in a qualitative manner, the physical origin of this random frequency spread by means of the uncertainty principle. We continue to use a perturbation-theory viewpoint. In lowest order, the frequency of oscillation of  $L_3$  is determined by the externally applied field only (and, indeed, in lowest order there exists no damping). In higher order, the NO sees not only the external field but also the radiation field, and the frequency is now determined by the superposition of the two fields. Classically, the radiation field is well defined; it is the radiated field, determined by the oscillating components  $l_{\perp}$  and  $l_{\perp}$  of the angular-momentum vector, or, in terms of reduced variables, by  $L_{\star}$  and  $L_{...}$  Quantum mechanically, however, the situation is different. Consider, for simplicity, the time t = 0, at which both the NO energy and the field energy have their well-defined minima, respectively. There will now be an uncertainty in  $L_{1}(0)$  and  $L_{2}(0)$ , as well as in  $\alpha_{0}(0)$  and  $\alpha_{0}^{\dagger}(0)$ . since these dynamical variables do not commute with the energy variables of the NO and the field, respectively. The uncertainty in  $L_{+}$  and  $L_{-}$  will produce an uncertainty in the radiated field. The effective field acting on the NO-in perturbationtheory language-is the superposition of the driving field, the radiated field, and the zero-point field, the resultant being determined by both the magnitudes and relative phases of these three fields. There exists, thus, an uncertainty in the net, or effective, field, which will produce an uncertainty (or a spread among members of the quantummechanical ensemble) in the frequency of oscillation of  $L_3$ .

This explanation may be presented in a more formal manner. Let us replace  $L_{\star}$  and  $L_{-}$ , in Eq. (168), according to the approximation

$$L_{*}(t) \approx L_{*}(0) + A^{*} \int_{0}^{t} dt_{1} L_{3}(t_{1}) ,$$
 (175a)

$$L_{-}(t) \approx L_{-}(0) + A_{0}^{t} dt_{1} L_{3}(t_{1})$$
 (175b)

This substitution yields

$$\dot{L}_{3} = -\left[AL_{*}(0) + A^{*}L_{-}(0)\right] - \left[L_{*}(0)\hat{\alpha}_{0} + \hat{\alpha}_{0}^{*}L_{-}(0)\right]$$
$$- 2\alpha L_{*}(0)L_{-}(0) - \frac{3}{2}\alpha\Omega^{2}\left[\int_{0}^{t}dt_{1}L_{3}(t_{1})\right]^{2}$$
$$- \int_{0}^{t}dt_{1}O[L_{3}(t_{1})], \quad (176a)$$

where O is a linear operator that operates on  $L_3(t_1)$ ; it is given by

$$O = \Omega^{2} + 3\alpha \{ [AL_{*}(0)]_{I} + [A^{*}L_{-}(0)]_{r} \}$$
  
+  $[A \mathfrak{a}_{0}^{\dagger}(t_{1})]_{I} + [A^{*}\mathfrak{a}_{0}(t_{1})]_{r} + [A \mathfrak{a}_{0}^{\dagger}(t)]_{I}$   
+  $[A^{*}\mathfrak{a}_{0}(t)]_{r}, (176b)$ 

the subscripts "l" and "r" indicating that the corresponding terms are placed on the left- and on the right-hand sides of  $L_3(t_1)$ , respectively. In lowest approximation, this operator determines the frequency of oscillation of  $L_3(t)$  by specifying the square of the frequency. Considering, now, the dynamical variables to be random c numbers, we can regard this operator as the square of the effective frequency, which exhibits, explicitly, its dependence on the magnitudes of the random quantities  $L_{+}(0)$ ,  $L_{-}(0)$ ,  $\alpha_{0}$ ,  $\alpha_{0}^{\dagger}$ , and on their respective phase relationships with the driving field. These quantities, for the present initial conditions, vanish classically, <sup>25</sup> but constitute the uncertainties in the oscillation amplitudes-the zero-point oscillations-when the uncertainty principle is taken into account, and produce an uncertainty in the effective frequency of oscillation of  $L_3$ . We will not carry this formal analysis further, since one should not regard zero-point oscillations too literally in a classical sense. As was pointed out in an earlier paper,<sup>22</sup> such a course can lead to absurdities if taken too far. It does, however, indicate the formal (quantum-mechanical) origin of the frequency spread among the ensemble members.

We see, in the present instance, that the calculated quantum-mechanical expectation value cannot be considered even qualitatively illustrative of the results of an experiment, even though the system under consideration may be essentially macroscopic. In fact, some qualitative features of a single experiment (and in the macroscopic domain, one is interested in results of single experiments) are better described by the classical solution than by the expression for the quantum-mechanical expectation value. The greatest insight into the class of phenomena of the present type-one that includes a number of quantum-electronic processes-is, obviously, obtained by an analysis of both descriptions, as illustrated not only by the consideration of the forced oscillations, but also by that of the free decay.

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### APPENDIX A

Let the field under consideration be the electromagnetic field. It may be described in terms of the modes by

$$\vec{\mathbf{E}} = -\sum_{k} \left(4\pi\hbar\omega_{k}\right)^{1/2} \vec{\mathbf{u}}_{k}(\vec{\mathbf{r}}) p_{k}(t) , \qquad (A1)$$

$$\vec{\mathbf{H}} = \sum_{k} \left( 4\pi c^{2} \hbar / \omega_{k} \right)^{1/2} \vec{\nabla} \times \vec{\mathbf{u}}_{k}(\vec{\mathbf{r}}) q_{k}(t) , \qquad (A2)$$

where Gaussian units are used,  $q_k$  and  $p_k$  are dimensionless dynamical variables satisfying Eqs. (1) and (2), and  $\mathbf{\bar{u}}_k(\mathbf{\bar{r}})$  satisfies the equation  $\nabla^2 \mathbf{\bar{u}}_{k+} (\omega_k^2/c^2) \mathbf{\bar{u}}_k = 0$ , and is normalized over a sufficiently large volume. The same volume is used to impose boundary conditions on  $\mathbf{\bar{u}}$  that make the modes denumerable. This description corresponds to a standing wave for each mode; traveling waves can be obtained by a suitable superposition of standing waves. The decomposition into modes is not unique, of course.

If the NO is coupled to the field through a dipole moment, this moment can be described by  $^{26}$ 

$$\mathbf{d} = \mu (\mathbf{a}_1 l_1 + \mathbf{a}_2 l_2 + \mathbf{a}_3 l_3),$$

where the  $\vec{a}$ 's are dimensionless vectors of the order of unity and  $\mu$  is the dipole-moment strength, either magnetic or electric. (For a magnetic moment due to spin or orbital angular momentum, the  $\vec{a}$ 's are orthonormal vectors.) Setting the interaction energy to be either  $-\vec{d} \cdot \vec{H}$  or  $-\vec{d} \cdot \vec{E}$ , depending on whether the coupling to the field takes place through the q or the p coordinate, the coupling constants used in Eq. (5) are given by

$$\gamma_{ik} = - \left(4\pi c^2/\hbar\omega_k\right)^{1/2} \mu \, \ddot{\mathbf{a}}_i \cdot \vec{\nabla} \times \vec{\mathbf{u}}_k(\vec{\mathbf{r}}_N), \quad i = 1, \ 2, \ 3 \tag{A3}$$

for q-type (magnetic) coupling, and

$$\gamma_{ik} = (4\pi\omega_k/\hbar)^{1/2} \,\mu \bar{a}_i \cdot \bar{u}_k(\bar{r}_N), \quad i = 1, \ 2, \ 3 \qquad (A4)$$

for *p*-type (electric) coupling, where  $\mathbf{\tilde{r}}_N$  indicates the location of the NO. If the NO were described by a current rather than by a dipole moment, the  $\omega_k$  dependence of the coupling constants would be different.<sup>17</sup>

Consider now a number of identical two-level systems ("molecules") coupled to the field. If we label the variables of the mth molecule by a parenthetical superscript, then the interaction Hamiltonian referring to a single mode is given by

$$H_{kN} = \sum_{m} H_{km} = \sum_{m} \hbar q_{k} \left( \gamma_{1k}^{(m)} l_{1}^{(m)} + \gamma_{2k}^{(m)} l_{2}^{(m)} + \gamma_{3k}^{(m)} l_{3}^{(m)} \right), \quad (A5)$$

where  $\gamma_{ik}^{(m)}$  is given by Eq. (A3) with  $\mathbf{\tilde{r}}_N$  replaced by  $\mathbf{\tilde{r}}^{(m)}$ . If, and only if,  $\gamma_{ik}^{(m)}$  is independent of m,  $H_{kN}$  can be written in the form of Eq. (5), where

$$\gamma_{ik}^{(m)} = \gamma_{ik}, \quad \sum_{m} l_i^{(m)} = l_i \quad . \tag{A6}$$

Now, the dependence of  $\gamma_{ik}^{(m)}$  on *m* arises only from the factor  $\mathbf{\tilde{u}}_k(\mathbf{\tilde{r}}_m)$ . If one is concerned with only a single mode, then the requirement that  $\gamma_{ik}^{(m)}$  be independent of *m* is met approximately by either placing the molecules within a region small compared to a wavelength, or placing them at (periodic) locations where  $\mathbf{\tilde{u}}_{ik}(\mathbf{\tilde{r}}^{(m)})$  has approximately the same value. It can be shown<sup>14</sup> that if a single resonant traveling-wave mode—or a number of almost identical resonant traveling-wave modes— is considered, a trivial transformation of the molecular variables will make the coupling constant uniform in a region unrestricted-or almost unrestrictedin the direction of propagation. (This transformation essentially "phases" the molecules so that the phase relationship between the oscillation of the mth molecule and that of the mode at  $\vec{r}^{(m)}$  is the same for all m.) If, however, one is concerned with a large number of modes haveing a wide range of characteristic properties, then the molecular coupling constants will be approximately independent of m only for modes which have wavelengths significantly larger than the region where the molecules are located. The collective effect of the molecules on modes with shorter wavelength (with the exception of the resonant modes mentioned above) will be reduced, and this reduction may be incorporated approximately in the present theory by a reduction in the absolute value of the effective  $\gamma_{ik}$ , or, essentially, by a cutoff.

The model used by Bonifacio *et al.*, <sup>14,16</sup> who consider a single traveling-wave mode, appears to impose the least restriction on the location of the molecules, and, therefore, on the number of molecules which behave cooperatively. These authors consider a waveguide closed at one end and open at the other, so that field radiated by the molecules escapes, and they assume the single mode to be that of a traveling wave. This model is subject to the criticism that it implicitly neglects all other modes that exist in such a waveguide structure, modes arising from the fact that a combination of traveling waves is required to form a field node at the closed end, and a range of frequencies is allowed by the open end.

#### APPENDIX B

We consider the nonlinear differential equation (68),

$$\ddot{L}_{3} - 3\alpha L_{3}\dot{L}_{3} + \Omega^{2}L_{3} = 0 , \qquad (B1)$$

with the initial conditions  $L_3(0) = -L_0$ ,  $L_3(0) = 0$ . The change of variable  $L_3 = x$ ,  $L_3 = y$ , permits this equation to be written as a pair of first-order equations,

$$\dot{x} = y , \qquad (B2a)$$

$$\dot{y} = (3\alpha y - \Omega^2)x , \qquad (B2b)$$

$$x(0) = -L_0, \quad y(0) = 0,$$
 (B2c)

which leads to

$$\frac{dy}{dx} = \frac{(3\alpha y - \Omega^2)x}{y}, \qquad (B3)$$

and

$$\frac{y\,dy}{3\alpha\,y-\Omega^2} = x\,dx \ . \tag{B4}$$

Integration of both sides yields

$$x^{2} = L_{0}^{2} + \frac{2}{(3\alpha)^{2}} \left[ 3\alpha y + \Omega^{2} \ln \left( 1 - \frac{3\alpha}{\Omega^{2}} y \right) \right].$$
(B5)

Consider, now, a graph of the solution in the x-yplane. Equations (B3) and (B5) imply the following statements: (i) The graph is symmetrical with respect to the y axis. (ii) The slope is zero at the y intercept(s). (iii) The slope is infinite at the xintercepts  $\pm L_0$ . Tracing the graph with time from t=0, we start at  $x=-L_0$ , y=0, and go up into the second quadrant (since  $\dot{y} > 0$  for y = 0, x < 0). The slope decreases as y increases, from Eq. (B3), and becomes zero as we reach the x axis. One has to show, of course, that the factor  $3\alpha y - \Omega^2$ remains negative, and does not change sign before x does. This can be done by noting that  $y = \Omega^2/3\alpha$ is a solution of Eqs. (B2a) and (B2b). In view of the theorem that the graphs of two solutions (corresponding to different initial conditions) cannot join.<sup>27</sup> we have

$$y < \Omega^2/3\alpha$$
, (B6)

and  $3\alpha y - \Omega^2$  is everywhere negative.

Proceeding clockwise with the graph into the first quadrant, we obtain the mirror image of that in the second quadrant. As we enter the fourth quadrant,  $x \approx L_0$ , y < 0. The slope, therefore, starts to decrease from  $\infty$ , and, as y decreases (or increases in absolute value) x approaches zero, at which point the slope becomes zero. The graph in the third quadrant is a reflection of that in the fourth quadrant, and when we reach the x axis, at  $-L_0$ , the graph is closed and a cycle has been completed. x and y thus execute periodic oscillation, the period being

$$T = \oint dx / y(x) . \tag{B7}$$

One can show, either by expanding the logarithm in Eq. (B5) in powers of  $\alpha$ , or by applying perturbation theory to Eq. (B1) with  $\alpha$  as the perturbation parameter, that, to first order in  $\alpha$ , the period T is independent of  $\alpha$ , and given by  $T = 2\pi/\Omega$ . While the time of rise of  $L_3$  from  $-L_0$  to  $L_0$  is increased approximately by  $\alpha L_0/2\Omega^2$ , the succeeding time of fall from  $L_0$  to  $-L_0$  is decreased by approximately the same amount. The physical explanation is obvious: On the way up, the power gained by the NO is the power absorbed from the driving field minus that radiated, while on the way down, the power loss is the power given up to the driving field plus that radiated.

#### APPENDIX C

A method for obtaining the successive derivatives  $\langle L_3^{(1)}(\tau) \rangle$ ,  $\langle L_3^{(2)}(\tau) \rangle$ , ...,  $L_3^{(n)}(\tau)$  [where a parenthetical superscript (n) indicates the *n*th derivative with

respect to  $\tau$ ] as expectation values of a polynomial in  $L_3$  will be developed. We need, first, an expression for  $(d/d\tau) \langle L_3^{\flat} \rangle$  (*p* being an ordinary power index). Consider

$$\frac{d}{d\tau} \langle L_3^{p+1} \rangle = \sum_{s=0}^{p} \langle L_3^s L_3^{(1)} L_3^{p-s} \rangle$$
$$= -\alpha^{-1} \sum_{s=0}^{p} \langle L_3^s (L_* \alpha - \alpha^{\dagger} L_*) L_3^{p-s} \rangle , \quad (C1)$$

where Eqs. (87c) and (89) have been used in the second step. Since all variables refer to the same time,  $\alpha$  can be moved to the extreme left-hand side, and  $\alpha^{\dagger}$  can be moved to the extreme right-hand side, and then replaced by the expression from Eqs.

(88). The expectation values of the terms containing  $\alpha_0$  and  $\alpha_0^{\dagger}$  vanish [see Eq. (34)], and the result is

$$\frac{d}{d\tau} \langle L_3^{p+1} \rangle = -\sum_{s=0}^{p} \langle L_3^s L_* L_3^{p-s} L_- + L_* L_3^s L_- L_3^{p-s} \rangle .$$
(C2)

The commutation relationships of Eqs. (15) lead to

$$L_3^n L_2 = L_2 (L_3 - 1)^n , (C3)$$

which, when utilized in Eq. (C2) to bring  $L_{\star}$  and  $L_{\star}$  adjacent to each other, together with the relationship

$$L_{+}L_{-} = \frac{1}{2}(L^{2} - L_{3}^{2} + L_{3}), \qquad (C4)$$

yield

$$\frac{d}{d\tau} \langle L_3^{p+1} \rangle = \langle (L_3^2 - L_3 - L^2) [ L_3^{p+1} - (L_3 - 1)^{p+1} ] \rangle.$$
(C5)

If we start with a set of polynomial coefficients  $a_m^{(n)}$  such that

$$\langle L_3^{(n)} \rangle = \left\langle \sum_{m=0}^{n+1} a_m^{(n)} L_3^m \right\rangle, \tag{C6}$$

then, we obtain, from Eq. (C5),

$$\langle L_{3}^{(n+1)} \rangle = \langle (L_{3}^{2} - L_{3} - L^{2}) \rangle \sum_{m=1}^{n+1} a_{m}^{(n)} \left[ L_{3}^{m} - (L_{3} - 1)^{m} \right],$$
(C7)

which permits the determination of the next-higher set of polynomial coefficients. Equation (99) is thus obtained from Eq. (96). In order to be able to obtain a set of coefficients  $a_m^{(n+1)}$  from the previous set  $a_m^{(n)}$ , it is convenient to break the process into two steps:

$$A_{p}^{(n+1)} = \sum_{t=0}^{n+1-p} (-1)^{t} {\binom{p+t}{1+t}} a_{p+t}^{(n)} , \qquad (C8)$$

and

$$a_{0}^{(n+1)} = -L^{2}A_{1}^{(n+1)},$$

$$a_{1}^{(n+1)} = -A_{1}^{(n+1)} - L^{2}A_{2}^{(n+1)},$$

$$\vdots$$

$$a_{r}^{(n+1)} = A_{r-1}^{(n+1)} - A_{r}^{(n+1)} - L^{2}A_{r+1}^{(n+1)}, \quad 2 \leq \gamma \leq n$$

$$a_{n+1}^{(n+1)} = A_n^{(n+1)} - A_{n+1}^{(n+1)},$$
  

$$a_{n+2}^{(n+1)} = A_{n+1}^{(n+1)}.$$
(C9)

The A's have a computational significance of their own, since

$$\langle L_{3}^{(n+1)} \rangle = \sum_{p=1}^{n+1} \langle L_{3}^{p+1} - L_{3}^{p} - L^{2} L_{3}^{p-1} \rangle A_{p}^{(n+1)} .$$
 (C10)

This expression is useful in calculating  $\langle L_3^{(n)} \rangle$  when the NO is in an energy state. In particular, for  $\langle L_3(0) \rangle = L_0$ , we obtain

$$\langle L_{3}^{(n+1)}(0) \rangle = -2 \sum_{p=1}^{n+1} A_{p}^{(n+1)} L_{0}^{p}$$
 (C11)

It should be noted that the coefficients themselves involve  $L_0$ . Equation (C11) was used to obtain the first six derivatives presented in Eq. (100).

### APPENDIX D

An expression will be derived for  $\langle L_3^2(t) \rangle$  up to order  $\alpha$  (that is, up to second order), with the NO initially in the ground state. We utilize the operator expression for  $L_3(t)$  given by Eqs. (169). It is clear that those terms in  $L_3(t)$  which yield zero when operating on the ground state both from the left- and the right-hand sides [note that  $\langle |L_{+}(0) = L_{-}(0)| \rangle = 0$ ] can be neglected for present purposes. Thus, we drop the term containing K in Eq. (169a) immediately. Using a parenthetical superscript to indicate perturbation-theory order in the present Appendix, the expressions we need are  $L_3^{(0)}$ ,  $L_3^{(1)}$ , and  $L_3^{(2)}$ , with  $\langle L_3^2 \rangle$  given in terms of these by Eq. (171). From Eq. (164), we can write

$$L_{3}^{(0)} = L_{3}(0) \cos\Omega t - M(0) \sin\Omega t , \qquad (D1)$$

where, for notational simplicity, we set

$$M(0) \equiv \dot{L}_{3}^{(0)}(0) = \Omega^{-1}[AL_{+}(0) + A^{*}L_{-}(0)].$$
 (D2)

From Eqs. (168), we obtain

$$L_{3}^{(i)} \rightarrow \Omega^{-1} \int_{0}^{t} dt_{1} \sin \Omega (t - t_{1}) Z^{(i)}(t_{1}), \quad i = 1, 2$$
(D3)

the arrow indicating that terms (not necessarily all) which will not contribute to  $\langle L_3^{(2)} \rangle$  up to order  $\alpha$  are dropped, with  $Z^{(1)}$  and  $Z^{(2)}$  given by

$$Z^{(1)} = -\left[A \,\mathfrak{a}_{0}^{\dagger} L_{3}^{(0)} + A^{*} L_{3}^{(0)} \mathfrak{a}\right] - \frac{d}{dt} \left(L_{*}^{(0)} \mathfrak{a}_{0} + \mathfrak{a}_{0}^{\dagger} L_{-}^{(0)}\right) ,$$
(D4)

$$Z^{(2)} = -\left[A \mathfrak{a}_{0}^{\dagger} L_{3}^{(1)} + A^{*} L_{3}^{(1)} \mathfrak{a}_{0}\right] - \frac{d}{dt} \left(L_{+}^{(1)} \mathfrak{a}_{0}^{\dagger} + \mathfrak{a}_{0}^{\dagger} L_{-}^{(1)}\right) \\ - \alpha \left(A L_{+}^{(0)} L_{3}^{(0)} + A^{*} L_{3}^{(0)} L_{-}^{(0)} + 2 \frac{d}{dt} \left(L_{+}^{(0)} L_{-}^{(0)}\right)\right).$$

(D5)

It is convenient to express  $L_{+}^{(i)}(t)$  and  $L_{-}^{(i)}(t)$  in terms of  $L_{3}(t)$  by the equations

$$L_{+}^{(0)}(t) = L_{+}(0) + A^{*} \int_{0}^{t} dt_{1} L_{3}^{(0)}(t_{1}) , \qquad (D6a)$$

$$L_{-}^{(0)}(t) = L_{-}(0) + A \int_{0}^{t} dt_{1} L_{3}^{(0)}(t_{1}) , \qquad (D6b)$$

$$\begin{aligned} L_{+}^{(1)}(t) &= A^{*} \int_{0}^{t} dt_{1} L_{3}^{(1)}(t_{1}) + \int_{0}^{t} dt_{1} \mathfrak{C}_{0}^{\dagger}(t_{1}) L_{3}^{(0)}(t_{1}) , \\ (D6c) \\ L_{-}^{(1)}(t) &= A \int_{0}^{t} dt_{1} L_{3}^{(1)}(t_{1}) + \int_{0}^{t} dt_{1} L_{3}^{(0)}(t_{1}) \mathfrak{C}_{0}(t_{1}) . \\ (D6d) \end{aligned}$$

Making the appropriate substitution, again dropping terms that will not contribute to the result we seek, and performing an integration by parts, we obtain

$$L_{3}^{(1)}(t) \rightarrow -\Omega^{-1} \int_{0}^{t} dt_{1} \sin\Omega(t-t_{1}) L_{3}^{(0)}(t_{1})$$

$$\times [A \alpha_{0}^{\dagger}(t_{1}) + A^{*} \alpha_{0}(t_{1})] - \int_{0}^{t} dt_{1} \cos\Omega(t-t_{1})$$

$$\times [A \alpha_{0}^{\dagger}(t_{1}) + A^{*} \alpha_{0}(t_{1})] \int_{0}^{t_{1}} dt_{2} L_{3}^{(0)}(t_{2}) . \quad (D7)$$

Using similar techniques, we also obtain

$$L_{3}^{(2)}(t) \rightarrow -(3\alpha/\Omega) \int_{0}^{t} dt_{1} \sin\Omega(t-t_{1}) \left[AL_{*}(0)L_{3}^{(0)}(t_{1}) +A^{*}L_{3}^{(0)}(t_{1})L_{*}(0) + \frac{1}{2}\Omega^{2} \int_{0}^{t_{1}} dt_{2} \left\{L_{3}^{(0)}(t_{1}), L_{3}^{(0)}(t_{2})\right\}\right].$$
(D8)

Since 
$$L_3(0)|\rangle = -L_0|\rangle$$
 and

$$\langle M^2(0) \rangle = \Omega^{-2} |A|^2 \langle L_0()L_0() \rangle = \frac{1}{2} L_0$$
, (D9)

one obtains, from Eq. (D2),

$$\langle L_3^{(0)2} \rangle = L_0^2 \cos^2 \Omega t + \frac{1}{2} L_0 \sin^2 \Omega t$$
 (D10)

One also has, immediately,

$$\langle \{L_3^{(0)}, L_3^{(1)}\} \rangle = 0$$
, (D11)

in view of the fact that  $\alpha_0$  and  $\alpha_0^{\dagger}$  (which are the zeroth-order field variables) commute with the zeroth-order NO variables.

\*Present address: Physics Department, Technion-Israel Institute of Technology, Haifa, Israel.

<sup>1</sup>I. R. Senitzky, Phys. Rev. A <u>3</u>, 421 (1971).

<sup>2</sup>The individual atoms are regarded here as identical two-level systems; in other words, it is the atomic oscillation with respect to a single pair of levels that is under consideration. The two-level systems may be regarded as spin- $\frac{1}{2}$  systems, and a number of such systems may be regarded as a single system with an angular momentum obtained according to the angular momentum addition theorem. Implicit in the use of a large (multilevel) angular momentum system coupled to the field as a model for a large number of identical atoms coupled to the field is the assumption that there is no coupling between the atoms themselves other than through the field, and that the coupling between the atoms and the field is identical for all the atoms. (See Appendix A for a more detailed discussion of the coupling.)

<sup>4</sup>V. Weisskopf and E. Wigner, Z. Physik 63, 54 (1930).

In the calculation of  $\langle L_3^{(1)2} \rangle$ , we encounter the expectation value of a product of two zeroth-order field variables. The only expectation value which does not vanish is  $\langle a_0(t_1)a_0^{\dagger}(t_2) \rangle$ . From Eq. (19), we obtain

$$\langle \alpha_{0}(t_{1}) \alpha_{0}^{\dagger}(t_{2}) \rangle = \frac{1}{4} \sum_{k,k'} \gamma_{k}^{*} \gamma_{k'} \langle A_{k}(0) A_{k'}^{\dagger}(0) \rangle e^{i (\nu_{k'} t_{2} - \nu_{k} t_{1})}$$

$$= \frac{1}{4} \sum_{k} |\gamma_{k}|^{2} e^{i \nu_{k} (t_{2} - t_{1})} , \qquad (D12)$$

where, it is recalled,  $\nu_k \equiv \omega_k - \omega$ . Using the same notation as that of Eq. (22), and familiar approximation methods, one gets

$$\begin{aligned} \langle \boldsymbol{\alpha}_{0}(t_{1})\boldsymbol{\alpha}_{0}^{\dagger}(t_{2}) \rangle &\approx \frac{1}{4} \int_{0}^{\infty} d\omega_{k} \left| \gamma(\omega_{k}) \right|^{2} \rho(\omega_{k}) e^{i (\omega_{k} - \omega) (t_{2} - t_{1})} \\ &\approx \frac{1}{4} \left| \gamma(\omega) \right|^{2} \rho(\omega) \int_{0}^{\infty} d\omega_{k} e^{i (\omega_{k} - \omega) (t_{2} - t_{1})} \\ &\approx 2 \alpha \delta(t_{1} - t_{2}) , \end{aligned} \tag{D13}$$

where, as previously, the subscript has been dropped from  $\alpha_1$ . The calculation of  $\langle L_3^{(1)2} \rangle$  also involves the integration of oscillatory functions and products of oscillatory functions. We approximate by retaining only secular terms in such integrations, that is, terms which (after the integration is carried out) contain factors that increase with time. This approximation is essentially equivalent to that of neglecting  $L_0/\Omega$  compared to t, and is acceptable if t satisfies the inequality (166). Using Eqs. (D7), (D13), and the above approximation, one obtains

$$\langle L_3^{(1)2} \rangle = \frac{1}{4} \alpha t L_0 [3 \cos^2 \Omega t + (4L_0 + 1) \sin^2 \Omega t].$$
 (D14)

The same approximation, using Eqs. (D1) and (D8), yields

$$\langle \left\{ L_3^{(0)}(t), \ L_3^{(2)}(t) \right\} \rangle = - 3\alpha t (\frac{1}{4} L_0 \sin^2 \Omega t + \frac{1}{2} L_0^2 \cos^2 \Omega t).$$
 (D15)

Equations (D10), (D11), (D14), and (D15) constitute Eqs. (172) of the text.

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<sup>18</sup>It might be argued that prescribing a *c*-number driving field is inconsistent with a quantum-mechanical treatment of the field and its interaction with the NO. It should be understood, however, that such a prescription merely indicates that the *source* of the driving field is undisturbed by the behavior of the NO, as discussed in detail in several earlier papers on quantum optics: I. R. Senitzky, Phys. Rev. Letters <u>15</u>, 233 (1965); <u>16</u>, 619 (1966); Phys. Rev. <u>155</u>, 1387 (1967). The reaction of the NO on the field is fully taken into account through the coupling of the NO to all the modes, which are referred to as the radiation field, with the word "radiation" sometimes omitted when no ambiguity is possible.

 $^{19}\text{The expression for } \alpha$  thus obtained consists of the uncoupled field (which is  $\alpha_0$ ) and the reaction of the field due to the presence of the NO. If one regards the field as a loss mechanism or a thermal reservoir, it is easily seen that this derivation is similar, in principle, to that for a more general dissipation mechanism treated in Ref. 3, and also in two later papers: I. R. Senitzky, Phys. Rev. 137, A1635 (1965), Sec. II A; 155, 1387 (1967), Sec. I. The present analysis differs from that of Dillard and Robl (Ref. 8) by the presence of the constant  $\alpha_{2}$ , and also terms which are dropped in the rotating-wave approximation used by them. (This accounts for the fact that they obtain no frequency shift.)  $\alpha_1 L_{-}$ may be regarded as a purely resistive, or dissipative, effect of the field of the NO, while  $i \alpha_2 L_{-}$  may be regarded (in the language of circuit theory) as a reactive effect. The expressions for  $\alpha_1$  and  $\alpha_2$  illustrate the fact that the dissipation is mainly a resonant phenomenon, due to the coupling to modes of approximately the same frequency, while the frequency shift is mainly a nonresonant phenomenon, due to the coupling to modes of significantly different frequency.

 $^{20}\mbox{It}$  is to be noticed that the frequency shift can be written as  $m\tilde{\alpha} - \alpha'$ , where  $\tilde{\alpha}$  and  $\alpha'$  are defined by Eqs. (37d) and (74d), respectively. As mentioned in connection with these equations, the frequency shift is determined mainly by terms that are discarded in the use of the rotatingwave approximation. It is found by G. S. Agarwal [Phys. Rev. A 4, 1778 (1971)], who used essentially a Schrödinger-picture description of the system, that the rotating-wave approximation affects the solution significantly only if the initial state of the NO has a well-defined phase. The physical reason for this effect becomes obvious in light of the present discussion. Oscillators with slightly different frequencies will be in much different "states" after a number of cycles if the "states" describe the instantaneous values of the oscillating coordinates, that is, if they describe the phase of the oscillation.

<sup>21</sup>The above expressions for  $\langle L_3 \rangle$  are equivalent to those of Dillard and Robl, Ref. 7, who derive, essentially,  $\langle L_3 \rangle$  for n = 2-5, 9. Their method may appear more complicated because they do not take explicit advantage of the fact that  $L^2$  is a constant of motion.

<sup>22</sup>I. R. Senitzky, Phys. Rev. Letters <u>19</u>, 1062 (1968). <sup>23</sup>The amplitude of oscillation of the NO for  $\langle L_3 \rangle$  in the neighborhoods of  $\pm L_0$  is very small, of course, but—on the one hand—these neighborhoods become relatively smaller as  $L_0$  increases, and—on the other—they are passed through more quickly as the driving field increases. <sup>24</sup>Leon Kotin (unpublished).

<sup>25</sup>The classical equation for the strong-driving-field case may be recovered by setting these quantities equal to zero, differentiating both sides, and noting that  $-\alpha \Omega^2 \int_0^t dt_1 L_3(t_1)$  is equal to  $\alpha \dot{L}_3$  up to order  $\alpha$ . <sup>26</sup>I. R. Senitzky, Phys. Rev. <u>134</u>, A816 (1964).

<sup>27</sup>See, for instance, W. Hurewicz, Lectures on Ordinary Differential Equations (Wiley, New York, 1958).

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## Quantum Theory of Diffusion with Application to Solid Helium

Allan Widom\* and M. G. Richards School of Mathematical and Physical Sciences, University of Sussex, Brighton, Sussex, England (Received 3 February 1972)

A quantum theory of diffusion is presented and applied to the diffusion of isotopic impurities in solid helium. For temperatures much less than the Debye temperature  $\Theta$  and much more than the impurity exchange temperature  $\hbar J/k_B$ , it is shown that the diffusivity is given by  $D = (Ja^4/\sigma^*x)$ . The effective cross section  $\sigma^*$  for the scattering of two mobile impurity atoms is of the order of a square lattice spacing  $a^2$ , and the mole fraction x of the impurity atoms is assumed to obey  $(\hbar J/k_B \Theta) (T/\Theta)^7 \ll x \ll 1$ . Observation of the concentration dependence  $D \propto 1/x$  would constitute strong evidence of quantum mobility, which has been of considerable theoretical interest in recent years.

### I. INTRODUCTION

In the limit of low temperatures, where thermally activated diffusion is virtually zero, the conventional picture of a crystalline solid is one of atoms immobile on equilibrium lattice sites. This picture cannot be precisely correct. For low concentrations of impurity atoms a simple counting argument shows that a macroscopic degeneracy would exist at zero temperature leading to an entropy of solid solution in violation of the third law of thermodynamics.<sup>1</sup> This apparent