

ting.¹⁶ When 80 steps were taken between planes (as they were for the third movie), there was about about 0.1 period per step. This is a little more than necessary, but from an information-theory

viewpoint the focusing inhomogeneity is not sampled frequently enough. This explains the unphysical roughness of the last frame. It was felt that further refinement was not justified.

¹G. N. Steinberg, *Phys. Rev. A* **4**, 1182 (1971). See also I. L. Kerr, in *Damage in Laser Glass*, edited by A. J. Glass and A. H. Guenther (American Society for Testing and Materials, Philadelphia, 1969) Spec. Tech. Publ. 469, p. 23.

²E. L. Kerr, *Phys. Rev. A* **4**, 1195 (1971). The first equation of Sec. IV C should have been divided by 4 instead of 2. This makes all values of the failure intensity I incorrect but they may be corrected by writing $2I$ for I everywhere it appears.

³R. Y. Chiao, E. Garmire, and C. H. Townes, *Phys. Rev. Letters* **13**, 479 (1964).

⁴P. K. Tien, J. P. Gordon, and J. R. Whinnery, *Proc. IEEE* **53**, 129 (1965).

⁵J.-C. Buges, J.-M. Jégo, A. Terneaud, and P. Veyrie, *Compt. Rend.* **264B**, 871 (1967); J. P. Budin and J. Raffy, *Appl. Phys. Letters* **9**, 291 (1966).

⁶J. H. Marburger and E. L. Dawes, *Phys. Rev. Letters* **21**, 556 (1968); and V. N. Lugovoi and A. M. Prokhorov, *Zh. Eksperim. i Teor. Fiz. Pis'ma v Redaktsiyu* **7**, 153 (1968) [*Sov. Phys. JETP Letters* **7**, 117 (1968)].

⁷C. R. Giuliano and J. H. Marburger, *Phys. Rev. Letters* **27**, 905 (1971).

⁸M. Bass and H. H. Barrett, in *Damage in Laser Materials: 1971*, edited by A. J. Glass and A. H. Guenther, Natl. Bur. Std. Spec. Publ. No. 356 (U. S. GPO Washington, D. C., 1971), p. 76.

⁹M. A. Duguay, J. W. Hansen, and S. L. Shapiro, *IEEE J. Quantum Electron.* **QE-6**, 725 (1970).

¹⁰E. L. Kerr, Perkin-Elmer Report No. 10804, 1971 (unpublished). The report includes FORTRAN source listings. Machine-readable copies of the programs are not available.

¹¹G. M. Zverev and V. A. Pashkov, *Zh. Eksperim. i Teor. Fiz.* **57**, 1128 (1969) [*Sov. Phys. JETP* **30**, 616 (1970)].

¹²M. M. Denariez-Roberge and J.-P. E. Taran, *Appl. Phys. Letters* **14**, 205 (1969).

¹³In the electrostatic equation $n = n_0 + \frac{1}{2}n_2 \langle E^2 \rangle$ the units of n_2 are $\text{cm sec}^2/\text{g}$. The value of n_2 may be converted to the mks units $\text{m sec}^2/\text{kg}$ by multiplying by ten. At the same time the above index equation should be rewritten in the mks system as $n = n_0 + 2\pi\epsilon_0 n_2 \langle E^2 \rangle = n_0 + (2\pi n_2/cn_0)I$. The additional constant $4\pi\epsilon_0 \equiv 10^7/c^2$ is required to avoid a dimensional conversion factor in the index equation. To report a value of n_2 in units such as m^2/V^2 is to redefine n_2 by writing the esu index equation directly in mks units without converting it.

¹⁴A. V. Shatilov, A. I. Stozharov, and E. M. Smirnov, *Sov. J. Opt. Tech.* **37**, 403 (1970).

¹⁵E. L. Kerr, *IEEE J. Quantum Electron.* **QE-7**, 532 (1971).

¹⁶*Handbook of Mathematical Functions*, edited by N. Abramowitz and I. A. Stegun (U. S. GPO, Washington, D. C., 1964), p. 879.

Quantum-Mechanical Saturation in Resonance Fluorescence

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A number of radiating two-level systems subject to an external resonant driving field are treated as a single angular momentum system. It is shown that the damping of the oscillation of the energy expectation value is the result of quantum-mechanical averaging over an ensemble of experiments, the energy oscillation in a single experiment being undamped.

In the case of spontaneous emission, a collection of identical two-level atomic systems in an idealized situation have been treated by a number of authors as a single angular momentum system (AMS) of large quantum number L_0 .¹⁻⁵ No such treatment appears to exist for resonance fluorescence. It is the purpose of the present paper to outline such a treatment for a strong incident field and discuss a result which not only requires modification of previous theory, but is also of interest from a general quantum-mechanical viewpoint and

illustrates the use of classical solutions in the interpretation of the quantum-mechanical results.

The advantages of the AMS method in the analysis of spontaneous emission are the following: (i) Certain correlations between the atomic systems, which are unaffected by the electromagnetic field, are automatically accounted for and preserved by the conservation of total angular momentum. (ii) A large AMS may be expected to behave in an essentially classical manner except when its energy is near maximum and the external field is weak.

Since the difference in experimental conditions between spontaneous emission and resonance fluorescence is the presence of an external driving field, the above advantages exist also in the case of resonance fluorescence, with the second advantage acquiring additional importance when the driving field is strong. In fact, it is the comparison of the classical and quantum-mechanical analyses that contains, at first glance, surprises, and affects the physical interpretation of the quantum-mechanical results.

The system under consideration is an AMS driven by a prescribed (*c*-number) field and coupled to a radiation field consisting of a large number of (denumerable) modes closely spaced in frequency. The Hamiltonian is given by

$$H = \hbar\omega l_3 + \sum_k \hbar\omega_k (a_k^\dagger a_k + \frac{1}{2}) + \frac{1}{2}\hbar \sum_k (\gamma_k a_k^\dagger l_- + \gamma_k^* a_k l_+) + \frac{1}{2}\hbar (\gamma a^* l_- + \gamma^* a l_+) \quad (1)$$

and is explained as follows: The AMS variables are the (dimensionless) angular momentum components l_1 , l_2 , and l_3 , satisfying $[l_1, l_2] = il_3$, etc., with $l_\pm = (1/\sqrt{2})(l_1 \pm il_2)$; the variables of the k th mode, of (angular) frequency ω_k are a_k and a_k^\dagger , satisfying $[a_k, a_k^\dagger] = 1$; a and a^* represent the prescribed driving field, of frequency ω ; the rotating-wave approximation has been used. The preceding commutators become Poisson brackets multiplied by i in the classical analysis. Introducing "reduced" variables A_k , A_k^\dagger , L_\pm , and L_3 , defined by

$$a_k \equiv A_k e^{-i\omega_k t}, \quad l_\pm \equiv L_\pm e^{\pm i\omega t}, \quad l_3 \equiv L_3, \quad (2)$$

and utilizing the additional definitions

$$\mathfrak{A} \equiv \frac{1}{2}i \sum_k \gamma_k^* A_k e^{-i\nu_k t}, \quad A \equiv \frac{1}{2}i\gamma a e^{i\omega t}, \quad (3)$$

$$\mathfrak{A}_0 \equiv \frac{1}{2}i \sum_k \gamma_k^* A_k(0) e^{-i\nu_k t}, \quad \nu_k \equiv \omega_k - \omega,$$

we obtain for the equations of motion, both classically and quantum mechanically,

$$\begin{aligned} \dot{L}_+ &= (A^* + \mathfrak{A}^\dagger)L_3, \quad \dot{L}_- = L_3(A + \mathfrak{A}), \\ \dot{L}_3 &= -[L_+(A + \mathfrak{A}) + (A^* + \mathfrak{A}^\dagger)L_-], \\ \dot{A}_k &= -\frac{1}{2}i\gamma_k L_- e^{i\nu_k t}, \quad \dot{A}_k^\dagger = \frac{1}{2}i\gamma_k^* L_+ e^{-i\nu_k t}. \end{aligned} \quad (4)$$

It is to be noted that A is a constant. One also notes that the square of the total angular momentum, given by

$$L^2 \equiv L_3^2 + L_+ L_- + L_- L_+, \quad (5)$$

is a constant of motion which is equal to L_0^2 classically and $L_0(L_0 + 1)$ quantum mechanically.

If we assume sufficiently weak coupling between the AMS and the radiation field so that the reduced variables vary slowly compared to $e^{i\omega t}$, we obtain, by integrating \dot{A}_k and \dot{A}_k^\dagger ,^{6,7}

$$\alpha(t) \approx \alpha_0(t) + \alpha L_-(t), \quad \alpha^\dagger(t) \approx \alpha_0^\dagger(t) + \alpha L_+(t), \quad (6)$$

$$\alpha \equiv \frac{1}{4}\pi |\gamma(\omega)|^2 \rho(\omega), \quad (7)$$

where $|\gamma(\omega)|^2$ is the average of $|\gamma_k|^2$ over all k 's in a small neighborhood about $\omega_k = \omega$, and $\rho(\omega)$ is the number of modes per unit interval of ω_k at ω . The significant approximations contained in Eq. (7) are the neglect of the imaginary part of α (the main effect of which is the production of a frequency shift in the AMS, a shift that has already been qualitatively affected by the rotating-wave approximation) and the neglect of the time dependence of α , such that α increases from an initial value of zero to the value given by Eq. (7) in a time period which is small compared with that for a significant change in the reduced variables but large compared to ω^{-1} .

Substitution from Eqs. (6) yields equations of motion for the AMS variables only;

$$\dot{L}_+ = (A^* + \mathfrak{A}_0^\dagger)L_3 + \alpha L_- L_3, \quad (8a)$$

$$\dot{L}_- = L_3(A + \mathfrak{A}_0) + \alpha L_3 L_+, \quad (8b)$$

$$\dot{L}_3 = -[L_+(A + \mathfrak{A}_0) + (A^* + \mathfrak{A}_0^\dagger)L_-] - 2\alpha L_+ L_- . \quad (8c)$$

We consider initial conditions in which the radiation field is in the ground state, that is,

$$\langle |\mathfrak{A}_0^\dagger = \mathfrak{A}_0| \rangle = 0 \quad (9)$$

quantum mechanically, and

$$\alpha_0 = \mathfrak{A}_0^\dagger = 0 \quad (10)$$

classically. One obtains, with some obvious approximation,

$$\langle \mathfrak{A}_0(t_1) \mathfrak{A}_0^\dagger(t_2) \rangle = 2\alpha \delta(t_1 - t_2) \quad (11)$$

quantum mechanically. The physical meaning of α is displayed by the consideration of a (quantum-mechanical) two-level system in absence of a driving field. For $A = 0$, Eq. (8c) yields, for such a system,

$$\langle \dot{L}_3 \rangle = -\alpha \langle (L_3 + \frac{1}{2}) \rangle. \quad (12)$$

It is seen that α is just the spontaneous transition probability per unit time or the Weiskopf-Wigner decay constant.

Setting $2|A|^2 \equiv \Omega^2$ and using the notation $\{A, B\} = AB + BA$, we obtain from Eqs. (8)

$$\begin{aligned} \langle \ddot{L}_3 \rangle + \frac{3}{2}\alpha\lambda \langle \dot{L}_3 \rangle + \Omega^2 \langle L_3 \rangle - \frac{3}{2}\alpha \langle \{\dot{L}_3, L_3\} \rangle \\ = \alpha^2 \langle [L_0^2 - L_3 + \lambda(L_0 + L_3)](L_3 - \lambda) \rangle \end{aligned} \quad (13)$$

quantum mechanically, with $\lambda = 1$. The corresponding classical equation is obtained by setting $\lambda = 0$ and dropping the expectation-value brackets. We consider only the case of a strong driving field, such that

$$\Omega \gg \alpha L_0. \quad (14)$$

Equation (13) can therefore be approximated by dropping the α^2 term, with the result

$$\langle \ddot{L}_3 \rangle + \frac{3}{2}\alpha\lambda \langle \dot{L}_3 \rangle + \Omega^2 \langle L_3 \rangle - \frac{3}{2}\alpha \langle \{\dot{L}_3, L_3\} \rangle = 0. \quad (15)$$

For simplicity, the AMS will be considered to be initially in the ground state, so that the initial conditions are

$$\langle L_3(0) \rangle = -L_0, \quad \langle \dot{L}_3(0) \rangle = 0. \quad (16)$$

Classically, the AMS energy is described by the differential equation

$$\ddot{L}_3 - 3\alpha L_3 \dot{L}_3 + \Omega^2 L_3 = 0, \quad (17)$$

with $L_3(0) = -L_0$ and $\dot{L}_3(0) = 0$. It is possible to analyze the trajectory of this equation in the L_3, \dot{L}_3 plane and show that this trajectory is a closed curve about the origin,⁷ which means that the solution is a periodic function of the time. One can also show, by perturbation theory, that the period is independent of α to the first order in α and is given by $2\pi/\Omega$, although the oscillation of L_3 is not sinusoidal. Thus when analyzed classically, the energy of the driven AMS oscillates periodically.

Quantum mechanically, we consider, first, a driven two-level system. In this case, $\{L_3, L_+ \} = 0$, so that Eqs. (4) yield $\{L_3, \dot{L}_3\} = 0$ and Eq. (15) becomes

$$\langle \ddot{L}_3 \rangle + \frac{3}{2}\alpha \langle \dot{L}_3 \rangle + \Omega^2 \langle L_3 \rangle = 0. \quad (18)$$

The solution is a damped oscillation which in view of inequality (14) can be written approximately as

$$\langle L_3 \rangle = -\frac{1}{2}e^{-3\alpha t/4} \cos \Omega t. \quad (19)$$

Although the period of the oscillating factor is the same as that of the classical oscillation, the amplitude of $\langle L_3 \rangle$ is damped and approaches zero.

Consider now a multilevel system. If we approximate the term $\langle \{L_3, L_3\} \rangle$ by $\langle \dot{L}_3 \rangle \langle L_3 \rangle$, Eq. (15) becomes a second-order nonlinear differential equation for $\langle L_3 \rangle$, which may be analyzed in the $\langle L_3 \rangle, \langle \dot{L}_3 \rangle$ plane by a method similar to that used for Eq. (17). This analysis shows that $\langle L_3 \rangle$ is a damped oscillation that approaches zero, the behavior being qualitatively similar to that of the two-level case. Another approach to Eq. (15) is one of perturbation theory. We consider the coupling to the radiation field (but not to the driving field) to be a small perturbation, and obtain $\langle L_3 \rangle$ to first order in α for t such that

$$L_0 \Omega^{-1} \ll t \ll \alpha^{-1}. \quad (20)$$

The result is given by

$$\langle L_3(t) \rangle \approx (1 - \frac{3}{2}\lambda \alpha t)(-L_0 \cos \Omega t). \quad (21)$$

It is seen that, quantum mechanically, the first-order expansion is consistent with an exponential damping of the oscillation of $\langle L_3 \rangle$ which is the same as that of the two-level system. Equation (21) also shows explicitly that, classically, the oscillations of L_3 are undamped at least to order α .

There appears, at first glance, a paradox: A large AMS subject to a strong field should be de-

scribable classically, and yet the behavior of $\langle L_3 \rangle$ is qualitatively different from that of the classical L_3 ! The explanation must be sought in the statistical aspect of $\langle L_3 \rangle$, associated with the uncertainty principle.⁴ The expectation value represents the average over an ensemble, each member of the ensemble being an AMS interacting with a radiation field. While a given member may behave in a manner only slightly different from that of the classical system, that is, exhibit an undamped oscillation of the energy, the differences among members (the quantum fluctuations, or the "uncertainties" of the uncertainty principle) may add in such a manner as to make the *average* behavior appear greatly different from the classical behavior. This is indeed the case for slight random variations in the frequency of oscillation of L_3 among members of the ensemble; as is well known, a small spread in frequencies will lead to a damping of the average.

The frequency of the AMS energy oscillation can be shown to contain a random part that is of quantum-mechanical (in the sense of nonclassical) origin. Consider a "classical" analysis in which quantities introduced by the uncertainty principle (i.e., the quantum fluctuations) are treated as random variables. (This type of analysis illuminates certain differences between classical and quantum-mechanical results, but may lead to difficulties when carried too far quantitatively.) Let us assume a perturbation theory viewpoint with respect to the coupling of the AMS to the radiation field. In lowest order (i.e., in the absence of coupling to the radiation field) Eq. (17) shows that L_3 oscillates with a frequency Ω , which is proportional to the amplitude of the driving field. In higher order, one can consider the frequency of oscillation of L_3 to be given approximately by Ω_{eff} , which is the corresponding quantity proportional to the effective, or total, field acting on the AMS. Classically, the total field is the superposition of the driving field and the radiated field, both of which are well defined. Quantum mechanically, the total field is the superposition of the driving field, the radiated field, and the zero-point field, with the last two fields subject to the uncertainty principle and *not* well defined. The randomness of the zero-point field requires no discussion. As far as the radiated field is concerned, consider, for simplicity, the time $t=0$. Since $L_3(0)$ is well defined, $L_+(0)$ and $L_-(0)$ are not. The radiated field, determined by \hat{A}_k and \hat{A}_k^\dagger in Eqs. (4), reflects the uncertainty in these quantities, and is therefore random with respect to the ensemble under consideration. Thus, the effective field acting on the AMS contains a random component due to both the zero-point and radiated fields, and produces a random component in the frequency of oscillation of L_3 .

In order to show that the customary practice of predicting the result of an experiment involving a large system by an expectation value is not applicable in the present instance, we examine the expression for $\langle L_3^2 \rangle$. With the same perturbation-theory approach as that used in obtaining Eq. (21), one derives (to order α) the relationship⁷

$$\langle L_3^2 \rangle \approx [L_0^2 - \frac{3}{2}\alpha t L_0(L_0 - \frac{1}{2})] \cos^2 \Omega t + [\frac{1}{2}L_0 + \alpha t L_0(L_0 - \frac{1}{2})] \sin^2 \Omega t. \quad (22)$$

(It may be noted that this expression yields the exact result for $L_0 = \frac{1}{2}$.) For large L_0 , we need consider only the L_0^2 terms. Since, from Eq. (21), $\langle L_3 \rangle^2 = L_0^2(1 - \frac{3}{2}\alpha t) \cos^2 \Omega t$ to order α , the term $\alpha t L_0^2 \sin^2 \Omega t$ cannot be reconciled with an ensemble of *damped* oscillations in which each member is approximated by the expectation value, and is, indeed, of the order of magnitude that would be expected for an ensemble of undamped oscillations for which the frequency spread introduces the indicated damping of the average.

One may ask where the fallacy lies in the viewpoint that a large number of two-level systems sub-

ject to a strong driving field should behave approximately independently, with the result being the sum of the expectation values. The answer is furnished by noting the fact that the deviations from the average are due to essentially spontaneous-emission effects, and the coupling between the two-level systems through the radiation field is precisely strong enough, in relation to these spontaneous emission effects, so that the deviations cannot be regarded as independent.

The amplitude of the *radiating* oscillation (at frequency ω) is determined by L_+ and L_- , which, in turn, are affected by L_3 according to Eq. (5). An oscillation of L_3 will produce an amplitude modulation in the radiation which, from a spectral point of view, introduces sum and difference satellite frequencies, the nonsinusoidal aspect of the modulation accounting for a number of such satellites. It is clear that an undamped oscillation of L_3 , predicted by the present analysis, will result in different radiation properties from those for a damped oscillation predicted in the literature^{6,8-10} on the basis of a calculation of $\langle L_3 \rangle$ for a two-level system.

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¹R. H. Dicke, Phys. Rev. 93, 99 (1954).

²J. H. Eberly and N. E. Rehler, Phys. Rev. A 2, 1607 (1970); 3, 1735 (1971).

³G. S. Agarwal, Phys. Rev. A 2, 2038 (1970).

⁴I. R. Senitzky, Phys. Rev. A 3, 421 (1971).

⁵R. Bonifacio, F. Haake, and P. Schwendimann, Phys.

Rev. A 4, 302 (1971); 4, 854 (1971).

⁶M. Dillard and H. R. Robl, Phys. Rev. 184, 312 (1969).

⁷I. R. Senitzky, following paper, Phys. Rev. A 6, 1175 (1972).

⁸Maurice C. Newstein, Phys. Rev. 167, 89 (1968).

⁹B. R. Mollow, Phys. Rev. A 2, 76 (1970).

¹⁰C. R. Stroud, Phys. Rev. A 3, 1044 (1971).