

PHYSICAL REVIEW LETTERS

VOLUME 34

16 JUNE 1975

NUMBER 24

Problem of Resonance Fluorescence and the Inadequacy of Spontaneous Emission as a Test of Quantum Electrodynamics*

H. J. Kimble and L. Mandel

Department of Physics and Astronomy, University of Rochester, Rochester, New York 14627

(Received 19 February 1975)

The interaction of a two-level atom with an external electromagnetic field is examined in the framework of quantum electrodynamics, without any assumption about Markoffian behavior or stationarity. It is found that spontaneous emission in the vacuum and the rate of fluorescence in an external field do not depend on quantum properties of the electromagnetic field, whereas the correlation functions do.

The problem of the interaction of an atom with a quantized electromagnetic field has received renewed attention in recent years,¹⁻¹³ partly because of the possibilities it offers for testing quantum electrodynamics (QED) at low energies. Such tests have become especially interesting since a number of semiclassical treatments of the atomic decay problem¹⁴ have succeeded in giving at least partially correct accounts of the observations. However, if one examines most of the current treatments of the problem by QED, it soon becomes apparent that certain presumptions about the answer are exceedingly common. Perturbation treatments based on the Weisskopf-Wigner method¹⁵ in effect start with an assumed answer, whereas the Heitler-Ma technique¹⁶ is usually applied only to single-photon transitions. Most treatments based on the solution of the Schrödinger equation either deal with an explicitly limited number of photons,^{6,12} or assume factorization conditions for the density operator of the combined atom-field system.³ Solutions derived from the Heisenberg equations of motion frequently are limited to a classical external field.^{1,2,7,8} Alternatively, Markoffian assumptions about the process are introduced to ensure that memory about the past dies out as the interaction proceeds, and have become standard in master-equation treatments,^{4,13,17} and approximately equivalent assumptions are frequently introduced in solutions of the Heisenberg equations of motion.^{2,10,11} However, the stochastic features and the resulting loss of memory are just what distinguish the interactions of quantum systems from most semiclassical interactions. If these features are consequences of QED, they should be revealed by solution of the equations of QED rather than inserted by *a priori* assumption.

In the following we show how the problem of the resonant interaction between a two-level atom and a quantized electromagnetic field may be tackled via the Heisenberg equations of motion without Markoffian assumptions. In certain respects our treatment is similar to that given by Senitzky⁸ and Ackerhalt and Eberly,¹⁰ except that we do not make use of mode decompositions. Although approximations are introduced in the course of the calculation, we avoid the usual assumptions about the nature of the

process, or the introduction of a classical driving field. We find that the quantum properties of the electromagnetic field do not play a significant role in spontaneous emission, and do not enter explicitly until we calculate two-time correlation functions of the fluorescence in the presence of an external field. Measurements of the light intensity in normal spontaneous atomic emission,¹⁹ or even in the presence of an exciting field, therefore appear to be inadequate as tests of QED.

As two atomic energy levels play a dominant role in this problem, we make the common simplification of representing the atom by a spin- $\frac{1}{2}$ system, having two energy eigenstates $|1\rangle$, $|2\rangle$ with energy separation $\hbar\omega_0$, and described by the three dynamical spin variables \hat{R}_1 , \hat{R}_2 , and \hat{R}_3 .²⁰ The Hamiltonian for the combined atom-field system is taken to be

$$\hat{H} = \frac{1}{2} \int [\epsilon_0 \hat{\mathbf{E}}^2(\vec{r}, t) + \mu_0^{-1} \hat{\mathbf{B}}^2(\vec{r}, t)] d^3x + \hbar\omega_0 \hat{R}_3(t) + 2\omega_0 \vec{\mu} \cdot \hat{\mathbf{A}}(0, t) \hat{R}_2(t), \quad (1)$$

when $\vec{\mu}$ is the transition dipole moment and the atom is located at the origin. We work in the Coulomb gauge in which $\hat{\mathbf{A}}(\vec{r}, t)$ is transverse. The Heisenberg equations of motion for the atomic operators then take the form

$$\dot{\hat{R}}_1(t) = -\omega_0 \hat{R}_2(t) + 2\omega_0 [\vec{\mu} \cdot \hat{\mathbf{A}}(0, t) / \hbar] \hat{R}_3(t), \quad (2)$$

$$\dot{\hat{R}}_2(t) = \omega_0 \hat{R}_1(t), \quad (3)$$

$$\dot{\hat{R}}_3(t) = -2\omega_0 [\vec{\mu} \cdot \hat{\mathbf{A}}(0, t) / \hbar] \hat{R}_1(t), \quad (4)$$

while the equations of motion for the field operators lead to Maxwell's equations. These may be combined in the form of the well-known inhomogeneous wave equation, whose retarded solution at the position of the atom, after decomposition into positive and negative frequency parts $\hat{\mathbf{A}}^{(+)}$ and $\hat{\mathbf{A}}^{(-)}$, may be expressed in the form

$$\vec{\mu} \cdot \hat{\mathbf{A}}^{(+)}(0, t) = (\hbar/\omega_0) (\beta - i\gamma) \hat{b}(t) + \vec{\mu} \cdot \hat{\mathbf{A}}_{\text{free}}^{(+)}(0, t), \quad t > 0. \quad (5)$$

Here $\hat{\mathbf{A}}_{\text{free}}(\vec{r}, t)$ is the solution of the homogeneous wave equation, $\hat{b}(t) \equiv \hat{R}_1(t) - i\hat{R}_2(t)$, $\beta \equiv (1/4\pi\epsilon_0)(2\mu^2\omega_0^3/3\hbar c^3)$, and γ is an (infinite) frequency shift parameter, corresponding to the Lamb shift for a real atom. The decomposition is not unambiguous for small t , so that the equation should be restricted to times t for which $\omega_0 t \gg 1$. Equation (5) is similar to one obtained by Ackerhalt and Eberly,¹⁰ and expresses the total field as a combination of free-field and source contributions, but was derived without Markoffian assumptions.

We now follow the procedure of Ackerhalt and Eberly¹⁰ in writing Eqs. (2) and (4) in normal order and using Eq. (5) and its conjugate to eliminate $\vec{\mu} \cdot \hat{\mathbf{A}}^{(+)}(0, t)$ and $\vec{\mu} \cdot \hat{\mathbf{A}}^{(-)}(0, t)$. We find it convenient to introduce the slowly varying dynamical variables defined by $\hat{b}_s(t) \equiv \hat{b}(t) \exp(i\omega_0 t)$ and $\hat{\mathbf{A}}_s^{(+)}(\vec{r}, t) \equiv \hat{\mathbf{A}}_{\text{free}}^{(+)}(\vec{r}, t) \exp(i\omega_0 t)$, etc. After discarding all rapidly oscillating terms, we obtain

$$\dot{\hat{b}}_s(t) = (-\beta + i\gamma) \hat{b}_s(t) + 2(\omega_0/\hbar) \hat{R}_3(t) \vec{\mu} \cdot \hat{\mathbf{A}}_s^{(+)}(0, t), \quad (6)$$

$$\dot{\hat{R}}_3(t) = -2\beta [\hat{R}_3(t) + \frac{1}{2}] - (\omega_0/\hbar) [\hat{b}_s^\dagger(t) \vec{\mu} \cdot \hat{\mathbf{A}}_s^{(+)}(0, t) + \text{H.c.}]. \quad (7)$$

We can immediately apply these equations to the problem of spontaneous decay of an excited atomic state, by taking the initial state to be the product of the electromagnetic vacuum and some arbitrary atomic state. Then the expectations of all terms in Eqs. (6) and (7) involving $\hat{\mathbf{A}}_s^{(+)}(0, t)$ and $\hat{\mathbf{A}}_s^{(-)}(0, t)$ vanish, and we obtain

$$\langle \hat{R}_3(t) \rangle + \frac{1}{2} = [\langle \hat{R}_3(0) \rangle + \frac{1}{2}] e^{-2\beta t}, \quad (8)$$

$$\langle \hat{b}_s(t) \rangle = \langle \hat{b}_s(0) \rangle e^{(-\beta + i\gamma)t}, \quad (9)$$

which are the usual solutions for the spontaneous emission problem in QED.¹⁰ However, the quantum nature of the electromagnetic field has actually played no role so far in the analysis, because Maxwell's equations hold equally in classical and quantum electrodynamics. The same argument would hold if the free-field $\hat{\mathbf{A}}_{\text{free}}$ were treated as a c -number, or classical, variable, provided the total field $\hat{\mathbf{A}}(0, t)$ at $\vec{r} = 0$ commutes with atomic operators, which is required by the transversality condition. Admittedly,

$\hat{A}(0, t)$ would then still not be a c number because of the quantum nature of the source, so that the treatment would fall outside the domain of these semiclassical theories¹⁴ in which the total field is classical. A theory in which \hat{A}_{free} is a c number would be semiclassical in the sense that all quantum features are contained within the Hilbert space of the atom alone²¹ (and it would lack internal consistency), but the solutions would remain unchanged.

Let us now examine the situation when an external electromagnetic field is present. This time we take the initial state of the system to be a product of the lower state $|1\rangle$ for the atom and the coherent quantum state $|\{v\}\rangle$ for the field,²² such that

$$\hat{A}_{\text{free}}^{(+)}(0, t)|\{v\}\rangle = \tilde{\epsilon}\mathbf{G} \exp[-i(\omega_1 t + \varphi)]|\{v\}\rangle \equiv \exp(-i\omega_0 t)\tilde{\mathbf{V}}(t)|\{v\}\rangle, \quad (10)$$

where $\tilde{\epsilon}$ is the unit polarization vector, \mathbf{G} and φ are an amplitude and a phase, and the frequency ω_1 is close to, but not necessarily coincident with, the atomic frequency ω_0 . It is readily shown that the expectation of the total light intensity registered by a distant light detector outside the exciting beam is proportional to $\langle \hat{b}^\dagger(t)\hat{b}(t) \rangle = \langle \hat{R}_3(t) \rangle + \frac{1}{2}$. To find $\langle \hat{R}_3(t) \rangle$ we may formally integrate Eqs. (6) and (7), substitute for $\hat{b}_s(t)$ in the $\hat{R}_3(t)$ equation, and take expectation values. With the help of Eq. (10) we then arrive at a simple Volterra-type integral equation for $\langle \hat{R}_3(t) \rangle$, that can be integrated exactly.²³ However, once again, no quantum properties of the free field are explicitly invoked in the derivation of the equation, other than that $\hat{A}^{(+)}(0, t)$ and $\hat{A}^{(-)}(0, t)$ commute with atomic operators, so that the measurement of the time development of the fluorescent light intensity again does not test QED explicitly.

Finally we turn briefly to the calculation of the two-time correlation function of the fluorescent light, which is proportional to $\langle \hat{b}^\dagger(t)\hat{b}(t+\tau) \rangle$, whose Fourier transform is the spectral density. On integrating Eqs. (6) and (7) and substituting for $\hat{R}_3(t)$ in the $\hat{b}_s(t)$ equation, we arrive at the following relation:

$$\begin{aligned} \langle \hat{b}_s^\dagger(t)\hat{b}_s(t+\tau) \rangle &= \frac{1}{4}\Omega^2 e^{(-\beta+i\gamma)\tau} e^{-2\beta t} \int_0^t dt' \int_0^{t+\tau} dt'' e^{(\beta+i\Delta)t'} e^{(\beta-i\Delta)t''} \\ &+ \frac{1}{4}\Omega^3 e^{(-\beta+i\gamma)\tau} e^{-2\beta t} \int_0^t dt' e^{(\beta+i\Delta)t'} \int_0^{t+\tau} dt'' e^{-(\beta+i\Delta)t''} \int_0^{t''} dt''' [\langle \hat{b}_s^\dagger(t''') \rangle e^{i(\omega_0-\omega_1)t'''} + \text{c.c.}] e^{2\beta t'''} \\ &+ \frac{1}{4}\Omega^3 e^{(-\beta+i\gamma)\tau} e^{-2\beta t} \int_t^{t+\tau} dt' e^{(\beta-i\Delta)t'} \int_0^t dt'' e^{-(\beta+i\Delta)t''} \int_0^{t''} dt''' [\langle \hat{b}_s^\dagger(t''') \rangle e^{i(\omega_0-\omega_1)t'''} + \text{c.c.}] e^{2\beta t'''} \\ &+ \Omega^2 (\omega_0/\hbar)^2 e^{(-\beta+i\gamma)\tau} e^{-2\beta t} \int_0^t dt' e^{-(\beta+i\Delta)t'} \int_0^{t'} dt'' [\tilde{\mu} \cdot \tilde{\mathbf{V}}^*(t'') \langle \hat{b}_s(t'') \rangle + \langle \hat{b}_s^\dagger(t'') \rangle \tilde{\mu} \cdot \hat{\mathbf{A}}_s^{(+)}(0, t'')] e^{2\beta t''} \\ &\times \int_0^{t+\tau} dt_1 e^{-(\beta+i\Delta)t_1} \int_0^{t_1} dt_2 [\langle \hat{b}_s^\dagger(t_2) \rangle \tilde{\mu} \cdot \tilde{\mathbf{V}}(t_2) + \tilde{\mu} \cdot \hat{\mathbf{A}}_s^{(-)}(0, t_2) \langle \hat{b}_s(t_2) \rangle] e^{2\beta t_2}, \end{aligned} \quad (11)$$

where $\Delta \equiv \gamma + \omega_1 - \omega_0$ is a measure of detuning and $\Omega \equiv 2\omega_0 \tilde{\mu} \cdot \tilde{\epsilon} \mathbf{G} / \hbar$ is the Rabi frequency.

Apart from the desired spectrum, two important general conclusions follow from this equation, at least for the case of a moderate to weak external field. For large values of t the answer can be shown to depend only on τ and not on t [although $\langle \hat{b}_s(t) \rangle$ depends on t], so that the process behaves as a quasi-stationary process in the wide sense. Secondly, if the electromagnetic field $\hat{A}_{\text{free}}^{(+)}(0, t)$ is treated as a c number $\tilde{\epsilon}\mathbf{G} \exp[-i(\omega_1 t + \varphi)]$, then $\langle \hat{b}_s^\dagger(t)\hat{b}_s(t+\tau) \rangle$ does not fall off with τ , and the fluorescent spectrum becomes a δ function. Only the contributions from the last term in Eq. (11) that explicitly involve the field commutation relations yield exponentially decaying terms, and therefore lead to a finite spectral width. The calculation will be published elsewhere.

We conclude that certain two-time correlation functions—or spectral distributions—of the fluorescent light should be measured in the presence of an exciting field, if the noncommuting properties of the electromagnetic field are to be tested.²⁴ Not even the time evolution of the light intensity in resonance fluorescence reflects explicit quantum features of the field. The experimental study of spontaneous emission appears to be much less appropriate as a test of QED.

*Work supported by the National Science Foundation.

¹M. Newstein, Phys. Rev. **167**, 89 (1968).

²M. Dillard and H. R. Robl, Phys. Rev. **184**, 312 (1969).

- ³B. R. Mollow, Phys. Rev. **188**, 1969 (1969); B. R. Mollow and M. M. Miller, Ann. Phys. (New York) **52**, 464 (1969).
- ⁴G. S. Agarwal, Phys. Rev. A **2**, 2038 (1970), and in *Springer Tracts in Modern Physics*, edited by G. Höhler (Springer, Berlin, 1974), Vol. 70, p. 111.
- ⁵C. S. Chang and P. Stehle, Phys. Rev. A **4**, 641 (1971).
- ⁶C. R. Stroud, Jr., Phys. Rev. A **3**, 1044 (1971).
- ⁷B. R. Mollow, Phys. Rev. A **5**, 1969, 2217 (1972).
- ⁸I. R. Senitzky, Phys. Rev. A **6**, 1171, 1175 (1972).
- ⁹R. K. Bullough, in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 121.
- ¹⁰J. R. Ackerhalt, P. L. Knight, and J. Eberly, Phys. Rev. Lett. **30**, 456 (1973); J. R. Ackerhalt and J. H. Eberly, Phys. Rev. D **10**, 3350 (1974); see also L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975), Chap. 7.
- ¹¹P. W. Milonni, Ph.D. thesis, University of Rochester, 1974 (unpublished).
- ¹²M. E. Smithers and H. S. Freedhoff, to be published.
- ¹³H. J. Carmichael and D. F. Walls, to be published.
- ¹⁴E. T. Jaynes and F. W. Cummings, Proc. IEEE **51**, 89 (1963); M. D. Crisp and E. T. Jaynes, Phys. Rev. **179**, 1253 (1969); C. R. Stroud and E. T. Jaynes, Phys. Rev. A **1**, 106 (1970); E. T. Jaynes, in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 35. See also the related work of G. Lanyi, Phys. Rev. D **8**, 3413 (1973).
- ¹⁵V. Weisskopf and E. Wigner, Z. Phys. **63**, 54 (1930); V. Weisskopf, Phys. Z. **34**, 1 (1933).
- ¹⁶W. Heitler and S. T. Ma, Proc. Roy. Irish Acad. **52**, 109 (1949).
- ¹⁷F. Haake, Phys. Rev. A **3**, 1723 (1971).
- ¹⁸I. R. Senitzky, Phys. Rev. Lett. **31**, 1955 (1973); P. W. Milonni, J. R. Ackerhalt, and W. A. Smith, Phys. Rev. Lett. **31**, 958 (1973); P. W. Milonni and W. A. Smith, Phys. Rev. A **11**, 814 (1975); G. S. Agarwal, Phys. Rev. A **10**, 717 (1974).
- ¹⁹J. M. Wessner, D. K. Anderson, and R. T. Robiscoe, Phys. Rev. Lett. **29**, 1126 (1972); in this connection, see also the much older work of E. Gaviola, Nature (London) **122**, 772 (1928); see also H. M. Gibbs, Phys. Rev. Lett. **29**, 459 (1972), and Phys. Rev. A **8**, 446, 456 (1973).
- ²⁰We distinguish Hilbert-space operators by the caret.
- ²¹Such treatments have been discussed by G. W. Series, in *Proceedings of the International Conference on Optical Pumping and Atomic Lineshape*, edited by T. Skalinski (PWN—Polish Scientific Publishers, Warsaw, 1969), p. 25; R. K. Nesbet, Phys. Rev. Lett. **27**, 553 (1971), and Phys. Rev. A **4**, 259 (1971); W. L. Lama and L. Mandel, Phys. Rev. A **6**, 2247 (1972), and in *Coherence and Quantum Optics*, edited by L. Mandel and E. Wolf (Plenum, New York, 1973), p. 273.
- ²²R. J. Glauber, Phys. Rev. **131**, 2766 (1963).
- ²³H. J. Kimble and L. Mandel, to be published.
- ²⁴Some experimental results have been reported by F. Schuda, C. R. Stroud, Jr., and M. Hercher, J. Phys. B: Proc. Phys. Soc., London **7**, L198 (1974); W. Hartig and H. Walther, Appl. Phys. **1**, 171 (1973); W. Lange, J. Luther, B. Nottbeck, and H. W. Schröder, Opt. Commun. **8**, 157 (1973).

Very Long ³He Nuclear Relaxation Times at 4 K Using Cryogenic Coatings

R. Barbé, F. Laloë, and J. Brossel

Laboratoire de Spectroscopie Hertzienne de l'École Normale Supérieure, F 75231 Paris Cédex 05, France

(Received 2 December 1974)

Longitudinal ³He nuclear relaxation times at low temperature have been measured by an optical-pumping technique, using solid H₂ coatings on the cell walls. Relaxation times longer than 2 days at 4.2 K were obtained with a 3-cm-diam cell. Preliminary results with other coatings (solid D₂, Ne, etc.) are reported.

Although the first optical-pumping experiment was performed with an atomic beam,^{1,2} it is often better to confine the atoms, ions, or molecules under study in a closed cell. This leads to more

intense and narrower resonance lines. The problem then is to find a suitable transparent container so as to minimize the effect of atom-wall collisions on the relaxation times of the relevant