

Experimental Limitations to the Validity of Semiclassical Radiation Theories*

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The polarization correlation of photons emitted in an atomic cascade is treated in the framework of a general semiclassical radiation theory. The resulting predictions are contrasted with those of orthodox quantum-radiation theory. Experimental evidence is discussed which is in agreement with the latter theory, but excludes semiclassical theories in general.

It is well known that calculations in a fully quantum-mechanical treatment of electromagnetic radiation lead to divergent expressions. Semiclassical theories which treat matter quantum mechanically, and radiation classically, are conspicuously free of these difficulties. This fact has recently led several authors¹⁻⁷ to investigate the possibility that semiclassical theories may warrant consideration in their own right as a general formalism for the description of radiation, matter, and their interaction. These authors with varying degrees of emphasis have suggested that the predictions of such a theory are compatible with all existing experimental data of atomic physics. Jaynes and his collaborators,¹⁻³ in particular, claim that to first order the theories are indistinguishable.

The successes of such a formalism are impressive, to be sure. Many effects, whose origins have long been thought to be intimately tied to a quantization of the radiation field, have been re-derived (or were originally derived) in a semiclassical framework. These include (a) spontaneous emission,¹⁻⁴ (b) absorption and stimulated emission,¹⁻⁴ (c) resonance fluorescence,^{1-3,5} (d) photoelectric effect,^{5,8} (e) Compton effect,⁷ (f) Lamb shift,¹⁻³ (g) vacuum polarization,⁸ and (h) blackbody radiation spectrum.¹⁻⁵

It is the purpose of this paper to show that there is at least one first-order effect that cannot be predicted by a semiclassical theory. Further, it will be shown that currently existing experimental data from two independent experiments exclude semiclassical theories in general. Thus there is justification for the usual caution with which such theories are treated. More important, they cannot be considered a solution to the problem of divergences in quantum electrodynamics.

In what follows we first discuss one of these experiments and its predictions from orthodox quantum theory of radiation. Next we calculate the predictions of a general semiclassical theory for the same experiment. Finally, we compare both with the experimental results. A second relevant experiment, discussed earlier by Bohm and Aharonov in a similar context, is included in Appendix A.

KOCHER-COMMINS EXPERIMENT

The pertinent experiment is one that measures the polarization correlation of photons emitted successively in an atomic cascade. Such an experiment has been performed by Kocher and Commins.^{9,11} In their experiment, the emitted photons were selected by interference filters and detected individually with photomultiplier tubes, as shown in Fig. 1. This experiment utilized a three-level cascade in calcium. The cascade proceeded from a $J=0$ level to a degenerate $J=1$ level, and terminated in a $J=0$ level, which is the atomic ground state.

In this process radiation, emitted at frequencies ν_A and ν_B , is passed by the two optical interference filters A and B , respectively. The polarization of the optical photons was measured by linear polarizers of the sheet type. For each decay there is a finite probability for both photomultipliers to register a count in coincidence. Kocher and Commins measured this coincidence rate as a function of relative polarizer orientation.

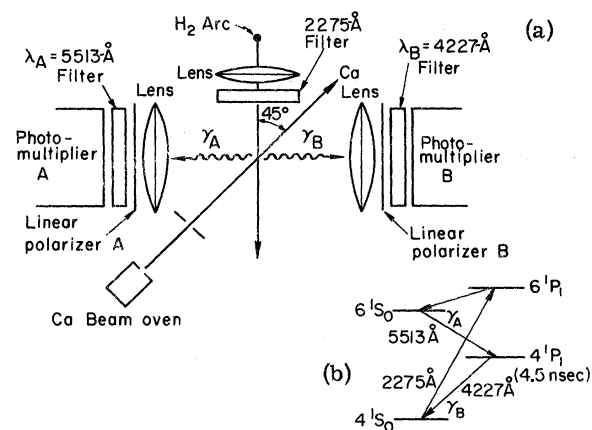


FIG. 1. (a) Schematic diagram of apparatus of Kocher and Commins. (b) Partial level scheme for calcium (after Kocher and Commins).

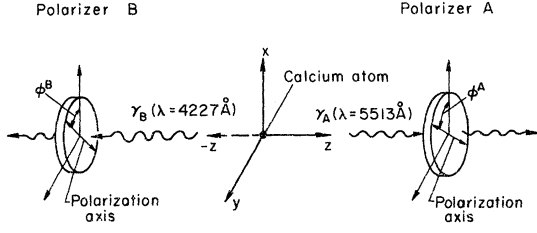


FIG. 2. Coordinate system for description of cascade-photon polarizations.

QUANTUM-MECHANICAL CALCULATION OF COINCIDENCE RATE

Consider two photons emitted into the polarizer-detector systems of the Kocher-Commins experiment. For simplicity of demonstration, assume the detectors subtend infinitesimal solid angle. Take the position of the emitting atom as the origin, with the detectors on the $+z$ and $-z$ axes (see Fig. 2). Use the ket vector $|\hat{p}_i, \mu\rangle$ to denote the photon state. Here $\hat{p} = R, L, x, y$ refers to the photon's polarization being right or left circular or linear in the x or y direction. The term $\mu = \pm$ denotes the photon's propagation direction along the $+z$ or $-z$ axes. Finally, $i = A$ or B refers to the photon's having, respectively, frequency ν_A or ν_B .

The most general polarization state for the two photons is given by

$$\begin{aligned}
 |2 \text{ photons}\rangle = & a |R_A, +\rangle |L_B, +\rangle + b |L_A, +\rangle |R_B, +\rangle \\
 & + c |R_A, -\rangle |L_B, -\rangle + d |L_A, -\rangle |R_B, -\rangle \\
 & + e |R_A, +\rangle |R_B, -\rangle + f |L_A, +\rangle |L_B, -\rangle \\
 & + g |R_A, -\rangle |R_B, +\rangle + h |L_A, -\rangle |L_B, +\rangle. \quad (1)
 \end{aligned}$$

The initial and final atomic states have zero angular momentum and the same parity, and are unaffected by a 180° rotation about the y axis. Similarly, the two-photon state must have zero angular momentum and even parity, and be unaffected by a rotation of 180° about the y axis. The only state satisfying these requirements is

$$\begin{aligned}
 |2 \text{ photons}\rangle = & \alpha (|R_A, +\rangle |L_B, +\rangle + |L_A, +\rangle |R_B, +\rangle \\
 & + |R_A, -\rangle |L_B, -\rangle + |L_A, -\rangle |R_B, -\rangle) \\
 & + \beta (|R_A, +\rangle |R_B, -\rangle + |L_A, +\rangle |L_B, -\rangle \\
 & + |R_A, -\rangle |R_B, +\rangle + |L_A, -\rangle |L_B, +\rangle). \quad (2)
 \end{aligned}$$

The two photons are distinguishable by their energies. The optical interference filter in the $+z$ direction transmits only photons of frequency ν_A . Likewise, in the $-z$ direction only photons of frequency ν_B are transmitted. Outside of these filters, then, the state vector is¹¹

$$|2 \text{ photons}\rangle = (1/\sqrt{2}) (|R_A\rangle |R_B\rangle + |L_A\rangle |L_B\rangle). \quad (3)$$

The redundant propagation-direction specification has been dropped, and the correct normalization inserted. A change of basis to linear-polarization states allows the state vector to be written

$$|2 \text{ photons}\rangle = (1/\sqrt{2}) (|x_A\rangle |x_B\rangle + |y_A\rangle |y_B\rangle). \quad (4)$$

The joint linear-polarization measurement made by polarizers oriented at angles ϕ_A and ϕ_B to the x axis projects the state of Eq. (4) onto the two linear-polarization states

$$|\phi^A\rangle = \cos\phi^A |x_A\rangle + \sin\phi^A |y_A\rangle$$

and

$$|\phi^B\rangle = \cos\phi^B |x_B\rangle + \sin\phi^B |y_B\rangle. \quad (5)$$

Thus, the coincidence probability in the Kocher-Commins experiment is proportional to

$$\begin{aligned}
 P_{\text{coinc}} \propto & |\langle\phi^A| \langle\phi^B| 2 \text{ photons}\rangle|^2 \\
 = & \frac{1}{4} + \frac{1}{4} \cos 2(\phi^A - \phi^B), \quad (6)
 \end{aligned}$$

with the idealization that the detectors subtend infinitesimal solid angle.

A more complete calculation,¹² which acknowledges the finite solid angle of the detectors and the nonideal efficiencies of the polarizers, yields, for the probability of a coincidence,

$$\begin{aligned}
 P_{\text{coinc}} \propto & \frac{1}{4} (\epsilon_M^A + \epsilon_m^A) (\epsilon_M^B + \epsilon_m^B) \\
 & + \frac{1}{4} (\epsilon_M^A - \epsilon_m^A) (\epsilon_M^B - \epsilon_m^B) F_1(\delta) \cos 2(\phi^A - \phi^B), \quad (7)
 \end{aligned}$$

where ϵ_M^A and ϵ_m^B are the maximum transmissions of the polarizers, ϵ_m^A and ϵ_m^B are their minimum transmissions, and $F_1(\delta)$ is a function of the half-angle δ subtended by the detectors. For the geometry of the Kocher-Commins experiment, $F_1 = 0.99$.

SEMICLASSICAL CALCULATION OF COINCIDENCE RATE

In a semiclassical theory, polarization effects of radiation must be described classically, since Maxwell's equations are to provide a complete description for the radiation field. Since Kocher and Commins observed coincidences, this result implies that single photons must be described as short pulses of classical electromagnetic radiation, emitted during a transition made by an atom from one state to another. The classical description implies that these pulses must have a well-defined space-time description for their electric and magnetic fields.

First consider a single such pulse put successively through a narrow bandwidth filter and a partial linear polarizer and into a fast photomultiplier tube. The photoelectric effect has been treated semiclassically by Franken⁵ and Lamb and Scully,⁶ who show that in this description the probability of electron emission is proportional to the intensity of the incident wave for low intensities. The pulse

entering the system has a probability of producing a photomultiplier count given by

$$p = \frac{1}{2} q(\epsilon_M - \epsilon_m) \cos 2(\phi - \theta) + \frac{1}{2}(q+s)(\epsilon_M + \epsilon_m). \quad (8)$$

In the above expression, ϕ , ϵ_M , and ϵ_m are, respectively, the orientation, maximum transmission, and minimum transmission of the polarizer, and θ is the orientation of the linear polarization of the pulse. Here q , θ , and s are classical random variables. For p to be a sensible normalized probability, we require that $0 \leq q, s \leq 1$.

Now consider the two pulses of radiation emitted during a $J=0 \rightarrow 1 \rightarrow 0$ atomic cascade. In the semiclassical theory of Jaynes *et al.*,¹⁻³ an atom exact in the highest level is in unstable equilibrium. A perturbation, perhaps associated with the excitation itself, gives it an admixture of other states and initiates a cascade.¹³ The perturbation determines the degree and direction of the linear polarization of the emitted pulses, and will vary from one emission to the next. According to this theory the radiation reaction field first accelerates and then damps the radiation process.

In any general semiclassical theory there must exist an asymmetry defining the polarization of a $J=0 \rightarrow 1$ transition.¹⁴ Since the radiation is to be treated classically, the parameters describing the polarization of the pair of emitted pulses over an ensemble of such pairs must be classical random variables with a classical joint probability density. Denote this by

$$\rho(\theta_A, q_A, s_A; \theta_B, q_B, s_B) \equiv \rho(\omega).$$

The probability of a delayed coincidence count in the two photomultiplier tubes of the Kocher-Commins experiment may be calculated by using the usual methods of classical probability theory:

$$\begin{aligned} p(\phi^A, \phi^B) &= \frac{1}{4} \int [q_A(\epsilon_M^A - \epsilon_m^A) \cos 2(\phi^A - \theta_A) + (q_A + s_A)(\epsilon_M^A + \epsilon_m^A)] \\ &\quad \times [q_B(\epsilon_M^B - \epsilon_m^B) \cos 2(\phi^B - \theta_B) \\ &\quad + (q_B + s_B)(\epsilon_M^B + \epsilon_m^B)] \rho(\omega) d\omega. \quad (9) \end{aligned}$$

In the experiment of Kocher and Commins, data were taken keeping the relative orientation of the polarizers fixed, and averaging over rotations of the pair. When the above expression is similarly averaged, we have

$$\begin{aligned} p(\phi^A - \phi^B) &= \frac{1}{8} (\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B) [\cos 2(\phi^A - \phi^B) \\ &\quad \times \int q_A q_B \cos 2(\theta_A - \theta_B) \rho(\omega) d\omega + \sin 2(\phi^A - \phi^B) \\ &\quad \times \int q_A q_B \sin 2(\theta_A - \theta_B) \rho(\omega) d\omega] + \frac{1}{4} (\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) \\ &\quad \times \int (q_A q_B + q_A s_B + q_B s_A + s_A s_B) \rho(\omega) d\omega. \quad (10) \end{aligned}$$

Equation (10) can be put in the form

$$p(\phi^A - \phi^B) = \frac{1}{8} (\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B)$$

$$\begin{aligned} &\times \langle q_A q_B \rangle_{av} C \cos 2(\phi^A - \phi^B - \xi) \\ &\quad + \frac{1}{4} (\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) (\langle q_A q_B \rangle_{av} + D), \quad (11) \end{aligned}$$

where $0 \leq C, D, \langle q_A q_B \rangle_{av} \leq 1$. It is clear from Eq. (11) that a semiclassical theory predicts that the ratio of minimum-to-maximum coincidence rate must always be greater than

$$\left(\frac{R_{\min}}{R_{\max}} \right)_{\text{sc}} \geq \frac{(\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) - \frac{1}{2}(\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B)}{(\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) + \frac{1}{2}(\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B)}. \quad (12)$$

On the other hand, conventional quantum mechanics predicts from Eq. (7),

$$\left(\frac{R_{\min}}{R_{\max}} \right)_{\text{qm}} = \frac{(\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) - (\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B) F_1}{(\epsilon_M^A + \epsilon_m^A)(\epsilon_M^B + \epsilon_m^B) + (\epsilon_M^A - \epsilon_m^A)(\epsilon_M^B - \epsilon_m^B) F_1}. \quad (13)$$

The factors of $\frac{1}{2}$ rendering the two expressions different¹⁵ arise from a neglect in the semiclassical results of interference terms of one photon with the other. It is one of the most curious predictions by the quantum theory that this polarization interference persists, even when the photons are remote from each other, and have different frequencies. A discussion of this point may be found in Appendix A, along with mention of a second relevant experiment performed by Wu and Shaknov.

A detailed calculation of the dynamics of the $J=0 \rightarrow 1 \rightarrow 0$ cascade is performed in Appendix B in the specific semiclassical formalism of Jaynes *et al.* It yields predictions consistent with Eq. (11) for any set of initial conditions.

Inserting the measured efficiencies of the polarizers used in the Kocher-Commins experiment¹⁶ into Eqs. (12) and (13), one gets

$$(R_{\min}/R_{\max})_{\text{sc}} \geq 0.45; \quad (R_{\min}/R_{\max})_{\text{qm}} = 0.15.$$

The experimental result

$$(R_{\min}/R_{\max})_{\text{expt}} = 0.15 \pm 0.02$$

decisively contradicts the predictions of semiclassical theories.

CONCLUSION

The polarization correlation of photons emitted in a $J=0 \rightarrow 1 \rightarrow 0$ atomic cascade has been treated within the framework of a general semiclassical radiation theory. The predictions differ from those of an orthodox quantum-mechanical description of the electromagnetic radiation field. Results of the Kocher-Commins experiment (and the Wu-Shaknov experiment) are discussed; they appear to exclude semiclassical theories in general. Nor does it appear likely that a simple modification to these theories can bring them into agreement with experimental data. Only two fundamental assumptions have been required for this discussion:

- (i) For a classical electromagnetic wave of any

incident polarization, the intensity that is transmitted by a linear polarizer varies as $a + b \cos^2(\theta - \phi)$.

(ii) The probability of electron emission at a photomultiplier cathode is proportional to the incident intensity.

Both of these assumptions are rather well tested experimentally for classical electromagnetic radiation, and evidently cannot be modified within the framework of Maxwell's equations. The experiment of Kocher and Commins is, however, undergoing current refinement at this and other laboratories in an attempt to test general neoclassical theories of matter and radiation which do not stay within the framework of Maxwell's equations.¹² These results will be reported elsewhere.^{12a}

APPENDIX A: WU-SHAKNOV EXPERIMENT AND HYPOTHESIS STUDIED BY FURRY

In a famous paper¹⁷ Einstein, Podolsky, and Rosen (EPR) discussed correlations in spatially isolated systems. They pointed out that quantum theory predicts that such systems may interfere with each other despite their spatial separation. The nonlocal nature of this interference, they believed, is an unreasonable behavior for physical systems; it suggested that the correlations must arise through an incompleteness of the theory. Bohr in reply¹⁸ showed that such correlations give rise to no observable effects that are contrary to physical experience, and that quantum mechanics, in spite of the objections of EPR, can be considered a complete description of physical phenomena.

At the same time, Furry pointed out¹⁹ that a suitably modified theory in which this interference was eliminated necessarily led to significantly different predictions. He hypothesized that, in this modified theory, after the two systems had ceased to interact, each somehow evolved into a definite state. Following this evolution, the systems are in a correlated "mixture" of states, rather than a correlated "pure" state.

Bohm and Aharonov,²⁰ much later, appealed to experiment to test this hypothesis. They considered the Wu-Shaknov experiment,²¹ which was a measurement of the polarization correlation of γ rays emitted during the annihilation of singlet positronium. Their analysis, similar to the one in this work, showed that the observed correlation has the magnitude predicted by quantum theory, and not that of the hypothesis discussed by Furry.

A moment's reflection will convince the reader that a semiclassical treatment of the joint polarization of cascade photons is simply an alternative form of this same hypothesis. The classical electromagnetic waves emitted in a semiclassical description of a cascade cannot interfere with each

other. They have different frequencies, and will not pass each other's narrow bandwidth filter. Moreover, the nonlocal character of quantum-mechanical interference to which EPR objected so vehemently is quite foreign to classical electromagnetic radiation.

Thus the Wu-Shaknov experiment can also serve to test semiclassical theories. The Kocher-Commins experiment is used in this work, since it is a system for which semiclassical theories are more easily discussed, and for which exact solutions can be obtained.

Finally, a comment is warranted concerning the relation of this result to the measurement problem in quantum mechanics. It is well known that difficulties are encountered when one attempts to include the apparatus and the observer within the quantum formalism.²² The problem is associated with interfacing the classical domain of the observer with the quantum domain of the radiating atom. It has been shown that the quantum formalism must not be applicable at some point in the measurement chain which stretches from the atom through the observer. Semiclassical theories attempt to solve this difficulty by placing the transition from the quantum domain to the classical domain at the point where radiation is emitted. The above analysis demonstrates the error in such an attempt.

APPENDIX B: $J=0 \rightarrow 1 \rightarrow 0$ CASCADE IN SEMICLASSICAL SCHEME OF JAYNES *et al.*

In this Appendix we shall solve exactly the equations of motion proposed by Jaynes *et al.*¹⁻³ for a $J=0 \rightarrow 1 \rightarrow 0$ atomic cascade. Consider a system that has three levels and five states, labeled as shown in Fig. 3. The above system is assumed to start in a definite pure state, whose wave function is represented by a superposition of the wave func-

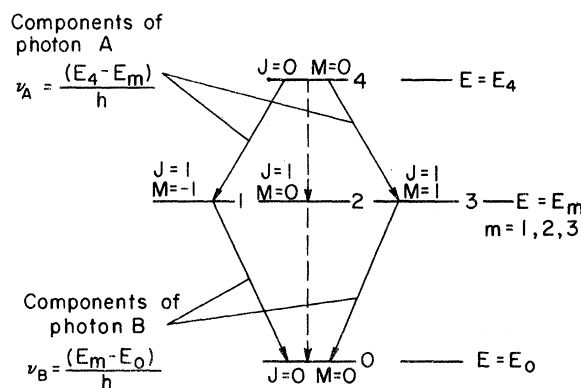


FIG. 3. States participating in cascade with appropriate labels, energy, and total angular momentum indicated. Transitions indicated by dashed lines are not observed in coordinate system chosen, as they give rise to longitudinal polarizations.

tions of the five states,

$$\psi(x, t) = \sum_{j=0}^4 a_j(t) \psi_j(x), \quad (\text{B1})$$

which satisfy Schrödinger's equation

$$\mathcal{H}^{\text{atom}} \psi_j = E_j \psi_j. \quad (\text{B2})$$

The time-dependent density-matrix elements are defined in the usual way as

$$\sigma_{im}(t) = a_i(t) a_m^*(t) \quad (\text{B3})$$

and a slowly varying part

$$\rho_{im} = \sigma_{im} e^{i\omega_{im}t}, \quad (\text{B4})$$

where

$$\omega_{im} = (E_i - E_m)/\hbar \quad (\text{B5})$$

and

$$\omega_{4m} \equiv 2\pi\nu_A \quad \text{and} \quad \omega_{m0} \equiv 2\pi\nu_B \quad \text{for } m=1, 2, 3.$$

The equations of motion for this five-state system coupled to the radiation field in the absence of the external fields are given by^{2, 3, 23}

$$\dot{\rho}_{im} = -\sum_j [\frac{1}{2}(A_{ij} - A_{jm}) - i(\Gamma_{ij} - \Gamma_{jm})] \rho_{jj} \rho_{im}, \quad (\text{B6})$$

where

$$A_{im} = -A_{mi} = 4\vec{\mu}_{im} \cdot \vec{\mu}_{mi} \omega_{im}^3 / 3\hbar c^3, \quad (\text{B7})$$

$$\Gamma_{im} = \Gamma_{mi} = \frac{e^2 \hbar}{2\pi^2 m^2 c^2} \iint \hat{\mathbf{k}} \times [\hat{\mathbf{k}} \times \langle l | e^{i\hat{\mathbf{k}} \cdot \mathbf{r}} \vec{\nabla} | m \rangle \cdot \hat{\mathbf{k}} \times [\hat{\mathbf{k}} \times \langle m | e^{-i\hat{\mathbf{k}} \cdot \mathbf{r}} \vec{\nabla} | l \rangle] d\Omega dk, \quad (\text{B8})$$

and

$$\vec{\mu}_{im}(t) = \int \psi_i^*(t) e^{-i\vec{x} \cdot \vec{\nabla}} \psi_m(t) d^3x. \quad (\text{B9})$$

These equations can be solved exactly if the Einstein A coefficient for transitions from the highest state to the intermediate state is the same as that for transitions from them to the ground state.²⁴ Thus we set

$$A_{4m} = A_{m0} = A \quad \text{and} \quad \Gamma_{4m} = \Gamma_{m0} = \Gamma \quad \text{for } m=1, 2, 3,$$

and the solutions for the diagonal elements of the slowly varying part of the density matrix $\rho_{jj}(t)$ in terms of the initial conditions $\rho_{jj}(t_0)$ are

$$\begin{aligned} \rho_{44}(t) &= \frac{1}{2} - \beta_0 \tanh[\beta_0 A(t - t_0 - t_{44})], \\ \rho_{33}(t) &= \rho_{33}(t_0) \rho'(t), \\ \rho_{22}(t) &= \rho_{22}(t_0) \rho'(t), \\ \rho_{11}(t) &= \rho_{11}(t_0) \rho'(t), \\ \rho_{00}(t) &= \frac{1}{2} + \beta_0 \tanh[\beta_0 A(t - t_0 - t_{00})]. \end{aligned} \quad (\text{B10})$$

In the above we have defined

$$\rho'(t) = \frac{\beta_0 \tanh[\beta_0 A(t - t_0 - t_{44})] - \beta_0 \tanh[\beta_0 A(t - t_0 - t_{00})]}{\rho_{33}(t_0) + \rho_{22}(t_0) + \rho_{11}(t_0)},$$

$$\beta_0 = \left[\frac{1}{2} - \rho_{00}(t_0) \rho_{44}(t_0) \right]^{1/2},$$

$$t_{44} = \frac{1}{A\beta_0} \tanh^{-1} \left(\frac{\rho_{44}(t_0) - \frac{1}{2}}{\beta_0} \right),$$

and

$$t_{00} = -\frac{1}{A\beta_0} \tanh^{-1} \left(\frac{\rho_{00}(t_0) - \frac{1}{2}}{\beta_0} \right).$$

For consistency with a probability interpretation, the initial conditions must satisfy the requirements

$$\rho_{jj}(t_0) \geq 0 \quad \text{and} \quad \sum_j \rho_{jj}(t_0) = 1.$$

The solutions for the off-diagonal elements are

$$\rho_{4j}(t) = \rho_{4j}(t_0) \cosh(\beta_0 A t_{44}) \operatorname{sech}[\beta_0 A(t - t_{44} - t_0)] \times (\text{oscillating terms}), \quad (\text{B11})$$

$$\rho_{j0}(t) = \rho_{j0}(t_0) \cosh(\beta_0 A t_{00}) \operatorname{sech}[\beta_0 A(t - t_{00} - t_0)] \times (\text{oscillating terms}).$$

The oscillating terms are identified as effectively generating a Lamb shift in radiating states.²⁵

The effective electric-dipole moment of the atom is given by^{2, 26}

$$\vec{M}(t) = \sum_{i < m} \operatorname{Re}(\rho_{mi} \vec{\mu}_{im} e^{i\omega_{im}t}). \quad (\text{B12})$$

In this expression, the electric-dipole moment is to be interpreted as due to an objectively existent charge distribution, whose oscillation generates the classical radiation field. This notion will be recognized as the old Schrödinger interpretation of quantum mechanics.

Following Franken⁵ and Lamb and Scully,⁶ we assume that the probability of a photomultiplier count is proportional to the time average of the intensity of the classical electromagnetic wave transmitted by the interference filter and the linear polarizer. Thus the system accepting light with frequency ν_A will register a count with probability proportional to the time average of the square of the component of the electric-dipole moment parallel to the linear polarizer direction ϕ^A . From (B11) and (B12) this is given by²⁷

$$p_A \propto r_{34}^2 + r_{14}^2 + 2r_{34}r_{14} \cos(\theta_{34} - \theta_{14} - 2\phi^A), \quad (\text{B13})$$

where $r_{im} \geq 0$ and θ_{ij} are both real and are defined by

$$\rho_{im}(t_0) = r_{im} e^{i\theta_{im}}.$$

Similarly, for frequency ν_B , we have

$$p_B \propto r_{03}^2 + r_{01}^2 + 2r_{01}r_{03} \cos(\theta_{03} - \theta_{01} + 2\phi^B). \quad (\text{B14})$$

Finally, the probability of a delayed coincidence is proportional to

$$p_{\text{coinc}} \propto \rho_{00}(t_0) \rho_{11}(t_0) \rho_{33}(t_0) \rho_{44}(t_0) \times \left(2[1 + \cos 2(\phi^B - \phi^A - \xi)] + \frac{\rho_{33}(t_0) + \rho_{11}(t_0)}{\rho_{11}(t_0) + \rho_{33}(t_0)} \right), \quad (\text{B15})$$

where

$$\xi = (E_0 + E_1 + E_3 + E_4)t_0 h^{-1} + \arg[a_0(t_0)a_1(t_0)a_3(t_0)a_4(t_0)].$$

Thus, for each decay the ratio of the minimum-to-maximum coincidence rate is given by

$$\frac{R_{\min}}{R_{\max}} = \frac{\rho_{33}(t_0)/\rho_{11}(t_0) + \rho_{11}(t_0)/\rho_{33}(t_0)}{4 + \rho_{33}(t_0)/\rho_{11}(t_0) + \rho_{11}(t_0)/\rho_{33}(t_0)} \geq \frac{1}{3}; \quad (\text{B16})$$

equality holds when $\rho_{11} = \rho_{33}$.

Relation (12), with the assumption of ideal

polarizers ($\epsilon_M = 1$, $\epsilon_m = 0$) yields the same lower limit for this ratio. Thus, as expected, the theory of Jaynes *et al.* is consistent with the more general results derived above.

Finally, it should be remarked that since (B15) holds for any initial pure state, this lower limit must be valid for an average over any statistical mixture of pure states. Hence, the assumed form for the density matrix is not critical.

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¹³When exactly in the highest level, the atom has no electric-dipole moment, but a small perturbation will create an asymmetry, and allow the resulting dipole moment to grow in time.

¹⁴For radiation to be treated classically, a $J=0$ atom must somehow be given a preferred direction to determine the polarization of the emitted radiation. Indeed,

the light from a $J=0 \rightarrow 1$ transition appears unpolarized when an ensemble of photons is observed. However, that each photon is polarized can be seen from the Kocher-Commins experiment. If additional optical path length is introduced so that the second photon of the cascade is observed first, the first photon must now be polarized parallel to the second. Moreover, existence of the second-half of the cascade is not necessary for this to be true. One could perform an experiment in which the atom itself, following the emission of a photon in a $J=0 \rightarrow 1$ transition, is state selected and detected in coincidence with the emitted photon. In this case the photon is still predicted to have a measurable polarization. Irrelevantly of the mechanism giving the atom its preferred direction, however, the arguments still apply. For an alternative possible origin of this asymmetry see Ref. 12.

¹⁵In the limit of infinitesimal-detector solid angle, the limit $F_1=1.0$ applies. The second terms in the numerators and denominators then differ by exactly a factor of $\frac{1}{2}$. This comparison is valid, since the lower limit for semiclassical theories holds for arbitrary solid angle.

¹⁶ $\epsilon_M^A=0.80$, $\epsilon_m^A=0.06$, $\epsilon_M^B=0.75$, and $\epsilon_m^B=0.06$ (see Ref. 11).

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²³Equation (B6) can be shown to apply despite the degeneracy of the intermediate level.

²⁴This specialization is approximately true for the cascade of the Kocher-Commins experiment, and will not affect the resulting polarization in any case. Hence, comparison of these predictions with the results of Kocher and Commins is valid.

²⁵In this formalism, it is the radiating states only that are Lamb-shifted with respect to nonradiating ones. See Refs. 2 and 3.

²⁶See Ref. 3, Eqs. (63) and (64).

²⁷Ideal polarizers and detectors with infinitesimal solid angle are assumed. Realizable polarizers and a finite solid angle will yield a lower amplitude correlation.