Spontaneous Decay of Coherently Excited Rb

Hyatt M. Gibbs
Bell Laboratories, Murray Hill, New Jersey 07974
(Received 24 April 1972; revised manuscript received 24 July 1972)

The observed dependence of Rb fluorescence upon the time after excitation and upon the area of the coherent optical pulse is consistent with quantized electromagnetic field theory and inconsistent with the neoclassical theory of radiation.

Quantum electrodynamics (QED) predicts that the incoherent[1] resonance fluorescence F from a two-level system is a maximum when only the excited state is occupied. A pure excited state can be produced in a sharp-line absorber by a short coherent optical pulse of area $\theta = (2p/\hbar) \times \int_{-\infty}^{+\infty} \mathcal{E}(t)\,dt$ equal to $\pi, 3\pi, 5\pi, \cdots$, where p is the electric dipole moment of the transition and $\mathcal{E}(t)$ is the slowly varying amplitude of the electric field.[2] The fluorescence is zero when only the ground state is occupied, for instance after excitation by a short pulse of area 0π, 2π, 4π, etc. This oscillatory dependence of the fluorescence upon input area has been demonstrated in Rb vapor under conditions closely approximating an ideal two-level system.[3] This demonstration of a simple coherent optical effect could be extended to the measurement of dipole moments, relaxation times, and lifetimes and may find application in the study of 0π pulses and chirping.[3] In addition, it disproves the neoclassical theory (NCT) of radiation in which the electromagnetic field is not quantized. NCT assumes that the expectation of the dipole-moment operator is an actual dipole moment which radiates according to classical electrodynamics.[4] NCT predicts fluorescence minima for input areas near $\pi, 2\pi, 3\pi, 4\pi, \cdots$, since in that theory a pure state is unable to radiate spontaneously.[4] No minima in F were observed at odd multiples of π, in contradiction with NCT.[5] NCT also predicts that the time dependence of the fluorescence deviates substantially from a single exponential decay if the average effective θ exceeds 45°; no deviations from single exponentials were observed.

The QED fluorescence rate is given by[4,5] $F_{\mathrm{QED}}(t) = (N_2\hbar\omega/\tau_{ab})\rho_{aa}(t)$, where N_2 is the number of two-level atoms with population $\rho_{aa}(t)$ in the excited state a. Spontaneous emission from state a to state b (with $\hbar\omega$ less energy) is characterized by the spontaneous emission rate[2] $\tau_{ab}^{-1} = \frac{8}{3}(\omega^3/\hbar c^3)p^2$; then $\dot{\rho}_{aa}(t) = -\rho_{aa}/\tau_{ab}$, in the absence of external fields. Coherent excitation through a tipping angle θ in a time short compared to relaxation times yields, in the sharp-line limit,

$$\rho_{aa}(t) = (\sin\tfrac{1}{2}\theta)^2 e^{-t/\tau_{ab}}, \quad \int_0^T F_{\mathrm{QED}}(t)\,dt \propto (\sin\tfrac{1}{2}\theta)^2.$$

The fluorescence is then a single exponential decay in time and an oscillatory function of pulse area.

The corresponding NCT equations are

$$F_{\mathrm{NCT}}(t) = (N_2\hbar\omega/\tau_{ab})|\rho_{ab}|^2 = (N_2\hbar\omega/\tau_{ab})\rho_{aa}\rho_{bb}, \quad \dot{\rho}_{aa} = \rho_{aa}\rho_{bb}/\tau_{ab}, \quad \rho_{aa}(t)\rho_{bb}(t) = \tfrac{1}{4}\mathrm{sech}^2[(t-t_m)/2\tau_{ab}],$$

$$t_m = \tau_{ab}\ln[\rho_{aa}(0)/\rho_{bb}(0)] = 2\tau_{ab}\ln(\tan\tfrac{1}{2}\theta), \quad \int_0^T F_{\mathrm{NCT}}(t)\,dt = \tanh(t_m/2\tau_{ab}) - \tanh[(t_m - T)/2\tau_{ab}].$$

$F(t)$ and $\int F(t)\,dt$ are shown in Figs. 1 and 2. For weak excitation (small θ) the two theories agree, but for areas between $\frac{1}{4}\pi$ and $\frac{7}{4}\pi$ the differences are considerable. An initial pure excited state is unnecessary to detect large discrepancies. The shape of the NCT decay depends upon the initial excitation.

The experimental system was an approximation to the ideal two-level system.[6] A single-transverse-mode, single-longitudinal-mode, pulsed ^{202}Hg II laser was locked within ± 2 MHz to the center of the 15-MHz residual-Doppler-absorption profile of a beam (0.27 mm thick, $\approx 10^{10}$ atoms/cm^3) of natural Rb in a 75-kOe magnetic field. A 6-nsec coherent optical pulse was extracted from the 1-μsec laser pulse by a Pockels cell and weakly focused to ≈ 200 μm diam, yielding a pulse of area up to $\approx 4\pi$. A uniform plane wave was approximated by imaging (and magnifying by ≈ 2) the excitation region upon a 100-μm output aperture which eliminated fluorescence not emanating from the uniform central portion. Because of the low absorption ($\approx 5\%$ total, $\lesssim 30\%$ peak), a solid angle of $(0.3\times 10^{-3})4\pi$, and the uniform-plane-wave aperture, 10^6 input photons

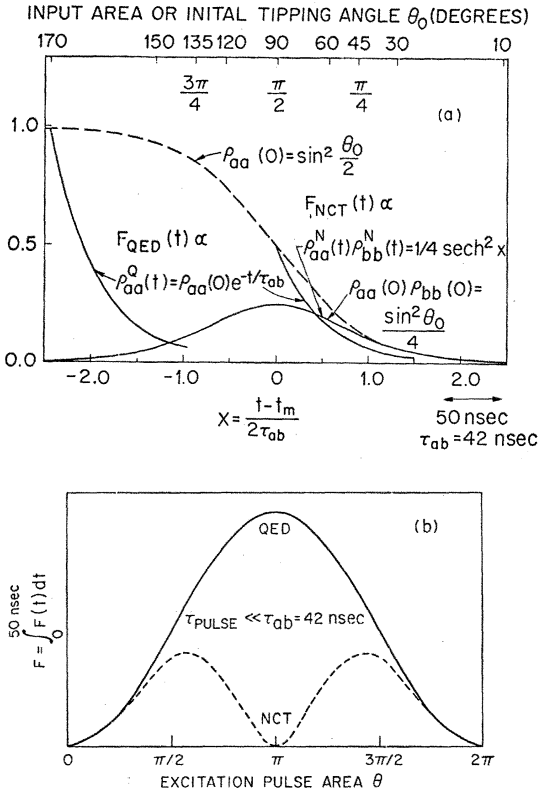

FIG. 1. (a) Comparison of the time dependence of the fluorescence from a sharp-line, two-level transition for a quantized (QED) and unquantized (NCT) electromagnetic field. (b) By observing fluorescence for only a fixed time interval after the excitation in which most of the QED fluorescence occurs, one excludes that portion ($\pi/2<\theta<3\pi/2$) of F_{NCT} which is considerably delayed.

were required for each fluorescence photon reaching the single-photon counting system (RCA 8552, time-to-height converter and coincidence circuitry, and PDP-8 computer).

The experimental system is very nearly in the sharp-line limit. But the spontaneous emission occurring during the 6-nsec pulse [$\approx$ 10 nsec full width at half-maximum for $\mathcal{E}(t)$ with a negative tail persisting to 35 nsec] to the two lower levels b and c is not negligible; see Figs. 2 and 3. These departures from the ideal two-level system were included in a computer simulation[6] of the experiment with

$$F_{QED}=(N_3\hbar\omega/\tau_{ab})\rho_{aa}(t),\quad \rho_{aa}+\rho_{bb}+\rho_{cc}=1,$$

$$\rho_{bb}(0)=\rho_{cc}(0)=\tfrac{1}{2},\quad (\dot\rho_{aa})_{SE}=-\rho_{aa}/\tau_{ab}-\rho_{aa}\tau_{ac},$$

$$(\dot\rho_{ab})_{SE}=-\rho_{ab}/2\tau_{ab}-\rho_{ab}/2\tau_{ac};$$

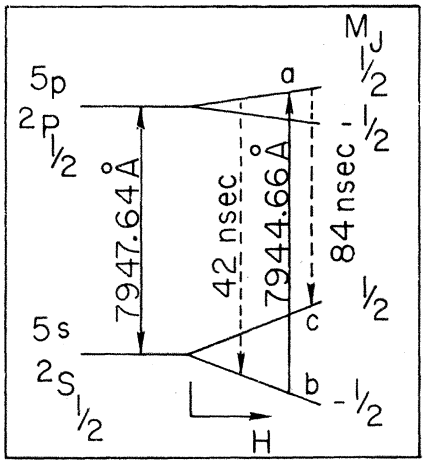

FIG. 2. Simplified Rb energy-level diagram.

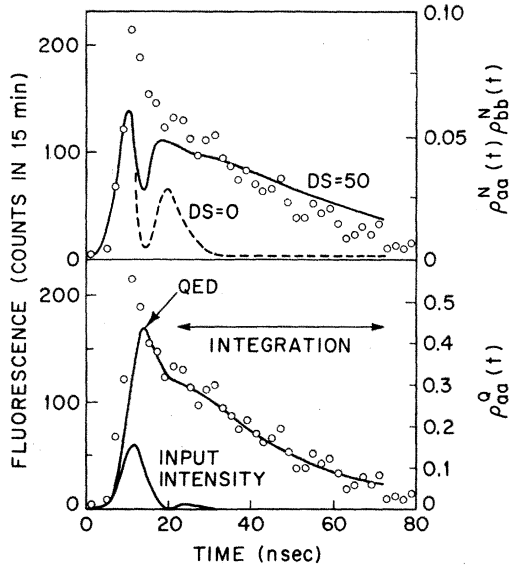

FIG. 3. Comparison of $F_{NCT}\propto\rho_{aa}{}^N(t)\rho_{bb}{}^N(t)$ and $F_{QED}\propto\rho_{aa}{}^Q(t)$ with experimental data for an input pulse area of π. Reflections of the excitation pulse into the detector added counts to the fluorescence signal in the 5–15-nsec region. The theoretical curves are computer simulations for the actual three-level system including 15-MHz absorption width, finite absorption, and the actual input pulse. DS refers to dynamic shift in MHz; NCT predicts the possibility of a DS of the atom's resonance frequency as it is excited. The DS for Rb would be difficult to calculate with confidence, so all values have been considered. The data clearly are not consistent with the NCT, DS=0 simulation using the equations in the text. The addition of a 50-MHz shift, which gives the best fit to the data of Fig. 5, gives a better but still poor fit. The QED and NCT, DS=50 curves are normalized to equal the 35-nsec datum point for which the external field is back to zero.

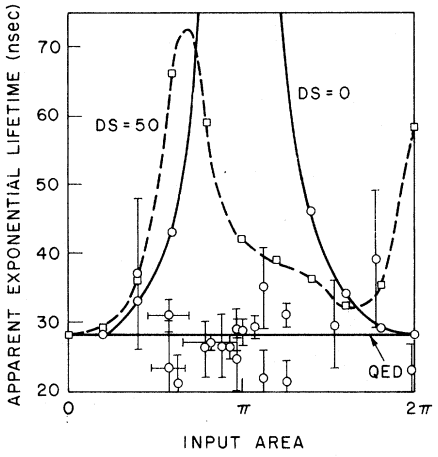

FIG. 4. Comparison of observed apparent exponential decay times with QED and NCT predictions. The experimental uncertainty in area is no more than ± 10% unless shown otherwise. The data and predictions were fitted by a single expontial over the range 40–70 nsec.

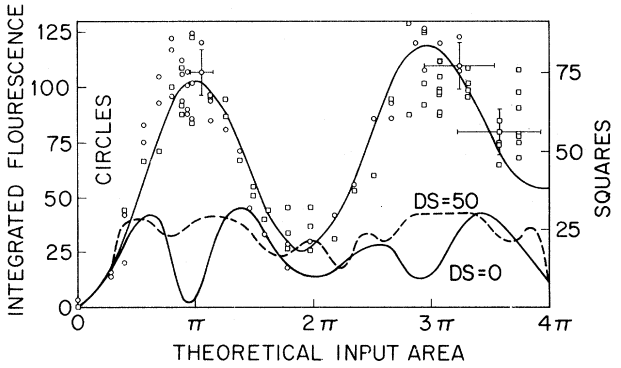

FIG. 5. Fluorescence from a to b integrated from 22 to 72 nsec as a function of input area. The squares and circles are counts in 100- and 80-sec intervals for peak cw absorption coefficients of 0.21 and 0.32, respectively. The QED curve is normalized to yield a minimum weighted variance with the circled points of 2.2. The NCT fluorescence is on the same scale as that of QED, so they agree for small θ. The weighted variance was a minimum of 16 for DS = 50 and 70 for DS = 0.

$\tau_{ab} = 42$ nsec, $\tau_{ac} = 84$ nsec, and $N_3 = 2N_2$, with N_2 atoms in b and in c initially. By NCT,[4]

$$(\dot{\rho}_{aa})_{SE} = -\rho_{aa}\rho_{bb}/\tau_{ab} - \rho_{aa}\rho_{cc}/\tau_{ac};\quad (\dot{\rho}_{ab})_{SE} = (\rho_{aa}-\rho_{bb})\rho_{ab}/2\tau_{ab} - \rho_{ab}\rho_{cc}/2\tau_{ac};\quad F_{NCT} = (N_3\hbar\omega/\tau_{ab})\rho_{aa}\rho_{bb};$$

τ_{ab} and τ_{ac} are respectively taken equal to 21 and 42 nsec in order that $\rho_{aa} = -\rho_{aa}/(28\text{ nsec})$ for weak excitation, in agreement with observations.[7] The computer simulation and experimental time dependences of the a-to-b fluorescence are shown in Fig. 3 for an input pulse area of π. The apparent exponential decay times as a function of input area are given in Fig. 4. Only the QED simulation agrees with the data. The fluorescence, integrated from 22 to 72 nsec as shown in Fig. 3, is displayed as a function of input area in Fig. 5. That the first minimum is for $\theta = 2\pi$ and not π was verified by observing the characteristic features of self-induced transparency with the same pulse in an optically thick beam. Again the results are seen to be consistent only with QED.

The NCT simulation, even with an arbitrary dynamic shift, is unable to account for the observed integrated fluorescence or apparent exponential lifetime as a function of input pulse area. In addition (and independent of this experiment), one must take a radiative lifetime of 14 nsec, in contradiction with the 28-nsec theoretical value, in order to explain the observed 28-nsec fluorescence decay time following weak excitation.

This demonstration of a simple coherent optical effect then disproves the neoclassical theory of spontaneous emission.[9] It also suggests the use of this technique in studying pulse areas and transition moments and relaxation times in the short-time regime where less frequency stability would be required.[3] In fact, anything altering the fluorescence as a function of input area, such as degeneracies, chirps, relaxation, or nonuniform-plane-wave conditions could potentially be studied by this resonance-fluorescence technique.

The technical assistance of G. Churchill and helpful discussions with M. D. Crisp, E. T. Jaynes, S. L. McCall, G. Salamo, R. E. Slusher, and C. R. Stroud, Jr., are gratefully acknowledged.

[1]Coherent resonance fluorescence is calculated to be $\lesssim 10\%$ of the incoherent fluorescence even at $\theta = \frac{1}{2}\pi$; see R. H. Dicke, Phys. Rev. 93, 99 (1954); I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. 141, 391 (1966); D. C. Burnham and R. Y. Chiao, Phys. Rev. 188, 667 (1969); F. T. Arecchi and E. Courtens, Phys. Rev. A 2, 1730 (1970). However, the coupled Bloch and Maxwell equations used in the computer simulation include coherent resonance fluorescence [E. L. Hahn, N. S. Shiren, and S. L. McCall, Phys. Lett. 37A, 265 (1971)].

[2]Abella, Kurnit, and Hartmann, Ref. 1; S. L. McCall and E. L. Hahn, Phys. Rev. 183, 457 (1969). The value of the dipole moment is derived in H. M. Gibbs and R. E. Slusher, Phys. Rev. Lett. 24, 638 (1970).

[3]H. P. Grieneisen, N. A. Kurnit, and A. Szöke, Opt. Commun. 3, 259 (1971). In this reference is described a similar experiment in which the oscillations of F versus θ are almost averaged out by level degeneracies and broad-line absorption.

[4]M. D. Crisp and E. T. Jaynes, Phys. Rev. 179, 1253 (1969), Eq. (30) in particular; C. R. Stroud, Jr., and E. T. Jaynes, Phys. Rev. A 1, 106 (1970); D. Leiter, Phys. Rev. A 2, 259 (1970); E. T. Jaynes, Phys. Rev. A 2, 260 (1970).

[5]Several articles claiming to disprove the NCT have appeared during this experiment: R. K. Nesbet, Phys. Rev. Lett. 27, 553 (1971); R. K. Nesbet, Phys. Rev. A 4, 259 (1971); J. F. Clauser, Phys. Rev. A (to be published); F. R. Nash and J. P. Gordon, to be published. These papers reanalyze previous experiments using NCT. The present experiment has the advantage of being the experiment suggested by Jaynes; it also demonstrates the validity of QED in a new regime.

[6]Many details are contained in Gibbs and Slusher, Ref. 2; R. E. Slusher and H. M. Gibbs, Phys. Rev. A 5, 1634 (1972); H. M. Gibbs and R. E. Slusher, "Sharp-Line Self-Induced Transparency" (to be published).

[7]This implies an electric dipole moment smaller by $\sqrt{2}$, in disagreement with theoretical calculations [O. S. Heavens, J. Opt. Soc. Amer. 51, 1058 (1961)] and the power required for a 2π self-induced transparency pulse (Ref. 6).

Neoclassical Diffusion in an Elliptical Torus

H. R. Strauss
Center for Plasma Physics and Thermonuclear Research, The University of Texas at Austin, Austin, Texas 78712
(Received 26 June 1972)

Neoclassical transport theory is applied to a plasma in an axisymmetric toroidal magnetic field whose flux surfaces have elliptical cross sections. The contribution of the poloidal field component is important and leads to a large fraction of trapped particles.

Neoclassical theory generalizes the classical diffusion process of plasma in a magnetic field to include drift-orbit effects, which become important at low collision frequencies. The model magnetic field used in most calculations is an axisymmetric toroidal field, with a small poloidal component providing circular flux surfaces.[1-3] This is a good representation of the magnetic field of a Tokamak.

It has been pointed out that there are advantages to building Tokamaks with elliptical, rather than circular, cross sections.[4,5] The stability conditions may be satisfied with a lower toroidal field B_φ for a given plasma current I. Elliptical cross sections can be made with $l=0$ windings above and below the plasma. This raises the possibility of an axisymmetric divertor.

In the present note we investigate neoclassical diffusion in a Tokamak with elliptical cross section. First we show that the calculations using circular magnetic surfaces may be trivially extended to the elliptic case, provided the assumption $\Theta \ll 1$ is retained. Here Θ is the ratio of poloidal to toroidal magnetic field.

It is just this assumption which is violated in the interesting case of $i/2\pi \sim 1$ and large ellipticity, where i is the rotational transform. In the limit $\Theta \gg 1$, most of the particles are trapped by the poloidal-field variation. We can obtain results analogous to those of Hazeltine[6] for the "fat torus."

To study toroidal magnetic surfaces with elliptic cross section, we introduce elliptic coordinates. Starting with cylindrical coordinates (R, Z, φ) we set

$$R - R_0 = \rho \cos\omega, \quad Z = \kappa\rho \sin\omega, \tag{1}$$

such that

$$(R - R_0)^2 + Z^2/\kappa^2 = \rho^2. \tag{2}$$

A magnetic surface is described by ρ = constant. The constant κ is the ratio of the axes of the ellipse. When $\kappa = 1$ we have circular surfaces.

The magnetic field has the form

$$\vec{B} = (I/R)\vec{\varphi} + \nabla\psi \times \vec{\varphi}/R, \tag{3}$$

where I and ψ are functions of ρ only.

The rotational transform divided by 2π is given by[7]

$$i/2\pi = d\psi/d\chi, \tag{4}$$

where $\psi(\chi)$ is the poloidal (toroidal) magnetic