

## Single-Pulse Superfluorescence in Cesium

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Superfluorescence is reported from an initially inverted, pure two-level system. Single-pulse emission occurs at low densities; neither homogeneous relaxation nor inhomogeneous dephasing nor linear diffraction can explain the absence of ringing. At higher densities multiple-pulse outputs occur. The experimental results are not in good quantitative agreement with present theories.

Superfluorescence<sup>1-3</sup> (SF) is the cooperative emission of a sample initially prepared in a completely inverted state. A carefully prepared transition in Cs is shown to possess near-ideal characteristics for observing SF. A regime of single-pulse emission is reported. It is the purpose of this Letter to describe the experiment and its results in sufficient detail to allow a meaningful comparison with various theories.

For pencil-shaped volumes large compared to the wavelength  $\lambda$ , early theories predicted radiation in a single pulse of squared hyperbolic secant shape,<sup>4,5</sup> with peak power proportional to the square of the density directed only along the sample axis in both directions. Arecchi and Courtens<sup>6</sup> emphasized that for a given density  $n$  and sample length  $L$  only a finite length  $L_c$  of the sample would be able to radiate cooperatively; for  $L > L_c$  the sample would break up into independently radiating parts.

The first SF experiment on rotational transitions in hydrogen fluoride demonstrated a dramatic reduction of the radiation time, highly directional emission, and  $n^2$  dependence of the peak power.<sup>7</sup> However, the output pulses clearly showed ringing which was attributed to Burnham-Chiao ringing.<sup>8,9</sup>

The question remained whether single-pulse output might be observed under conditions specified by Bonifacio and co-workers<sup>3</sup> for "pure" SF: (a) a pure two-level system; (b) cross-sectional area  $A$  of the sample such that the Fresnel number  $F = A/\lambda L$  equals 1; (c) a number of characteristics obeying the inequalities  $\tau_E < \tau_C < \tau_R < \tau_D < T_1$ ,  $T_2'$ ,  $T_2^*$  and  $\tau_P \ll \tau_D$ , where  $\tau_E = L/c$ ,  $\tau_C = (\tau_E \tau_R)^{1/2}$ ,  $\tau_D$  is the delay time,  $T_1$  and  $T_2'$  are the longitudinal and transverse homogeneous relaxation times,  $T_2^*$  is the inhomogeneous de-

phasing time, and  $\tau_P$  is the excitation pulse length. The SF time  $\tau_R$  is equal to  $\tau_0/\mu N$ , where  $\tau_0$  is the spontaneous-emission decay time,  $N$  is the total number of initially excited atoms, and  $\mu$  is the shape factor.<sup>5</sup> For Fresnel numbers of 1 or more,  $\tau_R \approx 8\pi\tau_0/3n\lambda^2L$ .<sup>5,10</sup> Recent SF experiments<sup>11</sup> do not satisfy these conditions, and so they provide no answer to the above question.

The present experiment in Cs has been undertaken to study the SF output under a wide range of experimental conditions; see Fig. 1 and Table I. In particular, the requirements<sup>3</sup> for "pure" SF have been met on the Cs  $7P_{3/2}$  to  $7S_{1/2}$  2931-nm transition: (a) In a transverse magnetic field<sup>12</sup> of about 2.8 kOe and with  $\sigma$  polarization of the pump, the sublevel  $7P_{3/2}(m_J = -\frac{3}{2}, m_I = -\frac{5}{2})$  is populated selectively; it can superfluoresce only to the final level  $7S_{1/2}(m_J = -\frac{1}{2}, m_I = \frac{5}{2})$ , emitting  $\sigma$ -polarized radiation. From the relevant dipole moments<sup>13</sup> it can be concluded that the competing

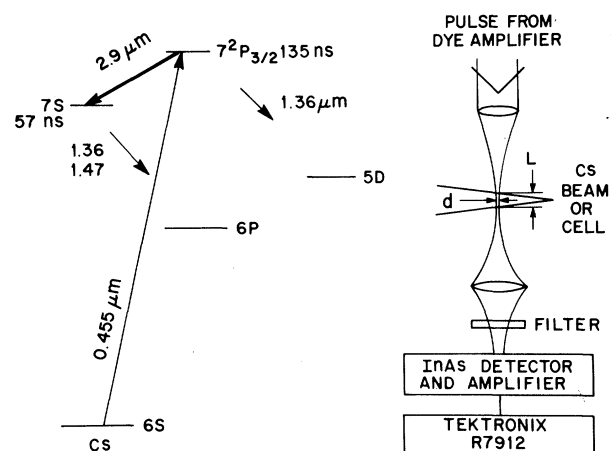


FIG. 1. Schematic diagram of cesium level scheme and of the experimental apparatus.

TABLE I. Survey of experimental conditions. The amplitude gain at the center of the atomic line is  $\alpha L = T_2/\tau_R$ . All times are in nanoseconds. The full width of the pump beam at  $I_{\max}/e$  is  $d$ .

	$L$ (cm)	$d$ ( $\mu\text{m}$ )	$T_2^*$	$\tau_E$	$\tau_R$	$\tau_D$	$\alpha L$
Cell	5.0	432	5	0.17	0.15-1	6-20	35-5
Beam	3.6	366	18	0.12	0.1-1.8	5-35	180-10
Beam	2.0	273	32	0.07	0.15-1.3	6-25	215-25
Cell	1.0	193	5	0.035	0.12-0.5	5-12	45-10

and cascading transitions are unimportant. A true two-level transition results. (b) The sample consists of Cs atoms in a cell or in an atomic beam of variable length; a Fresnel number close to 1 for the SF wavelength is realized by adjusting the diameter of the pump beam. (c) Finally, the time inequalities are satisfied:  $\tau_E = 0.067$  ns,  $\tau_c = 0.18$  ns,  $\tau_R = 0.5$  ns,  $\tau_D = 10$  ns,  $T_1 = 70$  ns,  $T_2' = 80$  ns, and  $T_2^* = 32$  ns;  $\tau_p$  is 2 ns. The values of  $T_1$ ,  $T_2'$ , and  $\tau_0 = 551$  ns are calculated from the lifetimes and branching ratios of the relevant levels.<sup>14</sup> The sample length and inversion density are adjusted to determine  $\tau_E$  and  $\tau_R$ . In a cell,  $T_2^* = 5$  ns from Doppler broadening. By use of an atomic beam  $T_2^*$  was lengthened to 32 ns, including contributions from magnetic-field inhomogeneity.

The  $7P_{3/2}$  level is excited from the ground state  $6S_{1/2}$  with a dye-laser pulse of 2 ns duration and 500 MHz bandwidth at 455 nm (Fig. 1). The pump has a peak intensity on axis of about  $10 \text{ kW/cm}^2$ . The transverse intensity has been studied by projecting the beam on a TV camera tube and by scanning a narrow slit through the beam. The profile was always smooth and nearly Gaussian.

Accurate knowledge of the initial excited density is of prime importance in the comparison with theory. The determination of that density constitutes a major difficulty in this experiment and probably in any experiment on SF. The atomic density in the cell or beam was measured carefully,<sup>15</sup> and the excited-state density was then calculated assuming complete saturation of the pump transition.<sup>16</sup> Excited-state densities quoted are accurate to  $(-40, +60)\%$ .

The SF output is detected with an InAs detector of 1-ns response time (Judson J 12 LD), an Avante amplifier, and a Tektronix Transient Digitizer R7912. The SF source has a diameter of less than 0.5 mm; it is imaged onto the detector with a magnification of 0.25. Since the diameter of the detector is 0.15 mm, essentially all of the

energy emitted in the forward direction is collected. For the experiments with  $F = 1$  described here, the energy is found to be emitted into a solid angle close to the diffraction-limited value. Care is taken to avoid feedback from windows, filters, and lenses. Pulses of comparable energy were observed simultaneously in the forward and backward directions with equal delay times. Quantitative estimates indicate that at least 20% of the stored energy is emitted in each direction.

Normalized output pulse shapes and delay times are shown in Figs. 2 and 3. Single pulses are always observed for delay times beyond 7 ns. For shorter delay times, multiple pulses occur with shapes which fluctuate greatly from pulse to pulse even for the same delay time of the first pulse. The observed value of  $\tau_R$  at which the transition from single pulses to multiple pulses

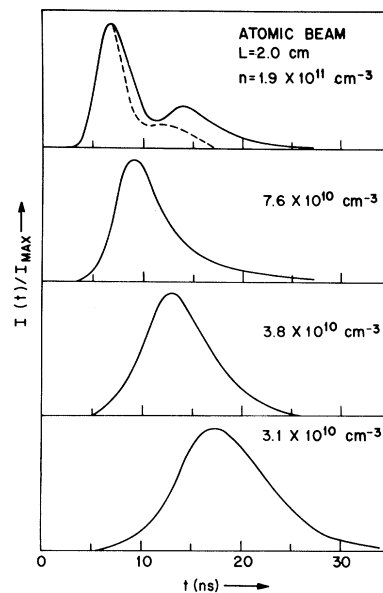


FIG. 2. Normalized single-shot pulse shapes for several densities  $n$ . Uncertainties in the values of  $n$  are estimated to be  $(+60, -40)\%$ .

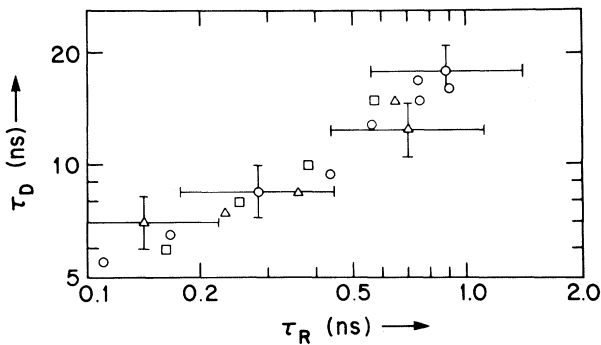


FIG. 3. Delay time  $\tau_D$  of the pulse as a function of  $\tau_R$ . Triangles, 2.0-cm beam; circles, 3.6-cm beam; squares, 5.0-cm cell.

takes place is approximately  $2\tau_E$ , i.e.,  $L \approx L_c$ . The fact that multiple-pulse emission appears rather abruptly suggests that it is different from Burnham-Chiao ringing which changes very slowly with density.<sup>8,9,17</sup> Whether or not it is related to Arecchi-Courtens<sup>6</sup> cooperature-length effects or Bonifacio-Lugiato<sup>3</sup> oscillatory SF remains to be established.

The occurrence of single pulses cannot be explained by the destruction of coherent ringing by relaxation processes. In a 2-cm atomic beam, single pulses are observed for a delay time of 8 ns, 4 times smaller than  $T_2^*$  and nearly 10 times smaller than  $T_1$  and  $T_2'$ . Furthermore, the peak power of the output pulses has been studied for an atomic beam of 3.6 cm and has been found to be proportional to  $\tau_D^{-2}$  for delay times between 8.5 and 40 ns, without any indication of accelerated decay due to dephasing. Hence dephasing is certainly negligible in the atomic-beam experiments. On the other hand, multiple pulses are found in a 5-cm cell for delay times of 10 ns, twice as large as  $T_2^*$ . So the atomic-beam single pulses are certainly not multiple pulses averaged into one by dephasing. It is important to note that the pulses shown in Fig. 2 are the most common shapes, but single, almost symmetric, squared hyperbolic-secant-pulses do occur frequently. They are the narrowest pulses observed, and so the asymmetric pulses may be symmetric pulses smeared by transverse effects but not vice versa.

A detailed comparison of the data with the many existing theories is outside the scope of this Letter. A few brief remarks may suffice. These data agree qualitatively with the Bonifacio-Lugiato<sup>3</sup> mean-field theory in that a single-pulse regime is observed which gives way to multiple-

pulse emission for  $L \approx L_c$ . The observed widths are about  $10\tau_R$  compared with the mean-field prediction of  $3.5\tau_R$ . The mean-field theory neglects entirely spatial variations within the sample which lead to pulse broadening and ringing according to plane-wave Maxwell-Schrödinger simulations. In such simulations the buildup from the quantum noise of spontaneous emission is often incorporated in an effective initial tipping  $\theta_0$  of the polarization vector or input pulse of small area. Both the delay and the amount of ringing depend logarithmically on  $\theta_0$ ; a small value of  $\theta_0$  gives long delays and strong ringing.<sup>8,17</sup> Theoretical predictions of  $\theta_0$  for our case range from  $3 \times 10^{-6}$  (Ref. 9) up to 0.08 (Ref. 5). Numerical solutions reproduce both width and delay of the single pulses for  $\theta_0 \approx 10^{-2}$ , with some ringing left. Even with allowance for the uncertainty in the density, a fairly large value of  $\theta_0$  seems unavoidable.

The numerical simulations do not reproduce the experimental data in detail for any  $\theta_0$ . There are possible explanations. First, it is not clear that the quantum-classical transition can be reduced to a single parameter  $\theta_0$ . Second, dynamical<sup>18</sup> transverse effects have not been included but could be important.

In summary, SF has been observed under near-ideal conditions, and a regime of single-pulse SF has been found and studied. It is hoped that these data will be useful in refining theoretical treatments of SF pulse shapes and that future experiments will exhibit features arising from the intrinsically quantum initiation.

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<sup>1</sup>R. H. Dicke, Phys. Rev. **93**, 99 (1954).

<sup>2</sup>For a general introduction see L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley, New York, 1975).

<sup>3</sup>R. Bonifacio and L. A. Lugiato, Phys. Rev. A **11**, 1507 (1975), and **12**, 587 (1975), and references therein.

<sup>4</sup>V. Ernst and P. Stehle, Phys. Rev. **176**, 1456 (1968).

<sup>5</sup>N. E. Rehler and J. H. Eberly, Phys. Rev. A **3**, 1735 (1971).

<sup>6</sup>F. T. Arecchi and E. Courtens, Phys. Rev. A **2**, 1730 (1970).

<sup>7</sup>N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. **30**, 309 (1973); I. P. Herman, J. C. MacGillivray, N. Skribanowitz, and M. S. Feld, in *Laser Spectroscopy*, edited by R. G. Brewer and A. Mooradian (Plenum, New York, 1974).

<sup>8</sup>D. C. Burnham and R. Y. Chiao, Phys. Rev. **188**, 667 (1969); S. L. McCall, thesis, University of California, 1968 (unpublished).

<sup>9</sup>J. C. MacGillivray and M. S. Feld, Phys. Rev. A **14**, 1169 (1976).

<sup>10</sup>A numerical integration of Eq. (5.16) of Ref. 5 gives  $\mu = 0.78 [(3/8\pi)/(\lambda^2/A)]$  for  $\lambda = 2931$  nm,  $L = 2$  cm, and  $F = 1$ , but this 0.78 correction has not been applied to any value of  $\tau_R$  quoted herein.

<sup>11</sup>M. Gross, C. Fabre, P. Pillet, and S. Haroche, Phys. Rev. Lett. **36**, 1035 (1976); A. Flusberg, T. Mossberg, and S. R. Hartmann, Phys. Lett. **58A**, 373 (1976).

<sup>12</sup>Without a magnetic field several sublevels of  $7P_{3/2}$  are excited leading to beats in the SF output: Q. H. F. Vreken, H. M. J. Hikspoors, and H. M. Gibbs, Phys. Rev. Lett. **38**, 764 (1977).

<sup>13</sup>The dipole moments (Ref. 2) for the strongest transitions are, in  $10^{-18}$  esu cm,  $d_{SF} = 12$ ,  $d_{\text{pump}} = 0.5$ ,  $d_{7P-5D} = 1.75$ , and  $d_{7S-6P} = 4.4$ .

<sup>14</sup>S. Svanberg and S. Rydberg, Z. Phys. **227**, 216 (1969); P. W. Pace and J. B. Atkinson, Can. J. Phys. **53**, 937 (1975); O. S. Heavens, J. Opt. Soc. Am. **51**, 1058 (1961). The  $7P_{3/2}$  to  $7S_{1/2}$  partial lifetime is 275.5 ns; since the  $\Delta m_j = +1$  SF transition emits perpendicular to the magnetic field, the effective lifetime must be doubled. The relaxation times are calculated as follows. The upper state  $a$  of the SF transition may de-

decay to the lower state  $b$  or to other states  $c$ ;  $b$  may decay to lower states  $d$ . Using the experimental value  $\tau_a = 135$  ns, Heaven's calculated value  $\tau_b = 57$  ns, and Heaven's branching ratios, one has  $\tau_{ab} = 275.5$  ns,  $\tau_{ac} = 264.7$  ns, and  $\tau_{bd} = 57$  ns. A straightforward extension of the three-level decay formulas in R. E. Slusher and H. M. Gibbs, Phys. Rev. A **5**, 1634 (1972), yields  $1/T_1 = 1/\tau_{ab} + 1/2\tau_{ac} + 1/2\tau_{bd}$  and  $1/T_2' = 1/2\tau_a + 1/2\tau_b$  for the energy and coherence relaxation rates.

<sup>15</sup>Q. H. F. Vreken, in *Cooperative Effects in Matter and Radiation*, edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977).

<sup>16</sup>Although the pump pulse is neither coherent nor completely incoherent, it is believed that saturation occurs rather than coherent excitation. This conclusion is mainly based on the following observations: (a)  $\tau_D$  decreases monotonically with increasing pump power and converges to an asymptotic value at the highest pump power; (b) in an additional experiment the pulse duration was increased to 3 ns and the bandwidth to 1200 MHz without either quantitative or qualitative changes in the SF output pulses for a 1-cm cell.

<sup>17</sup>R. Saunders, S. S. Hassan, and R. K. Bullough, J. Phys. A **9**, 1725 (1976).

<sup>18</sup>H. M. Gibbs, B. Bölger, F. P. Mattar, M. C. Newstein, G. Forster, and P. E. Toschek, Phys. Rev. Lett. **37**, 1743 (1976).

## Stochastic Ion Heating by a Perpendicularly Propagating Electrostatic Wave

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The motion of an ion in the presence of a constant magnetic field and a perpendicularly propagating electrostatic wave with frequency several times the ion cyclotron frequency is shown to become stochastic for fields satisfying  $E/B_0 > \frac{1}{4}(\Omega/\omega)^{1/3}(\omega/\hbar)$ . This stochasticity condition is independent of how close  $\omega$  is to a cyclotron harmonic. Applications of current interest in supplementary heating of plasmas with rf power near the lower-hybrid frequency are suggested.

It is well known that a small- (infinitesimal-) amplitude electrostatic wave traveling across a constant magnetic field suffers linear damping on the ions only if its frequency  $\omega$  is an exact multiple of the ion cyclotron frequency,  $\Omega$ . At finite amplitudes of the wave, nonlinear effects become important and we can expect that this resonance is broadened. We show that a single wave leads to stochastic ion motion at an amplitude which is independent of how close  $\omega$  is to a cyclotron harmonic,  $n\Omega$ . This provides a mechanism whereby the ions in a plasma can be heated by such coherent waves.

There are two, physically distinct, nonlinear

mechanisms operative in this interaction. The first consists of a transient trapping of the ions in the potential of the wave; this leads to a rapid heating of the ions near the lower boundary of the stochastic region.<sup>1</sup> The second is due to nonlinear resonances that arise from the perturbed cyclotron motion of the ions in the wave field, and which produce stochastic ion motion in a region of ion velocity space extending from the trapping region to an upper bound determined by the field amplitude; this leads to a slower heating of the ions up to the upper boundary of the stochastic region.

In current schemes for the supplementary heat-