Quantum fluctuations in superfluorescence delay times

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Measurements are reported of the quantum fluctuations in superfluorescence delay times. The fluctuations depend strongly on the Fresnel number F of the sample. For $F \approx 1$ fair agreement with recent theories is obtained, while for larger F the fluctuations are smaller. A qualitative explanation for this F dependence is proposed. No correlation has been found between the delay times of pulses emitted from opposite ends of the same sample.

Superfluorescence (SF) is the cooperative emission of a large number of two-level atoms, initially all prepared in the excited state. For pencil-shaped samples, the power is radiated in two narrow beams along the axial directions. The intensity, very weak at first, grows rapidly to reach a peak value I_{max} after a delay time τ_D. For a given sample length I_{max} is proportional to n^2, and τ_D to $1/n$, where n is the initial number density of the excited-state atoms. These characteristic properties, first observed by Skribanowitz *et al.*[1] have been confirmed by several experiments.[2,3] Many theoretical papers have been devoted to SF since the original work of Dicke.[4] Full references can be found in the paper by Polder *et al.*[5] According to recent treatments[5,6] the SF emission can be visualized as follows. Eventually, a macroscopic coherent pulse develops. Initially, however, the evolution of the pulse is driven by the quantum fluctuations in the vacuum electromagnetic field, and in the polarization of the fully inverted atoms. As a result the macroscopic parameters of the SF pulses, such as the delay time, the peak intensity, and the pulse shape, show fluctuations. Each individual pulse represents one element out of an ensemble. The ensemble can be scanned by repeating an experiment many times with fixed initial conditions.

The quantum fluctuations are among the most interesting aspects of SF. Direct measurements of their average strength have recently been reported.[7] In this Communication experiments on the quantum fluctuations in the delay time will be described. The experiments have been performed in cesium vapor.[3] A cell of length $L = 3$ cm is placed in an oven and a homogeneous transverse magnetic field is applied. A dye-laser pulse of wavelength 455 nm, duration 1.5 to 2 ns and bandwidth about 800 MHz passes through the cell and excites a thin pencil of atoms. Delay times are measured from the peak of the pump pulse. Cesium atoms are raised from the ground-state level $6S_{1/2}$, $m_J = -\frac{1}{2}$, $m_I = -\frac{5}{2}$ to $7P_{3/2}$, $m_J = -\frac{3}{2}$, $m_I = -\frac{5}{2}$, from which SF then occurs to $7S_{1/2}$, $m_J = -\frac{1}{2}$, $m_I = -\frac{5}{2}$. The pump beam has an approximately Gaussian transverse profile. The full width at half maximum (FWHM) diameter of the pump beam d defines a nominal Fresnel number $F^* \equiv S/\lambda L$, where λ is the SF wavelength (2.931 μm) and $S = \pi d^2/4$. The actual Fresnel number of the excited volume F could not be measured directly. It is certainly larger than F^* because of the strong saturation of the pump transition. From the intensity of the pump pulse and from the divergence of the SF beam it is concluded that F exceeds F^* by a factor of 3 to 4. Values quoted for F are estimated to be accurate to within a factor of 2. The cesium density is typically adjusted for an average delay time between 7 and 10 ns. For such delay times it has been found that the peak intensities are only weakly affected by inhomogeneous broadening ($T_2^* = 5$ ns from Doppler dephasing).

In principle the measurement of the delay time fluctuations is very easy. The sample is excited repeatedly and the distribution of the delay times is collected. Unfortunately, however, the pump pulse is not completely reproducible. As a result the density of excited-state cesium atoms varies from shot to shot, and thus *extrinsic* fluctuations, due to those density variations, are superimposed on the *intrinsic* fluctuations caused by the quantum initiation. The problem has been overcome by making a two-beam experiment as illustrated in Fig. 1. The pump beam is split into two parallel beams with the help of a beam divider. Two samples are thus prepared simultaneously, and their SF delay times are measured separately by two detectors. The difference in the delay times on the same shot can be attributed to the intrinsic fluctuations alone, provided the two pump beams are identical. The relative intensity r of the two pump beams can be adjusted by a translation of the beam splitter. The latter consists of a plane parallel plate of optical quality. Its back has a reflectivity of 100%. One-half of its front is antireflection coated, whereas the reflectivity of the other half varies linearly in the direction normal to the plane of the drawing, allowing the adjustment of r mentioned above (the variation of the reflectivity over the cross

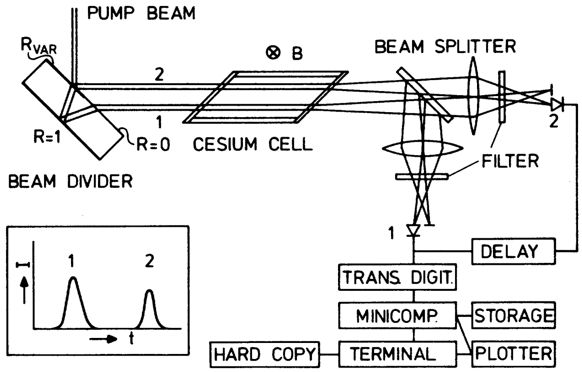

FIG. 1. Two-beam apparatus used to measure the delay time differences of SF pulses emitted by identically prepared samples. The relative intensity of the two pump beams can be adjusted with the help of the beam divider (see text).

section of the pump beam is smaller than 1%). It has been checked that the experimental results do not change significantly when r varies from 0.9 to 1.1, so that one may be confident that unintended differences in the pump beam intensities (certainly smaller than 5%) are of no importance. For each shot the two detector signals are recorded on the same trace of a Tektronix Transient Digitizer, after one of them has been suitably delayed. A waveform such as that sketched in the inset of Fig. 1 is then stored in the hard disk memory. From each trace two delay times, τ_1 and τ_2, are determined, and from these the parameters $\tau = (\tau_1 + \tau_2)/2$ and $\delta = (\tau_1 - \tau_2)$ are derived. Distributions of δ have been measured for several values of F. Examples are shown in Fig. 2. The width of the distribution depends strongly on F.

In a second set of experiments a single sample has been excited in the cell and the delay times of the SF pulses emitted in the forward and in the backward direction have been compared. In Fig. 3 the relative standard deviation of δ, $\sigma(\delta) \equiv \langle \delta^2 \rangle^{1/2} / \langle \tau \rangle$, is plotted as a function of F, both for the forward-backward and for the forward-forward (independent samples) experiments. For the two configurations the standard deviations are essentially the same; thus it may be concluded that the emissions from the two ends of the same sample are largely uncorrelated.

For a comparison with theory the distribution of the delay times τ_D arising from quantum fluctuations alone, or at least its relative standard deviation $\sigma(\tau_D)$, must be derived from the measured distribution of δ. Two complications should be considered. First, $\sigma(\delta)$ contains a contribution from instrumental effects, i.e., from the limited signal to noise of the detected pulses. Indeed, if the detectors are adjusted so that they both detect the pulses from the same beam, $\sigma(\delta)$ assumes a minimum value $\sigma_0(\delta) \approx 6\%$. A corrected value $\sigma_c(\delta)$ is obtained from $\sigma_c^2(\delta) = \sigma^2(\delta) - \sigma_0^2(\delta)$. Second, even though the delay-time differences δ are supposed to be due to quantum fluctuations alone, the distribution of δ does depend on the extrinsic fluctuations too. Fortunately it can be shown[8] that $\sigma(\delta)$ is only affected negligibly. Assuming statistical independence of the two samples one then finds $\sigma(\tau_D) = \sigma_c(\delta)/\sqrt{2}$. The following values are arrived at: $\sigma(\tau_D) = (10 \pm 2)\%$ for $F = 0.8$, $(6 \pm 2)\%$ for $F = 4$ and $< 4\%$ for $F = 18$.

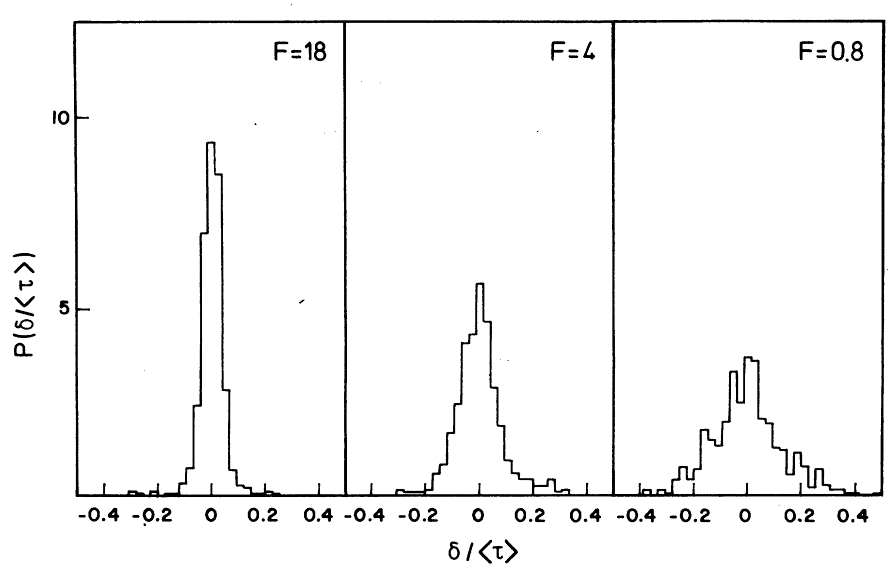

FIG. 2. Distribution of the relative delay-time differences $\delta / \langle \tau \rangle$ as observed for three values of the Fresnel number F. For easy comparison the plots have been normalized. The number of shots was 482, 393, and 468, respectively, for $F = 18$, 4, and 0.8, and the average delay time amounted to 6.8, 8.9, and 8.3 ns.

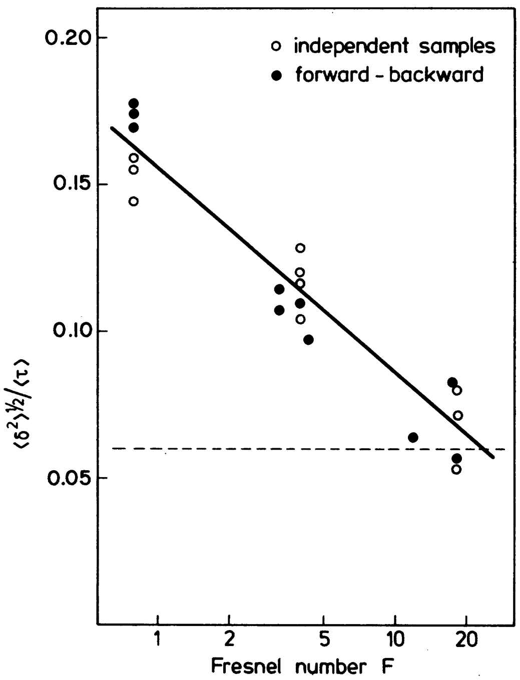

FIG. 3. The relative standard deviation in δ measured in a number of experiments for various values of the Fresnel number F. Closed circles represent pulses emitted from opposite ends of the same sample, open circles pulses emitted from two simultaneously prepared independent samples. The dashed horizontal line represents the instrumental contribution (see text).

Delay-time fluctuations in SF were first studied by Degiorgio[9] in the spirit of the mean-field treatment.[10] A value $\sigma(\tau_D)=1.2/\ln N$ was predicted, where N is the number of atoms in a volume of $F=1$. For the present experiment $1.2/\ln N=6.5\%$. More recently Polder *et al.*[5] have derived the expression $\sigma(\tau_D)=2.3/\ln N$ in the context of a quantum-mechanical theory which includes propagation effects; it amounts to 12.5% for the experiment reported here. Haake *et al.*[6,11] have numerically obtained a similar result. The above mentioned theories all assume that the amplitudes of the field and the polarization within the sample can be described by uniform plane waves propagating along the axis. It has also been assumed that the theories apply best to experiments on samples with $F=1$, the argument being that such samples will emit a diffraction-limited beam with a minimum of diffraction loss. For $F=0.8$ the experimental result is in reasonable agreement with the predictions of Refs. 5, 6, and 11. The absence of correlation between the delay times of forward and backward pulses is also to be expected. In the early stages of SF evolution the equations of motion are linear.[5,6] The amplitudes of the two oppositely traveling waves are superimposed and independent. Any correlation must arise from the interaction of the waves in the later nonlinear stages of the evolution. No strong interaction is expected because for the two waves the largest amplitudes are located near opposite ends of the sample.[12,13]

The dependence of $\sigma(\tau_D)$ on F has not been treated so far theoretically. We wish to suggest here that it is caused by the fact that for $F\geqslant 1$ the sample can radiate in several (approximately F^2) independent transverse modes. Extending the linear theory[5] to include off-axial modes we estimate that the delay-time fluctuations decrease roughly with the square root of the number of modes

$$\sigma(\tau_D)\simeq 2.3/[F\ln(N/F)] \ , \qquad (1)$$

where N is the number of atoms in a sample with $F=1$ for a given atomic density n and sample length L. The same argument also predicts that the average delay time $\langle\tau_D\rangle$ varies proportionally to $[\ln(2\pi N/F)]^2$. A reduction of $\langle\tau_D\rangle$ with increasing F at constant n and L has been reported by us before.[14] In the present experiment $\langle\tau_D\rangle$ decreased a factor 1.7 for F from 0.8 to 18, which is to be compared with a predicted factor 1.4. Substituting the experimental number $N=10^8$ into (1) one calculates $\sigma(\tau_D)=12.5\%$ for $F=1$, 3.4% for $F=4$, and 0.8% for $F=18$. The predicted dependence of $\langle\tau_D\rangle$ and $\sigma(\tau_D)$ on F is qualitatively in agreement with the experiments. Finally, it should be mentioned that Haake *et al.*[11] had predicted a marked dependence of the fluctuations on the inhomogeneous dephasing time T_2^*. Our data do not allow a test of his prediction.

In conclusion, delay-time fluctuations resulting from the quantum initiation of SF have been measured. The fluctuations depend strongly on the diameter of the sample. For the smallest Fresnel number used, $F=0.8$, the emission is nearly diffraction limited and the fluctuations are comparable in magnitude with the predictions of recent one-dimensional theories. For larger Fresnel numbers the fluctuations are significantly smaller. A qualitative explanation, based on the number of transverse modes participating in the emission, is proposed. No correlation has been found between the delay times of pulses emitted from the two opposite ends of the sample. An earlier conclusion to the contrary[15] was based on preliminary data and has been invalidated by the final results. The outcome of the present experiments emphasizes the limitations of one-dimensional theories. It is hoped that the data reported here will stimulate further theoretical studies of the three-dimensional nature of SF.

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[1]N. Skribanowitz, I. P. Herman, J. C. MacGillivray, and M. S. Feld, Phys. Rev. Lett. 30, 309 (1973).

[2]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); A. T. Rosenberger, S. J. Petuchowski, and T. A. DeTemple, in *Cooperative Effects in Matter and Radiation,* edited by C. M. Bowden, D. W. Howgate, and H. R. Robl (Plenum, New York, 1977), p. 15; A. Crubellier, S. Liberman, and P. Pillet, Phys. Rev. Lett. 41, 1237 (1978); J. Okada, K. Ikeda, and M. Matsuoka, Opt. Commun. 26, 189 (1978); 27, 321 (1978); J. Marek, J. Phys. B 12, L229 (1979); C. Brechignac and Ph. Cahuzac, J. Phys. (Paris) Lett. 40, L123 (1979); Ph. Cahuzac, H. Sontag, and P. E. Toschek, Opt. Commun. 31, 37 (1979).

[3]H. M. Gibbs, Q. H. F. Vrehen, and H. M. J. Hikspoors, Phys. Rev. Lett. 39, 547 (1977); Q. H. F. Vrehen, H. M. J. Hikspoors, and H. M. Gibbs, *ibid.* 38, 764 (1977).

[4]R. H. Dicke, Phys. Rev. 93, 99, (1954).

[5]D. Polder, M. F. H. Schuurmans, and Q. H. F. Vrehen, Phys. Rev. A 19, 1192 (1979); M. F. H. Schuurmans and D. Polder, in *Laser Spectroscopy IV,* edited by H. Walther and K. W. Rothe (Springer-Verlag, Heidelberg, 1979), p. 459.

[6]F. Haake, H. King, G. Schröder, J. Haus, and R. Glauber, Phys. Rev. A 20, 2047 (1979); F. A. Hopf, Phys. Rev. Lett. 42, 1740 (1979); F. Haake, in *Laser Spectroscopy IV,* edited by H. Walther and K. W. Rothe (Springer-Verlag, Heidelberg, 1979), p. 451.

[7]Q. H. F. Vrehen and M. F. H. Schuurmans, Phys. Rev. Lett. 42, 224 (1979); N. W. Carlson, D. J. Jackson, A. L. Schawlow, M. Gross, and S. Haroche, Opt. Commun. 32, 350 (1980).

[8]Q. H. F. Vrehen, in *Laser Spectroscopy IV,* edited by H. Walther and K. W. Rothe (Springer-Verlag, Heidelberg, 1979), p. 471.

[9]V. Degiorgio, Opt. Commun. 2, 362 (1971).

[10]R. Bonifacio and L. A. Lugiato, Phys. Rev. A 11, 1507 (1975); 12, 587 (1975).

[11]F. Haake, J. Haus, H. King, G. Schröder, and R. Glauber, Phys. Rev. Lett. 45, 558 (1980).

[12]J. C. MacGillivray and M. S. Feld, Phys. Rev. A 14, 1169 (1976).

[13]R. K. Bullough, R. Saunders, and C. Feuillade, in *Coherence and Quantum Optics IV,* edited by L. Mandel and E. Wolf (Plenum, New York, 1978), p. 263.

[14]Q. H. F. Vrehen, H. M. J. Hikspoors, and H. M. Gibbs, in *Coherence and Quantum Optics IV,* edited by L. Mandel and E. Wolf (Plenum, New York, 1978).

[15]Q. H. F. Vrehen and J. J. der Weduwe, J. Opt. Soc. Am. 70, 610 (1980).