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Quantum Beats in Superfluorescence in Atomic Cesium

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Superfluorescence has been observed at $3\ \mu\text{m}$ on the $7P$ -to- $7S$ transition in atomic cesium after 2-ns excitation of a $6S$ -to- $7P$ transition. Both in a magnetic field and in zero field, quantum beats have been observed. The beat frequencies correspond not only to initial-level splittings but also to combinations of initial- and final-level splittings. Superfluorescence beats are therefore basically different from single-atom quantum beats.

We report the first observation of quantum beats¹ in superfluorescence (SF),²⁻⁵ i.e., in the cooperative emission of an initially inverted system.⁶ SF beats and photon-echo beats^{1b} can arise either from a superposition of simultaneously excited upper levels (as for quantum beats in single-atom fluorescence^{1a}) or from the beating in the detector of the coherent emissions of two independent transitions (just as the outputs of two lasers can be mixed to produce beats).^{1b} The coherence required for beating of independent transitions is introduced by the excitation in the photon-echo case, but in the SF case it arises from the emission process itself.

SF quantum beats may have spectroscopic applications because, unlike single-atom quantum beats, they can occur at lower-state splittings as well as upper-state splittings. Also, almost all of the energy in the inverted transition is emitted along the excited cylinder and in a time much shorter than the natural radiative lifetime, yielding signals much larger than those for ordinary fluorescence. These advantages of coherent emission were also emphasized for photon-echo quantum beats.^{1b} The reduced emission time limits the accuracy of the frequency determination, but a signal increase of many orders of magnitude can be achieved on weak transitions⁴ before this limitation becomes important. The strong emission intensity in SF can lead to frequency chirps,

which must be understood before SF quantum beats can become a technique for high-resolution spectroscopy.

Quantum beats in spontaneous emission are well known and may be observed when two conditions are met.¹ First, the initial state must be prepared as a coherent superposition of substates at a well-defined point in space or time. Second, the various substates must radiate to a common final state (or states). Under such conditions the intensity of the spontaneous emission can reveal beats that correspond to level splittings of the initial state. Since the atoms decay in the absence of any driving field, accurate values for the splittings can be obtained.¹

In SF, many atoms decay coherently in their common reaction field. A coherent optical field is emitted at the frequencies of the transitions that take part in the SF process.^{1b} So the beat frequencies may reflect initial-level splittings, final-level splittings, or combinations of both. As a result, beats in SF can be quite different from those in spontaneous emission—the two conditions above need not be met.

In this Letter, the results of two different experiments are presented. In one experiment, a coherent mixture of initial substates is excited; the observed beats are well understood on the basis of calculations for the beats in spontaneous emission. In the other experiment, two initial

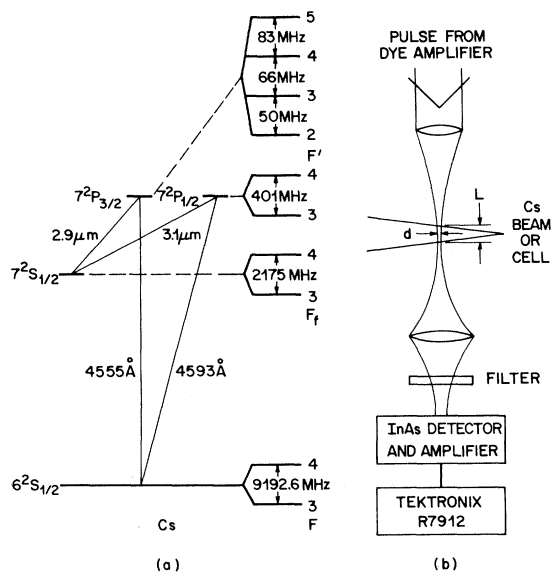


FIG. 1. (a) Relevant energy levels of Cs in zero magnetic field. See Ref. 7 for hyperfine structures. (b) Experimental apparatus.

substates are excited *incoherently* and these decay to two *different* final states. The beat frequency corresponds to the difference between the initial- and final-level splittings, which happens to be close to the final-level splitting.

The experiments are performed in Cs simply by saturating one of the $6S$ -to- $7P$ hyperfine transitions and observing SF to the $7S$ state (Fig. 1). This three-level technique⁴ results in complete population inversion. The 1–2-ns excitation pulses are produced by a N_2 -laser-pumped oscillator and amplifier with a Fabry-Perot in between for good frequency stability. Peak powers are a few hundred watts; the bandwidth is 400–500 MHz. The Cs absorber is either contained in a 30 to 100°C cell of 1 to 10 cm in length or sprayed from a 0.5×3 mm² slit in a 200 to 300°C oven containing a few grams of CS. Both in the cells and in the beam, thin cylinders are excited of Fresnel number equal to 1 within experimental error. In the atomic beam, the length of the cylinder has been varied from 1.3 to 3.5 cm. The SF signal is detected by an InAs photovoltaic detector with 2-mA bias current, resulting in a response time of about 1 ns (Judson J-12LD). The signals are amplified $22 \times$ with an Avantek 500-MHz amplifier and observed on a Tektronix R7912 transient digitizer. The pump radiation is removed with a germanium filter. Care is taken to avoid any feedback from windows, lenses, or filters. SF signals are observed simultaneously in the for-

ward and backward directions with equal delays.

The large mass of Cs and the long wavelength of the observed transitions, 2931 nm, make the Doppler broadening small and $T_2^* = 5$ ns in a cell; only weak beam collimation is needed to make $T_2^* > 20$ ns. The selected transition in Cs also has favorable radiative relaxation times, yielding $T_1 \approx 70$ ns and $T_2 \approx 80$ ns.⁸ An advantage of excitation with a wavelength much shorter than the SF wavelength⁴ is that the excitation absorption coefficient αL can easily be small even when the SF αL is large, as needed.⁹

In the first experiment, quantum beats were studied in zero magnetic field. From one of the hyperfine levels of the ground state, a coherent mixture of $7P$ hyperfine states is excited, which then radiates to the $7S$ hyperfine levels. Beat frequencies corresponding to the final-level splitting, about 2 GHz for $7S$, cannot be resolved by our detector, so the observed beats are entirely due to initial-level splittings, as in spontaneous emission. The depth and frequency of modulation depend upon the details of light polarization, the angular momenta, and the separations between the transition states. A formula for the emission intensity I_F for single-atom fluorescence for selective excitation is given by Selin and Pegg.¹⁰ Cooperative emission accelerates the decay into a SF pulse leading to beats.

In the case of D_1 excitation, the only beat frequency is the 400-MHz $7P_{1/2}$ hyperfine splitting; this modulates the SF pulse envelope [Fig. 2(a)].¹¹ As in single-atom beats, the phase of the beats is fixed by the initial phases of the coherent substates.

The case of D_2 excitation is more complicated, as previously observed for noncooperative fluorescence.¹ See Fig. 3 for $F = 3$ excitation; the fluorescence involves beats from the $7^2P_{3/2}$, $F = 2, 3, 4$ states. SF emission ceases or is greatly diminished at delay times of destructive interference. Similar effects occur for the D_2 , $F = 4$ excitation. This beating emphasizes the strong coupling of the polarization and the electric field. Destructive interference in the polarization may stop the SF emission, but it continues as soon as the polarization is constructive again.

In the second experiment, a magnetic field of 2.8 kOe was transversely applied to the cylinder excited by linearly polarized σ radiation [Fig. 4(a)]. The $M = -\frac{5}{2}$ atoms are excited and superfluoresce independently of the $M = -\frac{3}{2}$ atoms except for their coupling through the emitted field.

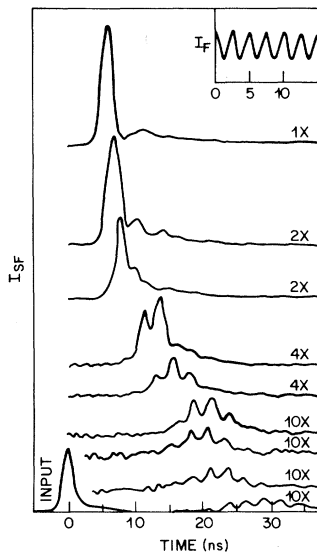


FIG. 2. Quantum beats in the superfluorescence in an atomic beam. Excitation D_1 , $F=3$, circular polarization. The inset shows I_F , the single-atom quantum beat fluorescence.

With the laser centered on the $M = -\frac{5}{2}$ transition, SF can be seen without beats. By increasing the excitation intensity until the nutation frequency exceeds the ground-state splitting of 1.3 GHz, the beats of Fig. 4(b) appear. No beats should be observable in spontaneous emission since the upper states are excited from different ground states and decay to different final states. The beat frequency measured on pulses such as Fig.

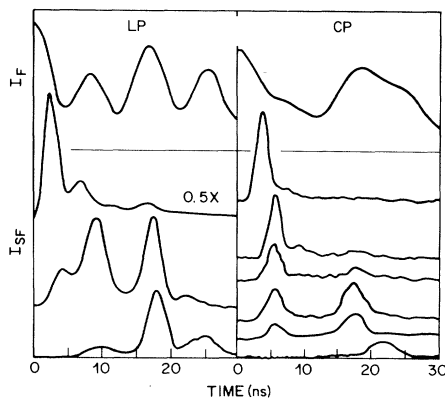


FIG. 3. Quantum beat SF with D_2 , $F=3$ excitation with linearly polarized (LP) light on a 10-cm cell and circularly polarized (CP) light on a 2.6-cm-pathlength atomic beam. The top (I_F) curves show calculated single-atom beats. The lower curves are the observed SF beats. SF is strongly suppressed near the minima in I_F . In the CP case the delays as a function of density jump discontinuously, avoiding the minimum at 12 ns.

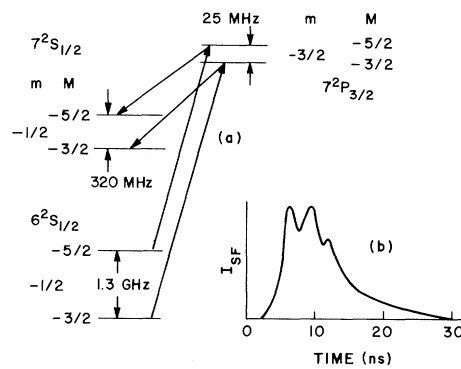


FIG. 4. (a) Relevant energy levels in a magnetic field of 2.8 kOe; m refers to the electronic and M to the nuclear magnetic quantum numbers. The $7P_{3/2}$ states are excited incoherently, namely from different ground states. (b) Quantum beats in a 2.0-cm-path-length atomic beam in 2.8 kOe.

4(b) is slightly higher than the calculated value 295 ± 5 MHz but not by as much as the ± 30 -MHz uncertainty. The phase of the beats is found to correspond to constructive interference at time zero, as might be expected.

It should be noted that in SF the atoms do not decay freely but are driven by a strong coherent electromagnetic field. Frequency chirps may occur, causing the beat frequencies in SF to deviate from those calculated from free-atom parameters. In fact, quantum beats could possibly provide a means for studying such chirps experimentally. High-resolution spectroscopy using quantum beats will require further understanding of the importance of such effects.

Although SF quantum beats may be useful for spectroscopy, they greatly complicate the analysis of SF data. Quantum beats could even be mistaken for SF ringing. We will describe a careful study of the SF of a nondegenerate two-level system in a later paper.

In summary, quantum beats have been demonstrated in superfluorescence. Such beats differ basically in nature from those in spontaneous fluorescence. They may be useful in the study of SF chirps. They may also extend the technique of quantum-beat spectroscopy to upper states with lifetimes so long that single-atom quantum-beat signals are unobservably weak, and to lower states which exhibit no single-atom quantum beats.

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Adsorbate and Substrate Characterization Using Extended X-Ray Absorption Fine Structure*

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We illustrate the usefulness of the extended x-ray absorption fine-structure effect for determining the chemical state, structure, and location of adsorbed species by a study of 0.2 monolayers of bromine adsorbed on graphite at room temperature. Bromine adsorbs as molecules with one atom fixed above the basal-plane hexagonal site while the molecular axis is free to flop around.

In this Letter, we illustrate the power of the extended x-ray absorption fine structure (EXAFS) technique¹⁻⁵ for determining the atomic structure and location of adsorbed species by studying bromine adsorbed on graphite.⁶ The capabilities of EXAFS that make it particularly suited for adsorption studies are the following: (a) measurement of the short-range order around a particular species of atom in both periodic and nonperiodic arrangements; (b) discrimination between types of surrounding atoms; (c) distinction between distances parallel and perpendicular to the surface by use of polarized x rays; (d) measurement of the variation about its average value of a given bond distance through a Debye-Waller effect; (e) determination of the chemical state of

the atom by edge shift and near-edge structure; and (f) relatively simple interpretation of EXAFS, since multiple scattering effects are precisely zero in a Fourier analysis of the first-neighbor shell and usually negligible for the next few shells.^{5,7}

We find EXAFS to be an extremely useful complement to low-energy electron diffraction (LEED) and other scattering techniques that probe long- and intermediate-range positional order of monolayer films. On suitable systems, EXAFS can determine positions and orientations of adsorbate molecules with respect to neighboring molecules of both the adsorbate and surface atoms. As initial test system we chose Br_2 on graphite, which was studied by Lander and Morrison using LEED.⁸