Yoked Superfluorescence

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(Received 25 May 1995)

A laser pulse two-photon resonant with the $6S_{1/2}$-$6D_{3/2}$ 442 nm transition in Cs vapor generates delayed superfluorescence (SF) on the upper $6D_{3/2}$-$6P_{1/2}$ 876 nm and lower $6P_{1/2}$-$6S_{1/2}$ 894 nm transitions. Yoked SF is observed in the forward (along the laser) direction as evidenced by the simultaneous appearance of SF on both transitions. Cascade SF is observed in the backward direction, where SF on the lower transition only evolves after it has terminated on the upper. The four SF signals exhibit quantum beats associated with the $6P_{1/2}$ hyperfine splitting.

PACS numbers: 42.50.Fx, 42.50.Md

An ensemble of two-level atoms, prepared with an empty ground state, will, if the optical gain is sufficiently high, develop a macroscopic transition moment and launch a superfluorescence (SF) burst which effectively depletes the upper state. This is a manifestation of what Dicke called superradiance, i.e., cooperative spontaneous emission [1]. For a three-level system the SF burst transfers the upper state population to the intermediate state, which then becomes the source of an SF burst to the ground state. This has been called cascade SF [2]. If, however, this system is prepared in a coherent linear superposition of the upper and ground states then the transition moment which develops on the upper optical transition must be accompanied by a transition moment on the lower optical transition [3]. This coupling gives rise to what we call yoked superfluorescence (YSF). It results in the simultaneous emission of SF bursts on the upper and lower transitions and differs from cascade SF where the SF bursts are temporally separated.

Coherent optical excitation of an *S-D* transition in an atomic vapor generally leads to a four-wave mixing (FWM) process with the result that coherent radiation is generated on the *D-P* and *P-S* legs. Mixed in with these induced radiation processes is the spontaneous process associated with ordinary fluorescence. Destructive interference in the optical pumping of the *P* level produced by the combined effect of the pump and amplified spontaneous emission (ASE) fields has been credited with suppressing gain for forward emission in addition to shifting line profiles for backward emission [4]. In a parallel manner the fields generated by the FWM process have been credited with the establishment of alternate pathways between the *S* and *D* levels and the consequent suppression of ASE [5]. These experiments were performed in Na vapor at high number density where (1) wave-vector matching could be achieved only with conical emission and (2) the temporal evolution of the induced radiation fields was effectively instantaneous.

We work in Cs vapor and investigate a related arrangement of reduced complexity, in which (for the first time) the excitation pulse is truly short and the number density, while still large, is lowered to the point where phase matching is collinear. For such a system the excitation pulse serves solely to set up an initial condition in which the *S* and *D* states are in a coherent superposition (two-photon coherence). Coherent radiation subsequently develops separately on each of the upper (U) *D-P* and lower (L) *P-S* legs in the forward (F) direction along the pump and in the backward (B) direction against. We find that these emissions are all delayed with respect to the initial excitation pulse by an amount which greatly exceeds the excitation pulse width. The first pulse (emission) to appear is the BU (along the backward direction and on the U transition) pulse followed by overlapping FU and FL pulses and followed again by the BL pulse. The delay of the FU pulse with respect to the BU pulse displays inhibition. The overlapping FU and FL pulses display YSF; the sequence of temporally separated BU and BL pulses displays cascade superfluorescence.

The superfluorescence problem in a two-photon pumped three-level system, where the top and bottom states are dipole coupled only to the middle state, was first analyzed by Ikeda, Okada, and Matsuoka [3] who predicted that the growth of superfluorescence in the forward direction should be inhibited by the two-photon coherence. Their analysis assumed immediate, uniform two-photon excitation and the absence of any excitation field during the plane-wave superfluorescence development. It was also assumed that superfluorescence develops independently in the forward and backward directions. Under these conditions they derived, from the coupled Maxwell-Schrödinger equations in the linear regime, the second order differential equations governing the Rabi frequency amplitudes Ω_{DP} and Ω_{PS} associated with the upper and lower transitions, respectively. Neglecting relaxation, we use their result to write

$$\frac{\partial^2}{\partial(z/L)\partial\tau}\begin{pmatrix}\Omega_{DP}\\ \Omega_{PS}\end{pmatrix}=\begin{bmatrix}1/T_{DP}^{r} & [\rho_{DS}(z)/\rho_{DD}]/T_{DP}^{r}\\ -[\rho_{SD}(z)/\rho_{SS}]/T_{PS}^{r} & -1/T_{PS}^{r}\end{bmatrix}\begin{pmatrix}\Omega_{DP}\\ \Omega_{PS}\end{pmatrix}, \quad (1)$$

0031-9007/95/75(18)/3265(4)$06.00

where z is the position, τ is the retarded time, S, P, and D are the ground, intermediate, and second excited state labels, respectively, L is the sample length, ρ_{jj} is the initial jth state population (we assume $\rho_{PP} = 0$), $\rho_{DS}(z)$ is the coherence induced by the two-photon pump, and $T_{ij}^r \equiv 8\pi(T_1)_{ij}/3\rho_{ii}n\lambda_{ij}^2 L = 2(T_2^*)_{ij}/\alpha_{ij}L$, $(T_2^*)_{ij}$, $(T_1)_{ij}$, n, λ_{ij}, and α_{ij}^{-1} are, respectively, the superradiant, inhomogeneous and spontaneous lifetimes, number density, wavelength, and Beer's length at line center of the ij transition. We have left out the noise source term which gets the superfluorescence started, and employ instead a constant coherent field $\Omega_{DP}^0 \equiv \Omega_{DP}(\tau, z = 0) = \theta_0/T_{DP}^r$ propagating in the direction of increasing z. The initial tipping angle is $\theta_0 = 2/\sqrt{N}$, assuming a Fresnel number $N_F \equiv 2A/\lambda_{DP}L$ of unity, where $N = \rho_{DD}nV$ is the initial D state population and $V = AL$ is the sample volume [6].

In the forward direction we express the effect of the resonant two-photon pump by an initial condition describing a spatially uniform coherence $\rho_{DS}(z) = \rho_{DS}(0)$. In the backward direction $\rho_{DS}(z) = \rho_{DS}(0)e^{4\pi iz/\lambda_{DS}}$, which spatially averages to zero. In either case, the eigenfunctions of Eq. (1) separately satisfy Bessel's equation, and it follows that in the backward direction $\Omega_{DP}^{\text{bkwd}}(z, \tau) = \Omega_{DP}^0 I_0(2\sqrt{z\tau/LT_{DP}^r})$ and $\Omega_{PS}^{\text{bkwd}} = 0$, while in the forward direction $\Omega_{DP}^{\text{fwd}}(z, \tau) = \Omega_{DP}^0[I_0(2\sqrt{(1-s)z\tau/LT_{DP}^r}) - s]/(1-s)$ and $\Omega_{PS}^{\text{fwd}} = (-s\rho_{DS}/\rho_{SS})(\Omega_{DP}^{\text{fwd}} - \Omega_{DP}^0)$. Here $s \equiv T_{DP}^r/T_{PS}^r = (\rho_{SS}/\rho_{DD})(\lambda_{PS}/\lambda_{DP})^2(T_1)_{DP}/(T_1)_{PS}$, $|\rho_{DS}|^2 = \rho_{DD}\rho_{SS}$, and I_0 is the zeroth order modified Bessel function.

These results show that in the backward direction SF should occur on the upper transition but not on the lower. In the forward direction SF on the upper transition induces simultaneous SF on the lower transition. At the same time the lower SF holds back the upper. We give the name *yoked* SF to this interplay of two transitions. In the backward direction, SF can occur spontaneously on the lower transition if the population of the intermediate state is sufficiently increased (by SF on the upper transition) relative to the ground state population. This latter development would be considerably delayed, however, from the SF that produces it. When present, we recognize it as cascade superfluorescence.

The above results also show that, for $s < 1$, the initial coherence developed by the pump reduces (via the parameter s) the argument of I_0 in the expression for $\Omega_{DP}^{\text{fwd}}(z, \tau)$. Thus SF should evolve more slowly in the forward direction than in the backward. For weaker excitation, $s > 1$, forward directed SF should not develop at all. If relaxation is admitted then the coherence ρ_{DP} will degrade and SF will appear.

The linear theory results provide the essential features of SF development in a three-level system which has been initially excited coherently. For relevant analyses within the nonlinear regime, see [3,7,8]. Some experimental results have been reported in [9,10].

In our experiment, a short, circularly polarized "pump" pulse at $\lambda = 885$ nm generates a coherent superposition between the $6S_{j=1/2,m_j=-1/2}$ and $6D_{j=3/2,m_j=3/2}$ states via resonant two-photon absorption. The intermediate $6P$ states are nonresonant. The pump pulse was obtained by frequency selecting and amplifying the output of a synchronously pumped dye laser. It was 10 ps long and close to transform limited as determined by autocorrelation and frequency spectrum measurements. Fluctuations in the amplifier output provided random samples of pump pulse energies. These pulses were focused to a diameter of 80 μm in a 1 cm long Cs vapor cell yielding N_F (at the D-P transition) close to unity. Cell temperatures ranged from 100 to 170 °C, corresponding to atomic number densities from 1.7×10^{13} to 5.4×10^{14} cm^{-3}. At these densities, the sample is optically thin at the pump wavelength, the SF transitions are Doppler broadened, and the D state population decays primarily to the $6P_{1/2}$ state via SF. The spontaneous $(T_1)_{DP}$ and inhomogeneous $(T_2^*)_{DP}$ lifetimes of the D-P transition are 79 and 1 ns, respectively. For our experiment only half the Cs atoms interact with the pump pulse, and so n only represents half the atomic density.

Both the total emitted energy and the temporal profile of the SF emissions and the pump pulse were separately measured. Photomultipliers (PMT), covered with interference filters, simultaneously fed gated integrators to record each BU, FU, BL, and FL SF emission. These records were stored in a computer and accumulated for later analysis. The temporal intensity profiles were obtained by 1 GHz avalanche photodiodes (APD). Each APD would measure in separate laser shots, first the SF and then the pump pulse, by inserting and then removing the interference filter which covered the detector. A separate APD was dedicated to monitoring the pump response alone in order to provide a reference for eliminating jitter. This reference was always combined with one of the other APD outputs and fed into a Tek7A29 1 GHz amplifier mounted in a Tek7104 oscilloscope mainframe. A TekDCS01 digitizing camera captured the oscilloscope trace for later analysis. SF delay measurements were made by referring the SF response peak to the peak of the pump pulse response from the same diode but recorded in a different trace. The system response was an order of magnitude slower than the 10 ps pump pulse duration but fast and stable enough to resolve a 40 ps relative delay of two short input pulses.

To establish the operating conditions we did the following: (1) We measured the fluorescence on the upper transition in the backward direction at 35 °C and found that it agreed with what we would expect from a D state population excited by a 10 ps pulse of measured energy. As expected a pump pulse energy of 0.01 μJ (6×10^{10} photons) produced maximal (94%) excitation of the D state at cell center. (2) We confirmed that the pump pulse produced a coherent superposition of the D and S states by observing second harmonic generation when a transverse dc magnetic field [11] was

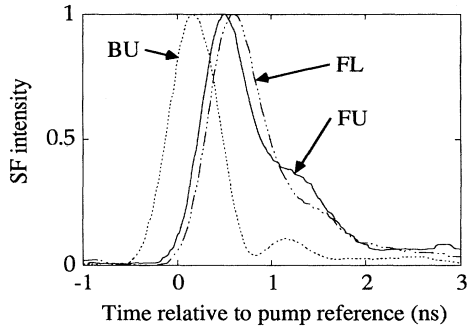

FIG. 1. Normalized SF responses for BU, FU, and FL transitions in Cs at 150 °C pumped with 5×10^{10} photons. The BL signal was too small to detect. With interference filters removed, the pump response peaks appear at 0 ns.

applied to the sample. (3) We confirmed that the second strongest channel of SF emission, through the $7P_{1/2}$ state, was not active (due to diffraction losses). (4) We confirmed that, for pump pulse energies less than 0.02 μJ (1×10^{10} photons), SF was confined to a single spatial mode. This was done by observing the response correlation of two APD detectors placed at different transverse positions in the SF beam.

Normalized temporal traces of SF signals are displayed in Figs. 1 and 2. The former was obtained at $T = 150$ °C pumping with 5×10^{10} photons and the latter at $T = 110$ °C with 1×10^{11} photons. Both figures show that SF is yoked in the forward direction; the FL signal at 894 nm always accompanies the FU signal at 876 nm. This cooperation inhibits SF in the forward direction, as the act of emission at 894 nm reduces the intermediate state population and the associated ρ_{DP} coherence. It is counterintuitive that SF on the lower transition, while helping to maintain the population inversion on the FU transition, nevertheless inhibits the development of SF. This is the essential difference between ordinary lasing and superfluorescence [1]. The former requires only gain, i.e., population inversion; the latter, a growing transition polarization, i.e., a temporally increasing lower-level state amplitude.

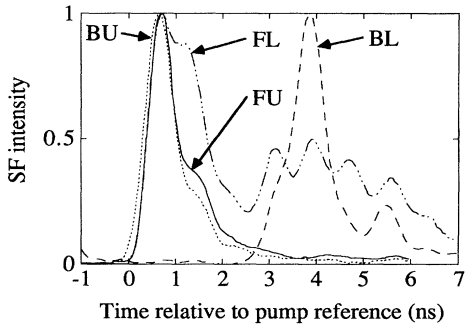

FIG. 2. Normalized SF responses for BU, BL, FU, and FL transitions in Cs at 110 °C pumped with 1×10^{11} photons. With interference filters removed, the pump response peaks appear at 0 ns.

Figure 2 was obtained at lower absolute but greater fractional population inversion density. As a consequence, the upper SF peaked after $(T_2^*)_{DS} = 0.5$ ns, and dephasing blurred the distinction between coherent and incoherent preparation. Thus inhibition was negligible. On the other hand, increased fractional population inversion resulted in increased gain on the BL transition after completion of BU SF with the result that cascade SF appeared on the BL transition.

The after-pulse in BU in Fig. 1 is due to ringing in the detector circuit. The slow modulation in Fig. 2 has to do with the interaction between the SF emissions and the induced macroscopic polarizations. We identify the fast component with the 1.17 GHz splitting of the intermediate $6P_{1/2}$ state [12]. In Fig. 3 we plot the position in time (referred to the pump pulse) of the fast modulation peaks of the BU and BL signals as a function of the pump energy for experiments carried out at 110 °C. Similar behavior was obtained for the FU and FL signals.

Although beating associated with a hyperfine splitting has already been observed [13], the question has been raised as to whether these beats were due to the interference of two coherent fields radiated from noninteracting subsets of the sample or to the interference of the coherent superposition of the two P hyperfine levels within a single atom [14]. If the former obtains, we would expect the beat phase to be random from one laser shot to the next, because the SF fields would develop independently. Our observation of phase stable beats from shot to shot argues for the quantum beat interpretation. The BL signal was sometimes delayed by up to 5 ns (four beat periods) relative to the BU signal, yet the beats of the two signals were locked in phase.

The delay of the SF emissions varied with experimental conditions as is evident from Figs. 1–3. This dependence is more readily analyzed by restricting attention to the FU and BU emissions which are presented as single pulses [15] carrying quantum beats. We therefore inferred the position of the SF peak intensity as the average of the peak (quantum beat) positions, weighted by their heights.

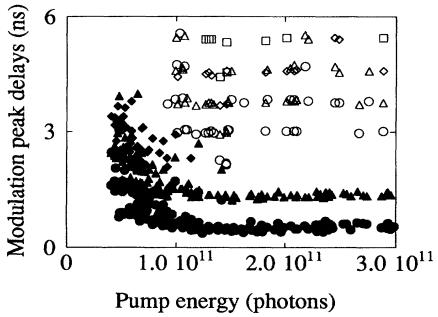

FIG. 3. "Fast" modulation peak delays (referred to pump) versus pump energy in Cs at 110 °C for the BU (solid shape) and BL (open shape) transitions. The sequence of shapes (circle, triangle, diamond, square) corresponds to the sequence of modulation peaks identified in a single response.

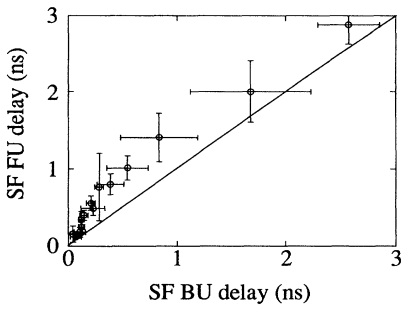

FIG. 4. Averaged SF delay on FU transition versus averaged SF delay on BU transition in Cs at 150 °C.

In Fig. 4 we compare the averaged FU and BU delays at 150 °C. In all cases, the FU delay exceeded the BU delay. For either short or long delays, the ratio of the FU, BU delays approached unity, indicating a reduction in the FU SF inhibition. The long delay limit obtains at very low pump energy inputs where the SF evolution develops on a time scale of $(T_2^*)_{DS}$ so that Doppler dephasing has time to degrade the two-photon coherence. The short delay limit is more complicated because it corresponds to very hard pumping. In this regime, excitation is a convoluted function of atomic position, and the influence of the two-photon coherence is reduced by interference between different domains.

Our discussion of the SF delay has been in terms of the BU transition which is supposed to follow $T_{DP}^{\text{delay}} \cong \frac{1}{4} T_{DP}^r \ln^2(\pi\sqrt{N})$ as long as inhomogeneous relaxation is negligible [16]. From separate measurements of fluorescence and excitation pulse energy we deduce T_{DP}^r so that we can calculate the BU delay directly. We find that for delays less than 0.8 ns [note that $(T_2^*)_{DP} \cong 1$ ns], the ratio of measured to calculated delay equals 2; above 0.8 ns, this ratio increases to a maximum of 3 when the BU delay is 2.5 ns. This increase may be attributed, in part, to the effect of inhomogeneous broadening [17]. We thus obtain the expected functional behavior for the SF delay and, in addition, a measure of its magnitude. The factor of 2 difference might be due to uncertainties in our determination of T_{DP}^r.

Other measurements over the range of 110 °C to 150 °C show that for a fixed value of BU delay, the FU delay increases and the ratio of the energy contained in the FU to that contained in the BU SF decreases with increasing temperature. Keeping the BU delay fixed while increasing the temperature (i.e., increasing number density) requires decreasing the pump energy. Reduced excitation increases the ratio $s = T_{DP}^r/T_{PS}^r \approx \rho_{SS}/\rho_{DD}$, thereby strengthening the coupling between FU and FL fields and increasing the SF inhibition. This explains the behavior of these measurements. Of course, the more the SF is inhibited the greater the disparity in the energies radiated in FU and BU.

The absence of SF in the forward direction as reported by Garrett [4] is certainly related to the inhibition we observe. For $s > 1$ there should be no SF growth until relaxation degrades the two-photon coherence. Perhaps the effect of his long pulse application is to continually reinitialize the *S-D* coherence. Speculation on the connection between our experiments is difficult because of their very different conditions (Garrett operated with weak excitation, $N_F \cong 60$, $L = 10$ cm, and $n > 10^{16}$ cm^{-3}). When we increased N_F to 7, we found, contrary to our expectations, that the inhibition effect increased. We have yet to understand the significance of this result.

In summary, we have observed YSF in the direction of a coherent two-photon pump and cascade SF in the opposite direction. All these emissions display quantum beats associated with the splitting of the intermediate state. The backward upper transition SF delay and energy agree very well with theory for a sample whose N_F is one.

We acknowledge stimulating discussions with professor R. Friedberg, Professor J. T. Manassah, and Professor F. Moshary. This work was supported by the Army Research Office and the National Science Foundation, Grant No. PHY-91-22388.

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