Omnidirectional Superfluorescence

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A dense sample of atomic Rb vapor excited by a 4 ps laser pulse two-photon resonant with the $5S$-$5D$ transition in a large Fresnel number geometry emits a delayed coherent omnidirectional ir pulse on the $5D$-$6P$ transition. This superfluorescence emission involves a coherent population transfer to the $6P$ level and results in simultaneous directional uv emission and coherent population transfer back to the ground $5S$ state. This uv emission is a manifestation of parametric time-delayed four-wave mixing. For a composite noncollinear, $|\vec{k}_1| = |\vec{k}_2|$, excitation, the uv radiation pattern is conical and the cone angle depends on the angle between the excitation pulses. [S0031-9007(99)09254-6]

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Amplified spontaneous emission (ASE) occurs whenever a system is pumped so that there is gain $\alpha L > 1$ along the sample length L [1]. For sufficiently high gain, $\alpha L \gg 1$, the fields developed become so strong that they modify the atomic state populations; the intensity of the emission becomes proportional to the square of the number of cooperating radiators and superfluorescence obtains. Classical superfluorescence experiments were performed in elongated samples which allowed development of two minimally competing superfluorescent modes, forward and backward [2–4]. In the present work, the sample is quasispherical and the gain is large in all directions. Now mode competition is significant and might be expected to either preclude the development of superfluorescence altogether or to at least restrict it to a few well isolated modes which would randomly fire from one shot to the next. Nevertheless, we find that superfluorescence develops and it is omnidirectional. This cannot arise from a spherical mode as the sample is large compared to the radiation wavelength. It must be that, in our sample, spontaneously emitted randomly directed photons are amplified and, in the process, generate separate antennas. These are macroscopic antennas (each consisting of a phased array of many oscillating atomic dipoles) which, if sufficiently excited, radiate a coherent burst of radiation in a time much shorter than the fluorescence lifetime T_1. It is the classic case of radiation damping [5]. Thus, a three-dimensional distribution of randomly phased (with respect to each other) antennas (a kind of pin cushion array), individually directed throughout 4π, is produced and each antenna separately and simultaneously produces a short (compared to T_1) coherent burst.

We work in Rb vapor and generate a high gain inversion on the $5D$-$6P$ transition by applying a short laser pulse two-photon resonant with the $5S$-$5D$ transition. This pulse is sufficiently short that (i) no appreciable superfluorescence takes place during its application and (ii) the atoms are left in a coherent superposition of states $|S\rangle$ and $|D\rangle$. Since the $5P$ state is not populated by the excitation pulse, both the $5D$-$6P$ and the $5D$-$5P$ transitions are inverted with the gain on the former dominating completely as it lies in the far infrared while the latter lies in the visible [5]. For our quasispherical sample, the gain in all directions is sufficiently high for superfluorescence to proceed everywhere; however, the slow response of our ir detector does not allow us to distinguish superfluorescence from ordinary fluorescence on the $5D$-$6P$ transition, and therefore the omnidirectional character of the superfluorescence must be established indirectly. This we do by making the two-photon excitation a composite of two angled but otherwise identical pulses and then exploiting the S-D coherent superposition they produce and the large disparity which exists between the wavelengths of the infrared and two-photon pumped transitions.

The coherent superposition initially established dephases in a time inverse to the Doppler width which in our experiment is long compared to the tens of picoseconds during which superfluorescence evolves. It follows that the coherent population transfer on the superfluorescence ir transition develops a coherent superposition of the $|S\rangle$ and $|P\rangle$ states which results in immediate uv emission along phase-matched directions. This directional uv emission has three components; one each along the separate two-photon resonant excitation directions and a third spread out on a cone whose apex angle depends only (since we worked at low number density) on the angular separation of the excitation components. As this angular separation is varied, phase matching picks out a uniquely determined component of the (4π) ir superfluorescence emission with which to combine.

The large wavelength disparity allows full (4π) coverage of the ir superfluorescence radiation pattern to be established while at the same time allowing the angular separation of the two-photon excitation pulses to be within the relatively narrow optical aperture of the apparatus. We

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find that, for the range of excitation pulse angles, the rim of each and every emitted cone is always filled. This observation, together with the knowledge that the ir superfluorescence is not enhanced in the phased-matched direction (as inferred from [6]), shows that the ir superfluorescence is taking place simultaneously in all directions.

The process leading to cone formation is one of time-delayed four-wave mixing where the four fields are not present simultaneously. In general, four-wave mixing (FWM) is a third order nonlinear (χ_3) phenomenon resulting from interaction of four fields such that $\omega_1 + \omega_2 = \omega_3 + \omega_4$ and $\vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{k}_4$, where ω_n and $\vec{k}_n$ are the frequency and wave vector of the nth wave. In a typical parametric four-wave mixing (PFWM) application, the system is pumped at $\omega_1 = \omega_2$ and emits at ω_3 and ω_4. The responsible interaction need not be resonant and generally all four fields are present simultaneously. Garrett and co-workers [7–9], and others [10,11], have observed and extensively studied PFWM in alkali metal vapors. In these articles, PFWM is resonantly enhanced via the nD-qP-mS channel while the system is two-photon excited in a low Fresnel number geometry near or on the mS-nD transition by a nanosecond laser pulse. These experiments were carried out at high number densities so that the FWM emissions were conical. It was found that when the interaction was resonant the associated ASE on the nD-qP transition was inhibited.

In a related experiment, Brownell *et al.* [6] resonantly excited the $6S$-$6D$ transition in Cs vapor using a short 10 ps laser pulse in a low Fresnel number geometry and observed superfluorescence on the $6D$-$6P$ transition (at ω_3) both in the forward (pump) and backward directions and super-radiant emission associated with the $6S$-$6P$ transition (at ω_4) in the forward direction. Here, also, the emission associated with the upper transition is inhibited, albeit only in the direction of the pump. *The distinguishing feature of this experiment (and ours) is that all emissions develop after the pump pulse has passed.* The label yoked superfluorescence (YSF) was coined as the $6P$-$6S$ super-radiance is due to and occurs simultaneously with forward superfluorescence on the $6D$-$6P$ transition. The connection between YSF and PFWM is self-evident. It was also observed that the phase-matched radiation (in the forward direction) at the $6D$-$6P$ and the $6P$-$6S$ wavelengths was delayed from the non-phase-matched $6D$-$6P$ superfluorescence in the backward direction. The superfluorescence appearing on the $6D$-$6P$ transition in the non-phase-matched direction was in fact more intense than that appearing in the phase-matched direction. It was pointed out that the favoring of the non-phase-matched direction was counterintuitive in that the depletion of the $6P$ level did not maintain favorable circumstances for superfluorescent emission [6]. Superfluorescence requires the presence of a macroscopic transition moment which in turn requires the participating atoms to be in a coherent superposition state. Depletion of the $6P$ level reduces the amplitude of the coherent superposition state.

Our experiment is a natural extension of the above work to the regime where the excitation pulses are noncollinear and the sample shape is not elongated. Here, one photon from each of the separately directed two-photon resonant excitation pulses can contribute to the generation of the $|S\rangle$ and $|D\rangle$ superposition state. We find that YSF is produced over all angular deviations of the pump beams, consistent with phase matching of the YSF transitions. These ~50 ps emissions are generally delayed some 50–300 ps from the 4 ps pump pulses. The inhibition of the upper transition ir superfluorescence emissions at ω_3 yoked to the uv emissions at ω_4 could not be observed as the ir detector rise time is slow compared with the lifetime of that transition. But the central point here is that, in the phase-matched direction, the ir emission is inhibited and not enhanced. The major consequence of this inhibition is that *the emission is conical only on the uv leg and not at all on the ir leg. This observation is novel and runs counter to what one might expect on the basis of the standard PFWM analysis for cw experiments* [12].

By choosing the angle between the directed pump beams, one selects the solid angle which contains that fraction of superfluorescence ir emission at ω_3 yoked to the uv emission at ω_4. When the pump beams are collinear it is the upper level superfluorescence in the forward direction which participates in the four-wave mixing. At the extreme angle of 63° at which the beams are introduced, it is the superfluorescence in the backward direction which participates. Sweeping through intermediate angles, one samples the full 4π in which the ir superfluorescence at ω_3 is contained.

Amplified spontaneous emission drives the YSF emissions we observe. This appears to be at odds with the notion that four-wave mixing inhibits amplified spontaneous emission when, as in our experiment, the pump fields are resonant [8,11]. But our pump pulses are so short that the four-wave mixed fields at ω_3 and ω_4 build up after the pump fields at $\omega_1 = \omega_2$ have passed. As a consequence they do not inhibit the $5D$ level population buildup which is attributed to the reported inhibition of ASE, and in our experiment ASE can develop without restriction. However, as noted earlier, there exists another inhibition process which was observed in previous YSF experiments in Cs vapor, which is the depletion of the $6P$ level due to the uv emission at ω_4 [6]. This reduces the amplitude of the $|P\rangle$-$|D\rangle$ superposition state which produces the ir superfluorescence pulse at ω_3 and thus inhibits it. In Rb vapor the efficiency of the radiation process at ω_3 far exceeds that at ω_4 with the result that emission at ω_4 does not significantly inhibit superfluorescence at ω_3.

Theory.—The intensity of the composite two-photon resonant excitation pulse is given by

$$\vec{S} = (\vec{E}_1 + \vec{E}_2) \times (\vec{H}_1 + \vec{H}_2), \qquad (1)$$

where $\vec{E}_j(\vec{H}_j) = \hat{\varepsilon}_j E_j(\hat{k} \times \hat{\varepsilon}_j H_j) e^{-i(\omega_j t - \vec{k}_j \cdot \vec{r})}$, and $\hat{\varepsilon}_j$ is the polarization vector. The terms in (1) involving products $\vec{E}_j \times \vec{H}_j$ lead to YSF emissions along $\vec{k}_j$ and have been dealt with before [6]. For the purpose of this paper we need only consider the cross terms; their magnitude varies as

$$S_{\text{cross}} \propto E_1 E_2 e^{-i[(\omega_1+\omega_2)t-(\vec{k}_1+\vec{k}_2)\cdot\vec{r}]}. \quad (2)$$

The excitation pulse is short and creates, via a two-photon process, the superposition state $a_D|D\rangle e^{-i[\Omega_{SD}t-(\vec{k}_1+\vec{k}_2)\cdot\vec{r}]} + a_S|S\rangle$; we have used $\omega_1 + \omega_2 = \Omega_{SD}$. The a's are normalization constants which depend on the magnitude and duration of the excitation pulse. Immediately after the excitation pulse has passed only the D and S states are populated, intermediate P states are empty, and there is population inversion of the associated D-P transitions. Spontaneous emission from D to P along $\vec{k}_3 = \omega_3/c = \Omega_{SP}/c$ builds to superfluorescence and the generation of a $b_P|P\rangle e^{-i[\Omega_{SP}t-(\vec{k}_1+\vec{k}_2-\vec{k}_3)\cdot\vec{r}]} + b_S|S\rangle$ superposition; the b's are normalization constants. This leads to YSF at Ω_{SP} if $\vec{k}_3$ (corresponding to one of the pin cushion antennas) is directed so that the S-P superposition is phase matched (see Fig. 1), i.e.,

$$|\vec{k}_4| = 2\pi/\lambda_{SP}, \quad (3)$$

where $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 - \vec{k}_3$. This is obtained when

$$k_m^2 = 4k^2\cos^2\alpha + k_n(k_n - 4k\cos\alpha\cos\alpha_n), \quad (4)$$

where $k = k_1 = k_2$, the indices (m, n) are either (3,4) or (4,3), $\cos\alpha = \frac{\vec{k}_1\cdot(\vec{k}_1+\vec{k}_2)}{k|\vec{k}_1+\vec{k}_2|}$, and $\cos\alpha_j = \frac{\vec{k}_j\cdot(\vec{k}_1+\vec{k}_2)}{k_j|\vec{k}_1+\vec{k}_2|}$. It follows that the YSF emission is confined to a cone whose apex angle is $2\alpha_j$.

In this work, by choosing α (the angle between the pump beams), one effectively selects the solid angle which contains that fraction of (4π) superfluorescence ir emission at ω_3 (on a cone of apex angle $2\alpha_3$) yoked to the uv emission at ω_4. In our experiment $k_4 > 2k_3$ and (4) yields an α for all values of α_3. Thus, by sweeping α, one samples the full 4π solid angle in which the ir superfluorescence at ω_3 is contained. The uv emission at ω_4, at the cone angle given by $2\alpha_4$, is used as a probe to establish the omnidirectional character of the upper transition SF.

The variation of α_3 and α_4 with α is shown in Fig. 2. The angle we experimentally measure is α_4 and it first increases with α and then decreases back to zero. The angle α_4 is zero when $\alpha = 0$ ($\vec{k}_1 = \vec{k}_2$) in which case all YSF emissions are collinear and directed along $\vec{k}_1 = \vec{k}_2$, maximum when $\vec{k}_3 \perp \vec{k}_4$, and zero again when $\alpha = 63°$ (at which $\alpha_3 = \pi$) and $\vec{k}_4$ is pointed in the "forward" direction, i.e., along $\vec{k}_1 + \vec{k}_2$.

Experiment.—A spectra-physics Ti:sapphire mode locked laser/regenerative amplifier system was set to work in the picosecond regime and tuned at 77 nm to be two-photon resonant with the $5S$-$5D$ Rb transition. It generated pulses at a 1 kHz repetition rate. These nearly transform limited pulses were 4 ps long and carried an average energy of about 0.5 mJ. The laser output was split into two 0.5 cm diameter beams of equal intensity and the beams were angled and spatially overlapped in a 1 cm long quartz cell containing saturated vapor of rubidium (Fig. 3a). The cell was situated in an oven and heated to 130–240 °C. The oven was specially designed so that it was capable of transmitting beams of up to 90° angular separation.

The upper $5D$-$6P$ transition was in the far ir (5 μm) and was detected by a LN2 cooled Ge:Au detector. The YSF pulses on the lower $6P$-$5S$ (420 nm) transition generated by the sample were separated from the pump laser via two narrow band interference filters. Behind the cell, two spots, corresponding to the two 420-nm

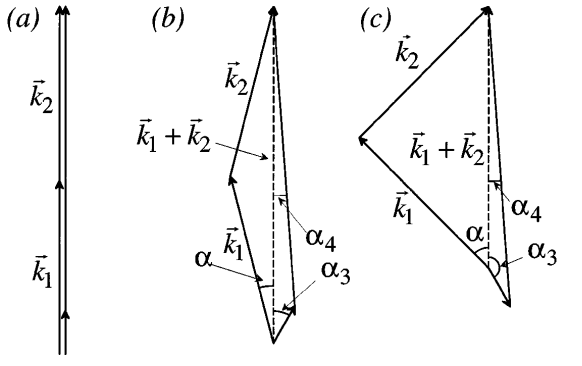

FIG. 1. Phase-matching diagram for two-pulse YSF in rubidium. (a) If there is only one pulse or the two pulses are collinear, both upper and lower transition YSF is also collinear with the laser; (b) if the excitation pulses are angled, both $\vec{k}_3$ and $\vec{k}_4$ have to be angled in order to phase match: YSF is emitted in a cone; (c) at large angles, the upper transition YSF is emitted in the direction practically opposite of the laser excitation: angle α_4 approaches zero again.

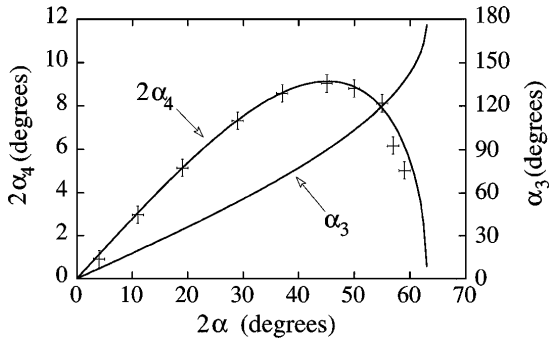

FIG. 2. Apex angle $2\alpha_4$ of the lower ($6P$-$5S$) YSF component and half apex angle α_3 of the corresponding direction of its higher ($5D$-$6P$) component plotted as a function of angular separation 2α between the excitation pulses. Experimental results are also presented here. Note: Whereas the uv radiation was observed only along $2\alpha_4$, the ir component appeared throughout our field of view and was not confined to $2\alpha_3$.

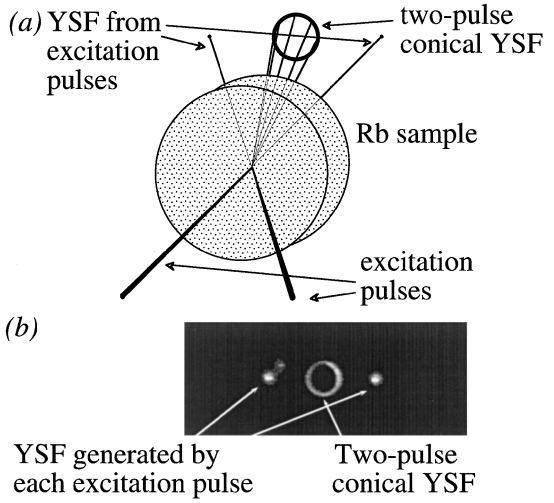

FIG. 3. (a) The optical setup diagram; (b) a photograph of conical YSF obtained with a CCD camera with an objective lens tubed to infinity. Conical YSF shows as a ring; also visible are the spots associated with the two YSF pulses generated by each laser beam alone.

YSF pulses collinear with the two excitation beams, could be observed. When the excitation pulses were applied simultaneously, a ring of conical 420-nm YSF emission also appeared (Fig. 3b). The photograph in Fig. 3b was taken via a Sony XC-77 CCD camera placed behind the sample facing the yoked emissions, with its 25 mm objective lens tuned to infinity.

Conical superfluorescence was observed over a wide range of beam angles, 2α, from 0° to 60°. The dependence (4) of α_4 on α was verified. To measure α, a mirror was placed temporarily on a horizontal rotation stage in front of each excitation beam. The mirror was turned so that the beam was reflected directly backwards and the angle of rotation was recorded. This allowed a measurement of α to within 0.5°. To measure α_4, a 250 μm pinhole was placed on a horizontal translation stage in the focal plane of a 75 mm lens located behind the cell. The diameter of the ring was measured by detecting the throughput of the two opposite sides of the ring through the pinhole. The angle of conical emission was obtained directly from the ring diameter, to within 0.5°. The result of the measurement is presented in Fig. 2, showing excellent agreement with theory.

The fact that uniform conical YSF at α_4 was observed for all α for which α_3 ranges from 0 to π demonstrates that the $5D$-$6P$ superfluorescence takes place simultaneously in all directions. We use the word uniform to mean that the conical emission is virtually uniformly distributed along the rim of the cone.

The delay of the peak intensity of the ring emission was recorded as the function of pump pulse intensities. The intensity waveforms were obtained using a fast photodiode (<300 ps rise time) and Tektronix 7104 oscilloscope. The observed delays were in the range from 50 to 300 ps and monotonically decreased with excitation intensities. Because of the limited detection bandwidth, we could obtain only an upper bound on the superradiance pulse width on the ($6P$-$5S$) transition; it was 50 ps. It can thus be concluded that the PFWM develops in the absence of the pump pulses.

The above observation shows that the macroscopic sample (0.5 cm $D \times 1$ cm L), emits an omnidirectional superflourescence flash, of duration less than 50 ps, that coherently transfers the $5D$ population to the $6P$ level.

In conclusion, we have established the existence of omnidirectional superfluorescence in a rubidium vapor sample of large Fresnel number. For two-pulse noncollinear excitation, this superfluorescence results in noncollinear time-delayed four-wave mixing signals substantially delayed with respect to the pump pulses.

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