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Observations of ultrafast superfluorescent beatings in a cesium atomic vapor excited by femtosecond laser pulses

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ABSTRACT

Picosecond and femtosecond superfluorescent beatings are observed from a dense atomic vapor. Cesium atoms are excited by femtosecond laser pulses via two-photon processes into their coherent superpositions of the ground 6S and excited 8S states. The yoked superfluorescent blue light at lower transitions of 6S – 7P is recorded and studied. Delayed buildup time of this blue light is measured as a function of the input laser beam power using a high-resolution 2 ps streak camera. At low power and density, a beating with a period of approximately 100 picoseconds is observed as we believe that represents the ground state splitting. The autocorrelation measurements of the generated blue light exhibit a beating with a quasi-period of 200 ± 60 fs corresponding to the splitting of the 7P level. Our experimental observations may hold a promising direction for investigation of cooperative radiation in many-body systems including solid-state materials.

1. Introduction

Spontaneous emission from excited atoms is a relatively slow and hardly manipulative process. Spontaneous emission is scaled in nanoseconds or microseconds intrinsically depending on the bandwidth of the excited energy levels of atoms and molecules in gas phase. With a laser excitation, the atoms and molecules are prepared in their collective state and eventually emit synchronously [1–6]. Synchronized spontaneous emission is commonly called superfluorescence (SF) and occurs in a much-reduced timescale compared to that of the unsynchronized spontaneous emission [7,8]. SF is observed in various atomic and molecular vapors [9–14] as well as other exotic materials [15–20]. A two-level model predicts the peak intensity and delay of SF pulses that are proportional to N^2 and N^{-1}, respectively, where N is the number of two-level atoms in the ensemble [7,8]. A three-level model exhibits the so-called cascade SF [21,22]. This process can be understood as a transient four-wave mixing process that involves two input laser pulses and two emitted pulses. If these two emitted pulses in the cascade transition did occur simultaneously then the emission in the lower transition is referred to as a yoked superfluorescence (YSF) [13,23,24].

Common beatings observed in spontaneous emissions involve directly excited energy sublevels only, however, the beatings that are measured in SF emissions can involve the excited energy sublevels both directly and indirectly [25,26] when pumped by the laser light. Another important difference between them is that the SF beating frequency can be shifted depending on many parameters including the input laser power and number density which rather obscures the beating interpretation. For example, SF beating demonstrates quasi-periodic behaviors where both red and blue shifts can be expected [25,27]. In the early works [25,26], cesium (Cs) atomic vapor was excited with nanosecond (ns) pulses and the SF emissions were detected on a nanosecond time scale. Later,

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a picosecond (ps) pulse excitation of the Cs vapor and the detection of the YSF on hundreds of picoseconds were reported [23,24]. However, to our knowledge, the detection of the YSF and its beating properties on a shorter time scale has not been reported yet. This can be achieved, e.g., by pumping the dense Cs atoms with femtosecond (fs) pulses and detecting the generated light signal with an ultrafast streak camera.

In this letter, we study the YSF and its beating behaviors in a dense Cs vapor pumped by 60-fs laser pulses. This research is complementary to our previous work on cooperative emissions from rubidium [5,6,13,28,29] and sodium vapors [14]. Particularly, our focus is to observe the ultrafast beatings. Next section introduces the experimental setup and detection methods. In section 3, the observed results will be discussed. The last section is conclusion.

2. Materials and methods

A Ti:Sapphire amplified laser system (Coherent Inc.) is used to produce a 60-femtosecond (fs) long pulses at 804 nm center wavelength (half width at half maximum – HWHM of 20 nm) at one kilohertz repetition rate. The maximum input power is about 800 mW (i.e., 0.8 mJ/pulse). A beam diameter is approximately half centimeter of the area of 0.2 cm^2. In this case, e.g., 200 mW power is converted to 0.2 mJ energy per pulse, thus, the pulse energy fluence becomes 1 mJ/cm^2. The laser beam enters a 3-inch long cylindrical cell with Cs atoms. The cell is heated up and stabilized (with a variation of less than 2 degrees during experiment) either at 217 C or 242 C. The main reason of this particular temperature selection is that the atomic density of approximately 3×10^{16} cm^{-3} at 217 C is approximately doubled (approximately 6×10^{16} cm^{-3}) at 242 C. As sketched in Fig. S1 in Supplemental Document, the atoms are efficiently excited from the ground state 6S via two-photon absorption through the intermediate level ($6P_{3/2}$ state is not shown here) into 8S state. The laser beam center wavelength line is about 9 nm off from its two-photon resonant transition on 6S – 8S. It is important to note that 6S – 7D transition is even further away (about 19 nm off from 804 nm). Since the HWHM is about 20 nm, therefore, the processes associated with 6S – 7D are substantially diminished. However, another competitive process involving $6S_{1/2}$, $8S_{1/2}$, $6P_{3/2}$, $6P_{1/2}$ may still be present, which was studied and reported in Refs. [23,24]. The emissions (761 nm, 795 nm, 852 nm and 895 nm), in this case, fall within the near infrared region and are filtered out together with the 804 nm input beam. The emissions in transition 7P – 8S are in mid infrared region and are filtered out. In addition, our streak camera and photodiode detector sensors are less or not sensitive in this region. This process is also referred to as a transient four-wave mixing (FWM) as in [23,24]. In our previous work [13], we demonstrated for rubidium vapor that when laser intensity increases (decreases) the coherence between ground and excited states (like in this case 8S and 6S) effectively increases (decreases), thus, the FWM emission increases (decreases). Two photons through intermediate $6P_{3/2}$ state excite atoms. The two-photon excitation eventually triggers simultaneous emissions on both 7P – 8S and 6S – 7P transitions. The emissions in transitions $6S_{1/2}$ – $7P_{3/2}$ at 456 nm and $6S_{1/2}$ – $7P_{1/2}$ at 459 nm are spectrally resolved, see Fig. S1 (C). However, the intensity ratio between the two is significant and 456 nm is mainly contributed. Both emissions represent YSF and have a common ground state [25] therefore, we expect beating in a 456 nm emission (blue light). The splitting of 7P level is 181 cm^{-1}, which corresponds to 184 fs. On the other hand, the splitting of the ground state is about 9 GHz which corresponds to 108 ps beating. Although, the construction of a detailed quantitative model of these laser-field interactions are challenging we believe that these beatings are observed, and our data are listed in the following section.

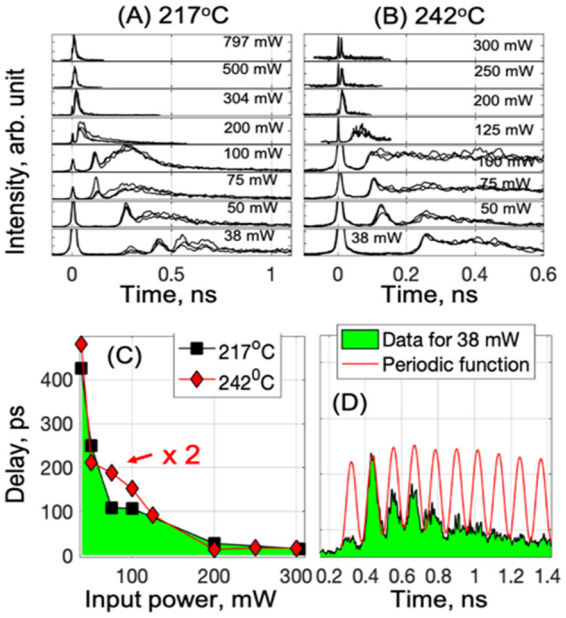

Fig. 1. Pulse temporal characteristics are recorded by the streak camera. (A) Data for 38, 50, 75, 100, 200, 304, 500 and 797 mW at temperature of 217 C. (B) Data for 38, 50, 75, 100, 200, 250 and 300 mW at temperature 242 C. (C) Scaled delay dependence on input power. Red (black) curves for temperature 217 C (242 C). All three data are overlaid for each input power. (D) The selected data (black curve) for 38 mW input power at 217 C compared with a sinusoidal function (dashed curve). (For interpretation of the colors in the figure(s), the reader is referred to the web version of this article.)

The generated blue light beam profiles (approximately 4 cm diameter) are shown for several different input laser powers (from 100 mW to 450 mW with an increment of 50 mW) at the temperature 242 C in Fig. S1 (D). For low power (<250 mW) the blue light divergence half angle is estimated to be less than one degree, however, for high power (>250 mW) the profile diameter is more than doubled due to the ring formation. For higher power, beam profiles have distinct ring and central spot structures. More rigorous theoretical and experimental studies are needed to explore the structured beam profiles and non-monotonic power dependence (see Fig. S1 (E)) as discussed in [30]. In this work, for the sake of transparency, we mainly focus on the data collected at lower power (<250 mW at the temperature 242 C). A detailed setup layout is sketched in Fig. S2 in Supplemental Document. The amplifier output pulses (with optional the Michelson interferometer for double pulses with a variable delay) enter the cell and generate blue light. The generated blue light is spectrally filtered and then detected either by the specially designed photodiode detectors or the 2-ps resolution streak camera. A response time of commercially available photodiode detectors (OPT101, Thorlabs Inc.) is intentionally increased to a half millisecond as to improve the signal to noise ratio by the time-integrated detection scheme. A digital storage oscilloscope (Tektronix) displays and records both input and generated light. A fast streak camera (Hamamatsu, c5680) is used to record temporal characteristics of the input and generated light.

3. Results and discussions

The generated blue light pulse characteristics are shown in Fig. 1. The data at 217 C (see, A) and at 242 C (see, B) and their comparison (see, C) are depicted for different input powers. The sharp peaks are input 804 nm pulses and their rising edge halves are aligned at zero. The following delayed pulses are the characteristics of the blue light pulses. As seen from both figures (A) and (B), buildup delays tend to elongate as the input power decreases. This is the main difference of the cooperative process from

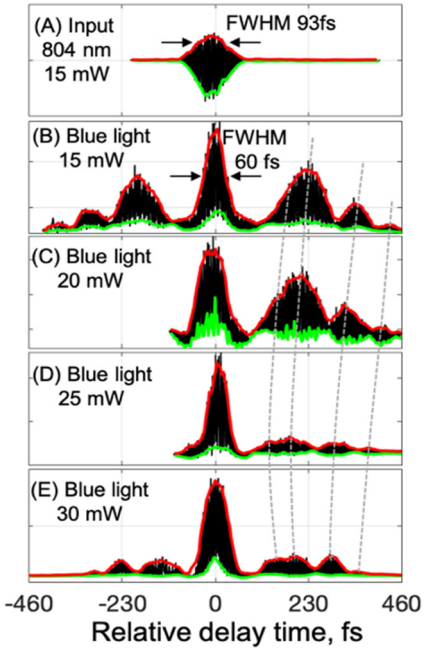

Fig. 2. Interferometric autocorrelation data for input beam (A), the generated blue beam at input power of (B) 15 mW, (C) 20 mW (D) 25 mW (E) 30 mW. Black, red and green curves represent measured signals, upper and lower envelopes of the signals, respectively.

non-cooperative processes. For example, at temperature 242 C, the pulse shapes and delay off-sets become insensitive for relatively high input powers (within a 2 ps resolution), see Fig. S3. As power gradually increases above 300 mW (at 242 C), beam profiles first become non-uniform consisting of the central spot and distinct ring formations, and then for relatively high powers (>450 mW), they become nearly uniform and collimated. Next, as mentioned above, these two temperatures correspond to the density ratio of two. Thus, in Fig. 1 (B) the scaled values of positions of the pulse peak halves at rising edges as functions of variable input power are plotted. In this case, the delay values for 242 C are scaled up by a factor of two. The delay time behaviors of the two quantitatively agree which indicates that they are proportional to N^{-1}. Another intriguing feature of the YSF is the beating that can be printed directly on temporal pulse characteristics [25,26]. The data for 38 mW power at 217 C exhibits this feature as compared to the sinusoidal function with a period of 100 ps, see Fig. 1 (D). The splitting of the ground state $6S_{1/2}$ is 9.19 GHz [25,26] corresponding to 108 ps. It is important to note that the quasi periodicity due to the SF beatings is different from the well-known Burnham-Chiao ringing [31], which explicitly depends on square root of time as was previously observed by the authors [6,13]. In our current experimental condition, this power of 38 mW is the lowest possible input power for the minimum observable signal intensity sufficient enough to be captured with the streak camera. All three data for this power distinctly exhibit the 100-ps beating. However, for a higher input power range, this beating, unfortunately, vanishes.

The streak camera resolution used here is limited, but (interferometric) autocorrelation measurements utilizing a pair pulse with variable time delay can reveal the beating further down to fs time scale [28,29]. All autocorrelation data are taken at temperature 242 C. As seen in Fig. S2, a Michelson interferometer is inserted on the path of input beam. One of the two end mirrors is facilitated to scan controlled by the AC function generator. The scope captures the signals in three channels: applied voltage, autocorrelation traces for the input and generated beams. Fig. 2 displays a set of the autocorrelation data for several different input powers. In Fig. 2 (A), the autocorrelation signal for input beam is measured to be 93 fs. Assuming Gaussian pulse shapes, the pulse temporal width corresponds to 60 fs. This width is confirmed with the measurement by the commercial second order autocorrelator (APE Inc.). We emphasize here that the above interferometric autocorrelation measurement demonstrates a linear interference, which is only sensitive to spectral intensities, but not phases. This one-to-one comparison of the first-order autocorrelation to the second-order autocorrelation concludes that the spectral phase of the input laser pulse is flat. The generated blue light autocorrelations are shown in Figs. 2 (B, C, D and E) for input powers of 15, 20, 25 and 30 mW, respectively. We note that the beating feature observed here not periodic corresponding to a single narrowband beat frequency. However, in this case, an important beating information can be extracted from these data. For example, the measured autocorrelation data exhibit the repeated bumps which have also certain temporal widths. Interestingly, the width of the central bump is measured to be 60 fs, which is shorter than the input beam width of 93 fs. However, the observed off-center bumps are directly related to SF. Unambiguously, maxima of the second bumps are located at a relative delay about 200 fs away from the center (zero delay) position with half width at half maximum of approximately 60 fs. This beating is expected since the splitting of 7P state corresponds to 184 fs. Finally, we note that as input power increases the nature of SF beating is eventually washed out, see the cases (D) and (E) in Fig. 2.

4. Conclusions

Spontaneous emission of excited individual atoms in vapor lasts nanoseconds, if not microseconds and the beatings in it involve directly excited energy sublevels. In contrast, the superfluorescent burst occurs, e.g., on a picosecond timescale. Since the generated superfluorescent light is free from dephasing and/or spontaneous emission decay processes, the beatings in superfluorescent light can involve not only directly laser excited sublevels but also indirectly excited and even ground energy sublevels. In this work, the ultrafast superfluorescent beatings that involve both excited and ground energy sublevels are observed. Cs atoms are excited by 60-femtosecond long, 804 nm laser pulses via two-photon near resonant processes into their coherent superpositions of the ground 6S and excited 8S states. As a part of the transient four wave mixing process, the yoked superfluorescent blue light pulses at lower transitions of 6S – 7P are recorded. Delayed buildup time of this blue light is measured as a function of the input laser beam power by the use of a high-resolution streak camera. Delays are appropriately scaled with the number of atoms as varying the temperature of the vapor. At low power and density, a beating with a period of 100 picoseconds corresponding to the ground state splitting is observed. The linear autocorrelation measurements exhibit a beating with a quasi-period of 200 $\pm$ 60 fs corresponding to the splitting of the 7P level primarily in the lower input laser power range. At increased laser power, the nature of the both observed beating is worn away. Temporal coherent control [32–34] using the ultrashort superfluorescent pulses [29] is still a challenging task. Our experimental observations may hold a promising direction for investigation of cooperative radiation in many-body systems including solid-state materials.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Appendix A. Supplementary material

Supplementary material related to this article can be found online at https://doi.org/10.1016/j.physleta.2022.127945.

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