Suppression of stimulated photon echo during excitation of coherent emission on an adjacent transition

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(Submitted 11 June 1991)
Pis'ma Zh. Eksp. Teor. Fiz. **54**, No. 3, 172–174 (10 August 1991)

A stimulated photon echo has been observed at the same time as the emission of coherent light on an adjacent transition in an LaF_3:Pr^{3+} crystal. Excitation of the emission exactly at the resonance completely suppresses the echo signal. A theory based on the Maxwell–Bloch equations gives a good description of all the basic features of the coherent emission.

The coherent excitation of any atomic system with an inhomogeneous broadening by a train of three ultrashort light pulses which are at resonance with some transition $|1\rangle$-$|3\rangle$ is known to give rise to a stimulated photon echo or "time-delay four-wave mixing."[1] On the other hand, each pump pulse may create a population inversion between the $|3\rangle$ excited state of the system and some lower-lying intermediate state $|2\rangle$, giving rise to coherent emission on this transition. In the present study we have observed the simultaneous occurrence of these two effects, and we have studied how they influence each other.

As the working medium we used an LaF_3:Pr^{3+} crystal 2 mm thick with plane-parallel faces. The concentration of the Pr^{3+} ion was 0.5 at.%. The crystal was cooled to 4.5 K and excited on the ${}^3H_4$–${}^3P_0$ transition of the Pr^{3+} ion by a tunable dye laser pumped by a XeCl* laser. An array of beam splitters and delay lines produced three pump pulses which were nearly square with a length of 17 ± 2 ns at their base. These pulses were separated from each other along the time scale by 45 ± 2 and 165 ± 2 ns. They were of the same intensity. The intensity at the entrance to the crystal could reach $I_L \simeq 100$ MW/cm^2; this figure corresponds to a pulse area $\theta_L \simeq 10\pi$. To satisfy the conditions for spatial synchronization, we directed the second and third pulses in opposite directions. In this case the stimulated-photon-echo signal propagated in the direction opposite the first pulse. The width of the lasing line was $\Delta\omega_L/2\pi c \simeq 0.08$ cm^{-1}, well below the measured inhomogeneous width of the ${}^3H_4$–${}^3P_0$ transition, which at $T = 4.5$ K was $\Delta\omega_H/2\pi c \simeq 0.4$ cm^{-1}.

When the pump frequency ω_L was tuned near the ${}^3H_4$–${}^3P_0$ resonance, we simultaneously detected a stimulated photon echo and coherent emission on the adjacent ${}^3P_0$–${}^3H_6$ transition (Fig. 1). The possible occurrence of this emission was first predicted in Ref. 2; it was observed previously in Ref. 3. The duration of this emission was 2–3 ns, and its excitation threshold was $I_{L1}^{\text{thr}} \simeq 18$ MW/cm^2. At $I_L \gg I_{L1}^{\text{thr}}$, this emission developed on the leading edge of the pump pulse; in this case a second emission pulse was observed on the trailing edge. As I_L was lowered, the pulse of stimulated light moved gradually toward the end of the pump pulse. A theoretical model based on the Maxwell-Bloch equations gives a good description of the observed temporal behavior of the stimulated emission on the ${}^3P_0$–${}^3H_6$ transition.

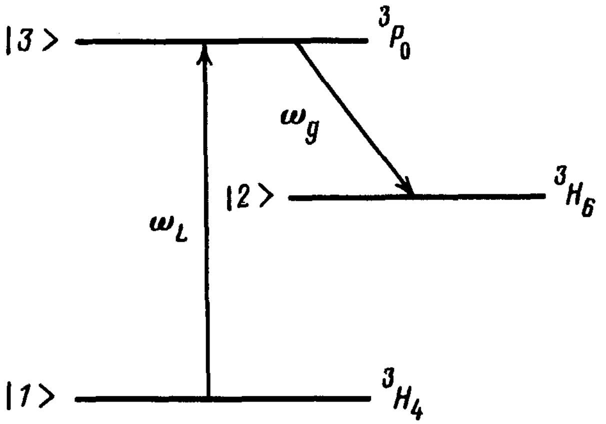

FIG. 1. Energy-level diagram of the Pr^{3+} ion in LaF_3.

The stimulated-photon-echo signal appears at values $I_{L2}^{\text{thr}} \simeq 600$ kW/cm^2 ($\theta_L \simeq 0.7\pi$). As I_L was raised, the intensity of this echo, I_e, also increased. After the appearance of the stimulated emission on the adjacent transition, however, the increase in I_e slowed and then gave way to a decrease. At the same time, the intensity of the stimulated emission, I_g, increased approximately exponentially with increasing I_L. This behavior is illustrated by Fig. 2, which shows the results of the I_e and I_g measurements versus I_L.

We also studied the behavior of I_e and I_g as a function of the detuning (Δ) of the pump frequency from the resonance ($\Delta = \omega_0 - \omega_L/2\pi c$, where ω_0 is the frequency of the $^3H_4$–$^3P_0$ transition). It was found that in the region of detunings Δ in which the

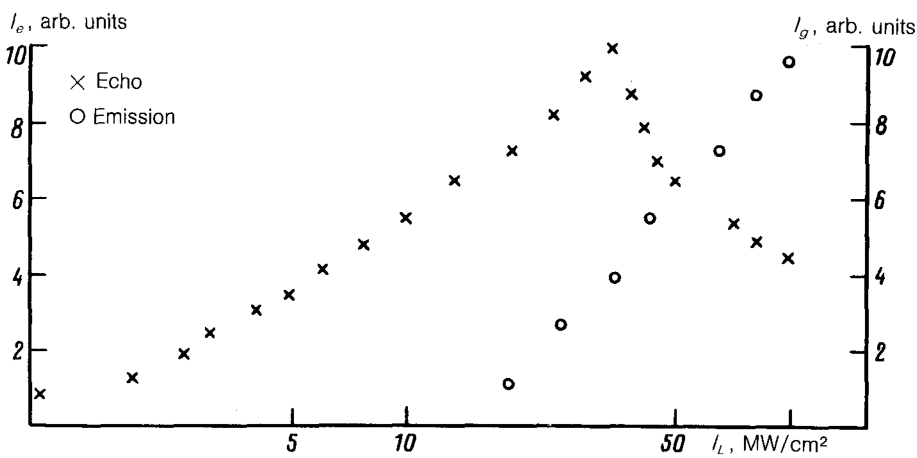

FIG. 2. The echo-signal intensity I_e and the intensity of the coherent emission on the adjacent transition, I_g, versus the intensity of the laser pump, I_L.

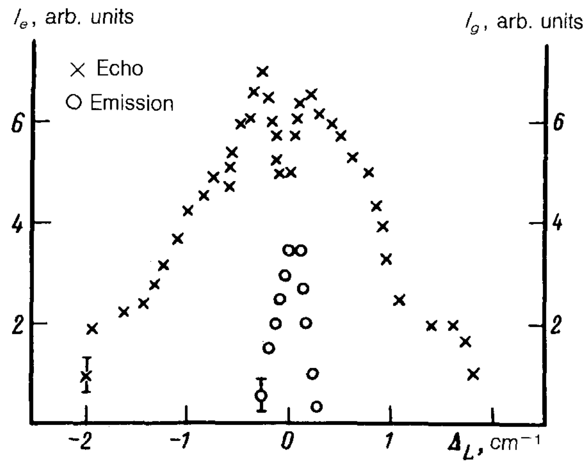

FIG. 3. The echo-signal intensity I_e and the intensity of the coherent emission on the adjacent transition, I_g, versus Δ, the detuning of the pump frequency from resonance with the $^3H_4$–$^3P_0$ transition.

coherent emission is excited the value of I_e decreases (Fig. 3). As expected, I_g reaches a maximum exactly at resonance. At $I_L < I_{L1}^{\rm thr}$, on the other hand, i.e., in a situation in which the emission was not excited, we did not observe a dip on the frequency dependence of the echo intensity.

Finally, we should point out that when we used an LaF_3:Pr^{3+} crystal with a higher concentration of the Pr^{3+} ion (1 at.%), in which case the emission on the adjacent transition was excited at a higher efficiency, we observed a complete suppression of the stimulated photon echo exactly at resonance.

One might suggest the following mechanism for this suppression. We know quite well that the intensity of a stimulated photon echo is determined by the presence of a three-dimensional population grating in the medium, which is formed in both the ground and upper levels by the first two pump pulses. The spreading out of this grating, which is usually linked with relaxation processes, leads to a decrease in the echo intensity.

Under our experimental conditions, the spreading out of the population grating in the $^3P_0$ level apparently resulted from the onset of emission from it to the intermediate $^3H_6$ level. This process goes at an exceedingly high rate, no less than (3–5)$\times 10^8$ s^{-1} under resonance conditions. The spreading out of the grating in the ground state apparently stems from fast relaxation processes from the $^3H_6$ level. Support for this suggestion comes from the emission observed experimentally, which develops at the beginning and end of the pump pulse under the condition $I_L \gg I_{L1}^{\rm thr}$. One can thus assert that the lifetime in the $^3H_6$ level is short (no longer than 15 ns).

The suppression of the stimulated photon echo upon the excitation of coherent emission on an adjacent transition which we have observed here might be turned to advantage to develop methods for rapidly erasing data in optical storage devices.

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Translated by D. Parsons