

PACS numbers: 42.50.Fx; 32.80.Bx; 71.20.Eh
DOI: 10.1070/QE2000v030n07ABEH001779

Optical superradiance in a Pr^{3+} : LaF_3 crystal

V A Zuikov, A A Kalachev, V V Samartsev, A M Shegeda

Abstract. Optical superradiance was experimentally obtained for the first time on the $^3P_0 - {}^3H_4(0)$ transition in a Van Vleck paramagnet – a Pr^{3+} : LaF_3 crystal. Superradiance at a wavelength of 477.7 nm was emitted by a sample 8 mm long, with resonant absorption coefficient $\alpha = 4.7$ cm^{-1}, excited by 10-ns laser pulses at a temperature of 2.2 K.

1. Introduction

Optical superradiance (SR) represents coherent highly directional spontaneous emission of photons by a system of inverted particles (mutually coupled by the radiation field and the noise field) with the intensity proportional to their number squared. This phenomenon has been theoretically predicted by R Dicke in 1954 [1] before the advent of lasers. The idea of the possibility of coherent spontaneous emission has played a considerable role in the development of coherent and quantum optics, and the potentialities of obtaining intense coherent radiation in a single passage through a medium (in the absence of feedback), which are based on this idea, are of great importance for practical applications. The first experiment on SR has been made in HF vapour in 1973 [2]. Then, a series of similar experiments were performed in other gases (see review [3] and monograph [4]).

First solid-state experiments on optical SR have been made in the early 1980s [5, 6] on O_2^- centres in a KCl crystal and on impurity pyrene molecules in a biphenyl crystal [7, 8]. A detailed analysis of these experiments in review [9] revealed characteristic features of SR in solids in comparison with gases. These features are associated with dense packing of impurity centres in a matrix (in contrast to rarefied gases), the crystalline environment, and a complex energy level diagram of the centres.

Because of this, each new solid-state experiment on SR is of considerable interest, especially an experiment made on a medium offering promise as a data carrier for optical processors. Among these media are Van Vleck paramagnets, which represent impurity crystals doped with rare-earth ions, in particular, a Pr^{3+} : LaF_3 crystal.

V A Zuikov, A A Kalachev, V V Samartsev, A M Shegeda E K Zavoiskii Physicotechnical Institute, Kazan Scientific Centre, Russian Academy of Sciences, Sibirskii trakt 10/7, Kazan, 420029 Russia; e-mail: samartsev@dionis.kfti.kcn.ru

Received 1 March 2000
Kvantovaya Elektronika **30** (7) 629 – 631 (2000)
Translated by A N Kirkin; edited by M N Sapozhnikov

In this work we observed for the first time and studied optical SR on the $^3P_0 - {}^3H_4(0)$ transition in a Pr^{3+} : LaF_3 crystal at a wavelength of 477.7 nm at 2.2 K. The theoretical estimates [10] showed the possibility of observing long-lived and trigger SR signals. Taking into account the fact that SR signals are considerably higher in power than long-lived photon echo, we hope to use them in optical memory systems. In our opinion, the experiment presented here may stimulate further studies in this field.

2. Experiment

The schematic of the experimental setup and the energy level diagram of Pr^{3+} : LaF_3 are presented in Figs 1 and 2. A sample (a Pr^{3+} : LaF_3 crystal with a praseodymium atomic concentration of 1%) of size $4 \times 4 \times 8$ mm was placed in a cryostat and cooled down to the liquid helium temperature ($T = 2.2$ K). It was optically excited by single laser pulses at the $^3H_4(0) - {}^3P_0$ transition (477.7 nm) at a pulse repetition rate of 12.5 Hz. Laser radiation was focused on the sample by a lens with focal distance of 30 cm, and the beam diameter in the sample was 0.1 mm. Pump pulses were 10 ns long, and their spectral width was 3 – 4 GHz, which is approximately two orders of magnitude narrower than the inhomogeneous width of the optical transition. The pulse power was changed from 5 to 35 kW. The optic axis of the crystal was oriented at an angle of 5° to the direction of propagation of pump pulses. The resonant-absorption coefficient was $\alpha = 4.7$ cm^{-1}.

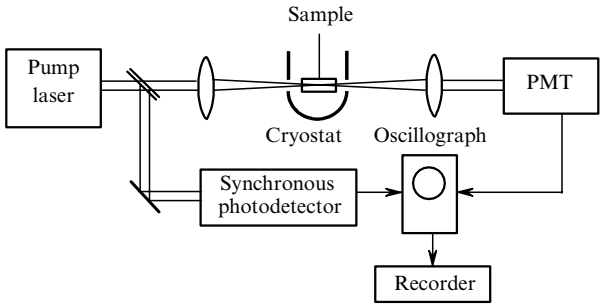

Figure 1. Schematic diagram of the experimental setup.

Emission of the resonance medium was detected by a PMT, and its output signal was observed on an oscillograph, which was triggered by a photodetector pulse.

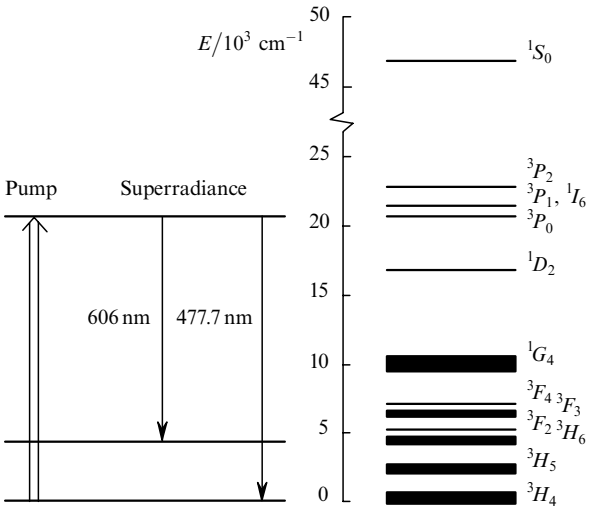

Figure 2. Energy diagram of working levels of the Pr^{3+} ion in a LaF_3 crystal.

3. Basic results

We studied the nature and properties of emission of the Pr^{3+} : LaF_3 crystal by gradually increasing the peak power of a pump pulse. When the laser pump power exceeded a threshold value ($\sim$ 10 kW), the resonance medium emitted a 10 – 12-ns optical coherent pulse, in the direction of a pump pulse (and in the opposite direction), which was delayed by 12 – 15 ns. We interpret it as a pulse of optical SR on the $^3P_0 - {}^3H_4(0)$ transition.

Oscillograms of this pulse (on the right) and of a single pump pulse (on the left) are presented in Fig. 3a. For pump pulse powers below 10 kW, no SR pulses delayed in time were observed. Fig. 3b shows SR pulses excited by a pulse train with a repetition rate of 12.5 Hz. The signals were detected in the direction opposite to the pump direction.

Note specific features of the signal observed in the experiments: (1) a high intensity, which considerably exceeds the incoherent spontaneous background; (2) the coherent emission time (10 ns) is considerably smaller than the lifetime of the $^3P_0$ state [11]; (3) a noticeable delay of the pulse; (4) the presence of statistical properties typical of SR; (5) a sharp directivity of emission, both in the direction of propagation of a pump pulse and in the opposite direction; (6) the disappearance of the signal with increasing temperature of the sample (above 4.2 K), which is typical of SR type cooperative processes and optical transient processes (the amplification of incoherent spontaneous emission in a medium is independent of temperature). These specific features suggest that the additional optical pulse observed after the pump pulse represented a SR signal.

In addition to this SR signal (in the blue region), the crystal emits another SR pulse (in the orange region) on the $^3P_0 - {}^3H_6$ transition. An oscillogram of this pulse is similar to the one presented above, but its delay time with respect to the pump pulse reached 30 ns (and even more), and the threshold pump power required for the excitation of the SR signal was as high as 30 kW. Thus, we observed two-colour SR in the given crystal.

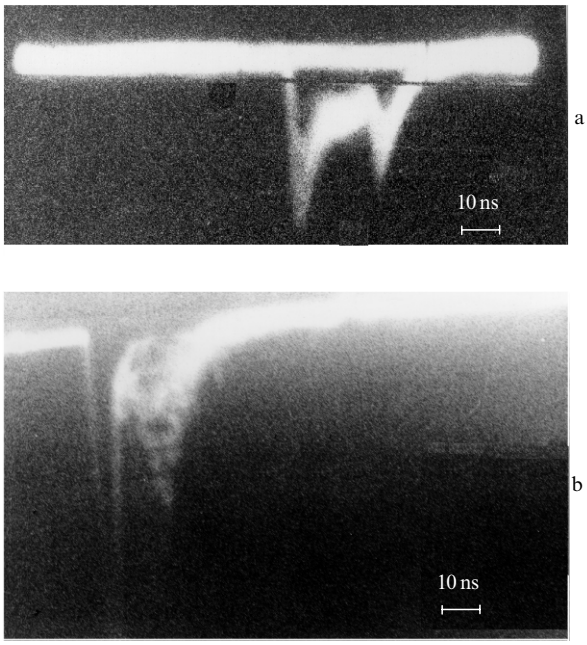

Figure 3. Oscillograms of SR signals (on the right) in an extended Pr^{3+} : LaF_3 sample excited by single pump pulses (a) and by a pulse train with the 12.5-Hz repetition rate (b).

4. Conclusions

The observation of SR in a crystal doped with a non-Kramers rare-earth ion (Pr^{3+}) is of principle importance for its use in optical processors, especially in the trigger excitation regime [12]. Taking into account the fact that a praseodymium impurity ion has long-lived hyperfine sublevels in the $^3H_4(0)$ ground state, it is reasonable to expect the excitation of long-lived trigger SR [13] in this crystal. We are going to realise these SR regimes in our future experiments.

Acknowledgements. This work was supported by the International Science and Technology Centre (Project No. 737), the Universities of Russia: Fundamental Studies Program (project No.990214), the Russian Foundation for Basic Research (project No.99-02-18495a), and the Physics of Quantum and Wave Processes Program, Fundamental Spectroscopy and Laser Physics Division, of the Russian Ministry of Science.

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