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Temporal evolution of a coherent stimulated radiation pulse in the three-level system in a Pr^{3+} : LaF_3 crystal

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***Abstract*. The temporal characteristics of coherent stimulated radiation at the $^3P_0 - {}^3H_6$ transition in the Pr^{3+} ion in a LaF_3 matrix are studied by tuning the pump frequency in the vicinity of the $^3H_4 - {}^3P_0$ transition. It is found that in the case of the exact tuning to the resonance, a laser pulse, consisting of a train of picosecond spikes of total duration about 10 ns, was delayed by 3 – 4 ns with respect to the pump pulse onset. As the pump pulse detuning was increased, the shape of the coherent laser pulse changes and its delay increased up to 10 ns. The experimental results are interpreted theoretically.**

Keywords: *radiation dynamics, coherent stimulated radiation, optical pumping.*

Coherent resonance excitation of a LaF_3 : Pr^{3+} crystal at the $^3H_4 - {}^3P_0$ transition of the Pr^{3+} ion, which was first performed in [1], was studied experimentally in a number of papers [2 – 5]. It was found that coherent pumping of this transition leads to the generation of stimulated radiation at the adjacent $^3P_0 - {}^3H_6$ transition, which has a number of interesting features, which are absent upon incoherent excitation. In particular, it was shown in [2] that when the pump power exceeded significantly the threshold power, the laser pulse consisted of two spikes of duration no more than 1 ns each, which was limited by the time resolution of the detecting system. It was shown in [3] that lasing at the $^3P_0 - {}^3H_6$ transition strongly affects the development of a stimulated photon echo in a Pr^{3+} : LaF_3 crystal. It was also found quite recently that coherent excitation of the $^3H_4 - {}^3P_0$ transition in the Pr^{3+} ion in a LaF_3 matrix gives rise to superradiance at the $^3P_0 - {}^3H_4(0)$ and $^3P_0 - {}^3H_6(1)$ transitions [4, 5], which can be considered as an alternative to lasing at the $^3P_0 - {}^3H_6$ transition.

In this paper, we studied in detail the temporal properties of stimulated radiation at the $^3P_0 - {}^3H_6$ transition produced upon coherent excitation of a Pr^{3+} : LaF_3 crystal at the $^3H_4 - {}^3P_0$ transition. Unlike [2], the study was performed with the picosecond time resolution.

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Excitation was performed by a tunable dye laser pumped by a XeCl excimer laser.

The laser beam was focused with a long-focus lens ($f = 50$ cm) to a 0.5×0.5 mm spot on a 2-mm thick Pr^{3+} : LaF_3 crystal with plane – parallel polished faces (the atomic concentration of Pr^{3+} ions was 1 %). The crystal was studied in a helium optical cryostat providing measurements in the temperature range from 4.2 K to room temperature.

The pump radiation incident on the crystal and transmitted through it, as well as stimulated coherent radiation excited in the crystal were directed through three fibres on the entrance slit of an Agat streak camera. The time resolution of the camera dependent on the sweep rate achieved 2 – 3 ps. The camera was triggered by an optical detector on which 4 % of the dye laser radiation was incident. In this case, the triggering instability was minimal because it was determined by only by the response jitter of the electronic circuit of the camera and allowed the use of maximal sweep rates. The delay of the camera sweep onset with respect to the triggering pulse equal to 80 ns was compensated by transmitting the exciting radiation from the dye laser through a delay line before directing it on the crystal.

The spectral properties of radiation were studied with a DFS-452 grating spectrometer and a Fabry – Perot interferometer with a free spectral range of 0.25 cm^{-1} and an instrumental function width of 0.004 cm^{-1}.

The dye laser frequency ω_L was tuned in experiments within the inhomogeneous linewidth $\Delta\omega_0/(2\pi c)$ of the $^3H_4 - {}^3P_0$ transition [$\omega_L/(2\pi c) =$ 20930.1 cm^{-1}] of the Pr^{3+} ion, which was equal to $\sim$1.8 cm^{-1} at 4.2 K. The laser linewidth $\Delta\omega_L/(2\pi c)$ did not exceed 0.06 cm^{-1}. The laser pulse was bell-shaped, with the width at the base $\tau_L = 15$ ns, which is substantially shorter than the longitudinal (T_1) and transverse (T_2) relaxation times for the $^3H_4 - {}^3P_0$ transition at liquid helium temperature, which are equal to 47 and 2.4 μs, respectively. The pump radiation intensity I_L on the crystal was limited by the radiation resistance of the latter and could achieve $\sim$40 MW cm^{-2}, which corresponds to the pulse area $\Theta_L \approx 1.5\pi$.

Coherent stimulated emission of the crystal was observed at the $^3P_0 - {}^3H_6$ transition frequency $\omega_g/(2\pi c) =$ (16708.6 ± 0.1) cm^{-1} already at the pump intensity $I_L \approx$ 20 MW cm^{-2}. The emission was excited both in the forward and backward directions and propagated along the pump beam, the width of the emission spectrum being $\Delta\omega_g/(2\pi c) \approx 0.04$ cm^{-1}. It was generated upon tuning ω_L

within the inhomogeneous width of the $^3H_4 - {}^3P_0$ transition and was observed up to 25 K.

Figure 1 shows the time sweeps of radiation incident on and transmitted through the crystal, as well as of coherent stimulated radiation. Figure 1a corresponds to the exact tuning of ω_L to the $^3H_4 - {}^3P_0$ transition frequency. In Figs 1b, c, the detuning $\Delta\omega/(2\pi c)$ from the resonance is 0.10 and 0.18 cm^{-1}, respectively. One can clearly see that for $\Delta\omega/(2\pi c) = 0$, the stimulated radiation pulse consists of a train of spikes with the FWHM of 0.7 ns separated by the time interval $\sim$1.5 ns. The delay between the pump pulse and the lasing onset is minimal, being $\sim$4 ns. Under these conditions, the pump radiation transmitted through the crystal also exhibits spikes, which are, however, irregular. Note that the appearance of the spikes is caused by the coherent nature of interaction of laser radiation with matter [6] and it was earlier observed in similar experiments with alkali-metal vapours [7]. As the detuning $\Delta\omega/(2\pi c)$ increased both to the blue and the red, the delay time of the laser pulse with respect to the pump pulse increased up to 10 ns, while the laser pulse duration decreased down to 2 ns, which is clearly seen in Figs 1b, c.

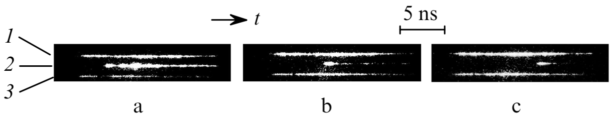

Figure 1. Chronograms of exciting radiation incident (*1*) on the crystal and (*3*) transmitted through it, and also of coherent stimulated radiation (*2*) for $\Delta\omega/(2\pi c) = 0$ (a), 0.10 (b), and 0.18 cm^{-1} (c). The time resolution is no worse than 100 ps.

The results were analysed using the self-consistent system of the Maxwell – Bloch equations describing the behaviour of a three-level Λ-configuration system (Fig. 2) excited by a light field at the frequency ω_L of the $|1\rangle - |3\rangle$; transition. Lasing is developed at the adjacent $|2\rangle - |3\rangle$ transition. It is assumed that $\mu_{12} \ll \mu_{13}$ (μ is the matrix dipole element of the corresponding transition) and $\tau_L \ll T_2 \ll T_1$, i.e., the condition of coherent excitation is fulfilled. It is also assumed that the working-medium thickness l is much smaller than the optical length $c\tau_L$ of the pulse. The latter circumstance allows us to ignore the spatial derivative in the Maxwell wave equation, by replacing it by the characteristic propagation time $T_p = l/c$ of the light wave in the crystal.

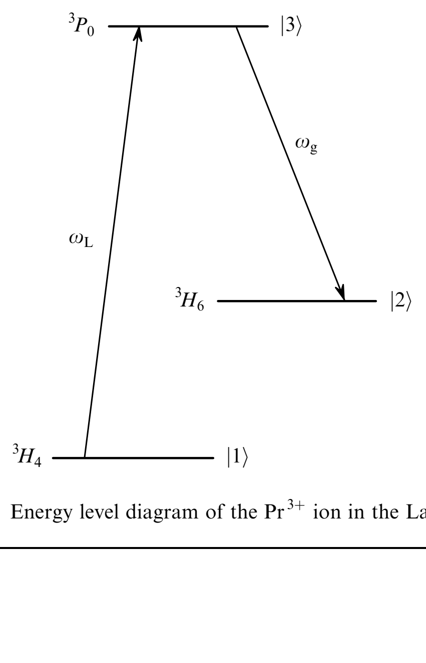

Figure 2. Energy level diagram of the Pr^{3+} ion in the LaF_3 matrix.

The self-consistent system of the Maxwell – Bloch equations was solved by the fourth-order Runge-Kutta method. It was found that, when the conditions

$$\frac{T_2}{T_p} < \alpha \ll 1 \quad \text{and} \quad \omega_R < \frac{\alpha}{T_p} \tag{1}$$

were fulfilled, where $\alpha = 2\pi\omega_{23}N_0\mu_{23}^2 T_2 T_p/\hbar$ and ω_R is the Rabi frequency at the $|1\rangle - |3\rangle$ transition, single-pulse lasing was observed with the characteristic duration $t_g \sim (2\pi\omega_{23}N_0\mu_{23}^2/\hbar)^{-1/2}$. If $\alpha \gg 1$ and $\omega_R > \alpha/T_p$, multi-pulse lasing is observed at the $|2\rangle - |3\rangle$ transition, which continues until a complete depletion of the $|3\rangle$ level at the end of each individual pulse.

In the case of low-intensity pumping, lasing develops at the pump pulse tail. As the pump intensity increases, the stimulated emission pulse divides into a few pulses, which shift to the exciting laser pulse.

Therefore, the model proposed in the paper well describes qualitatively the temporal properties of lasing at the $^3H_6 - {}^3P_0$ transition produced upon coherent resonance excitation of the $^3H_4 - {}^3P_0$ transition.

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