Photon echo in $CaWo_4$:Nd^{3+}

V. V. Samartsev, R. G. Usmanov, and I. Kh. Khad'yev

Kazan' Physico-technical Institute, USSR Academy of Sciences
(Submitted May 23, 1975)
Pis'ma Zh. Eksp. Teor. Fiz. **22**, No. 1, 32–35 (July 5, 1975)

Observation of photon echo in the crystal $CaWO_4$:Nd^{3+} is reported. The conditons and features of the formation of this coherent response are investigated.

PACS numbers: 42.65.D

The photon-echo method[1–3] is highly promising in the investigation of the mechanism of broadening of resonance lines of relaxation times, of dynamic generation of coherent responses, and of their waveforms and spectral composition in crystals. However, all such investigations of the conditions for the realization of this effect in solids were exclusively connected until recently with ruby, in view of the difficulties of obtaining resonance with the corresponding laser radiation. Therefore the observation of this phenomenon in other crystals is quite urgent. In this paper we report observation of photon echo in another crystal, namely $CaWO_4$: Nd^{3+}. We have investigated the conditions and features of the formation of this coherent response at 2.2 °K in the transition $^4I_{9/2}$—$^4F_{3/2}$.

The oscillograms of the experimentally observed signals are shown in Fig. 1. The upper oscillogram records the aggregate of two exciting pulses with the second pulse delayed by a time $\tau = 64$ nsec, and the photon-echo signal (first on the right) at the instant of time 2τ. The lower oscillograms demonstrate the absence of an echo signal in the case when one of the exciting pulses is not applied.

Figure 2 shows a block diagram of the experimental setup. The excitation at the wavelength $\lambda = 8767$ Å was produced with an organic-dye laser (1, 1′ diethylquinotricarocyanin iodide in dimethyl formamide). The pump system consisted of a ruby laser with passive Q switch-

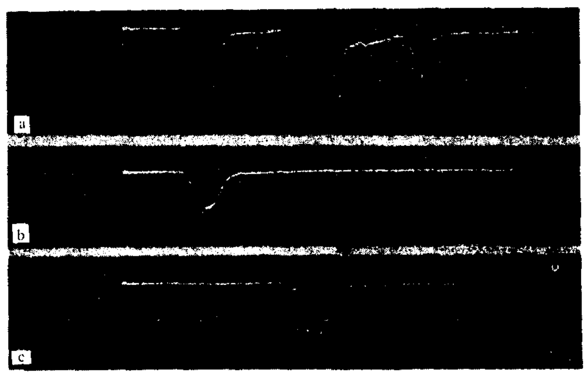

FIG. 1. Oscillograms of observed signals in $CaWO_4$: Nd^{3+}: a) two exciting laser pulses and photon-echo signal (first on the right) in the case $\tau = 64$ nsec; b) action of only the first laser pulse (there is no echo); c) action of only the second pulse (there is no echo).

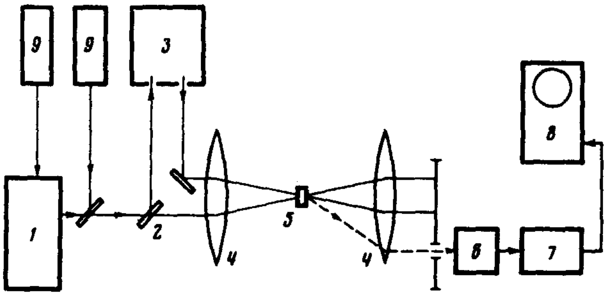

FIG. 2. Block diagram of the experimental setup used to observe photon echo in $CaWO_4 : Nd^{3+}$: 1—organic dye-solution laser; 2—beam-splitting plate, 3—optical delay line (τ = 20—120 nsec); 4—investigated sample; 5—Kerr cell with crossed polarization prisms; 7—high-speed photomultiplier ÉLU-FT; 8—high-speed I2-7 oscilloscope; 9—adjusting LG-75 helium-neon lasers.

ing, which generated at $\lambda_{ruby} = 6943$ Å optical pulses of duration $\Delta t \sim 20$—35 nsec and average radiation power ~40 MW. The time τ between pulses was varied with the aid of a delay line in the range 20—120 nsec. The signals were registered with a high-speed photomultiplier (ÉLU-FT) with a resolution not worse than 2.7 nsec.

The working sample, cut from a $CaWO_4 : Nd^{3+}$ laser rod, was a cube with cross section area $S = 0.25$ cm^2. The concentration of the Nd^{3+} ions was ≈0.5 wt.%. The optical axis of the crystal was in the plane of the sample and was perpendicular to the direction of the action of the exciting pulses. An optical helium cryostat together with the investigated sample was placed between Helmholtz coils, which made it possible to produce a constant horizontal magnetic field ~50—200 G. We were unable to record photon echo in a zero magnetic field.

The arrangement of the terms of the ^4I multiplets and the $^4F_{3/2}$ term of Nd^{3+} ion in the $CaWO_4$ crystal is shown in Fig. 3.[4]

The time T_1 of the longitudinal relaxation on the transition $^4I_{9/2} \to {}^4F_{3/2}$ amounts to $\approx 10^{-4}$ sec. Estimates of the time T_2 of the transverse irreversible relaxation, due to the magnetic and electric dipole-dipole interactions, yield values $\geqslant (5-7)\times 10^{-8}$ sec. The hyperfine and superhyperfine interactions are characterized by somewhat longer relaxation times. The estimates of the relaxation times due to the Orbach process $1/\tau_{orb} \sim \Delta^3 \exp(-\Delta/k_B T)$ and the Raman process $1/\tau_{Ram} \sim \Delta^{-4} T^9$ (where Δ is the splitting between any of the resonant energy sublevels and the nearest nonresonant sublevel, k_B is the Boltzmann constant, and T is the absolute temperature) lead to values not exceeding 10^3 sec^{-1}. An investigation of the luminescence and absorption spectra has made it possible to find the time of the transverse reversible relaxation, namely $T_2^* \sim 10^{-11}$ sec. Thus, only a fraction of the "spin packets" of the inhomogeneously-broadened line took part in the formation of the echo.

It follows from the foregoing that the conditions required for observation of photon echo were satisfied in our experiment.

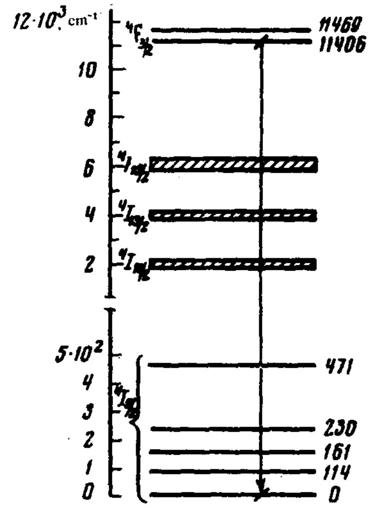

FIG. 3. Energy level scheme of the Nd^{3+} ion in $CaWO_4$.

It was established experimentally that the wave vector of the photon echo k_e statisfies the following spatial-synchronism condition: $\mathbf{k}_e = 2\mathbf{k}_2 - \mathbf{k}_1$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the exciting pulses. In addition, a correlation existed between the shape of the echo and the shape of the exciting pulses, this correlation being connected with the features of the formation of the photon echo in the case when $\Delta t \gg T_2^*$.[5,6] We note that theory[5,6] also attributes the smallness of the echo intensity (in comparison with the intensity of the pulses) to the presence, in the case of $\Delta t > T_2^*$, of the factor $3T_2^*/4\Delta t)^2$. Analysis has shown that when the polarization vectors of both linearly-polarized pulses are parallel to each other, the echo signal also has a linear polarization that coincides with the polarization of the pulses.

The laser emission linewidth was ~0.5—0.6 nm. When the excitation wavelength was changed by 0.5—0.8 nm, the photon-echo signal vanished. We call attention also to the fact that the echo was noticeably decreased when the direction of the optical axis of the crystal relative to $\mathbf{k}_1$ was changed by 90°.

The photon echo signal vanishes if the diffraction grating in the dye laser is replaced by a flat mirror (this broadens the emission spectrum to 30 nm). This circumstance is due to the fact that it becomes impossible to obtain $\pi/2$ and π pulses under these conditions.

[1] U. Kh. Kopvillem and V. P. Nagibarov, Fiz. Met. Metallogr. **15**, 313 (1963)]; U. Kh. Kopvillem, V. P. Nagibarov, V. A. Pirozhkov, V. V. Samartsev, and R. G. Usmanov, Fiz. Tverd. Tela **14**, 1794 (1972) [Sov. Phys.-Solid State **14**, 1544 (1973)]; ZhETF Pis. Red. **20**, 139 (1974) [JETP Lett. **20**, 60 (1974)].
[2] I. D. Abella, N. A. Kurnit, and S. R. Hartmann, Phys. Rev. Lett. **13**, 567 (1964); Phys. Rev. **141**, 391 (1966).
[3] S. Chandra, N. Takeuchi, and S. R. Hartmann, Phys. Lett. **41A**, 91 (1972).
[4] Ya. E. Kariss, A. M. Morozov, and P. P. Feofilov, Opt. Spectrosk. **17**, 887 (1964).
[5] A. G. Shagidullin, in: "Svetovoe ekho" (Photon Echo), Izd. KGPI, Kazan', p. 107.
[6] A. I. Alekseev and I. V. Evseev, Zh. Eksp. Teor. Fiz. **68**, 456 (1975) [Sov. Phys.-JETP **41**, No. 2 (1975)].