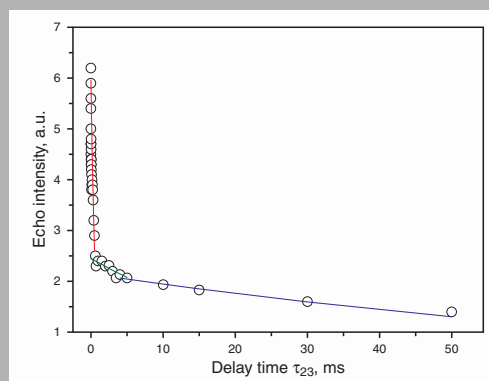


Abstract: Two-pulse, stimulated and accumulated photon echoes are investigated for the first time in a highly doped crystal Tm:YAG containing 10 at.% Tm³⁺. The decay curves of the two-pulse and stimulated photon echoes generated at 793.15 nm on the transition ³H₆(1)–³H₄(1) of the impurity ions are measured in the absence of an applied magnetic field and parameters describing the energy and phase relaxation are determined. The dependence of the intensity of the accumulated photon echo on the number of excitation pulse pairs is investigated.



The decay curve of stimulated photon echo in Tm:YAG (10 at.%)

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Optical echo-spectroscopy of highly doped Tm:YAG

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1. Introduction

Photon echo has been widely used in optical coherent spectroscopy for investigation of various resonant media [1–9], among which rare-earth doped solids are of special interest [10]. In particular, Tm³⁺:Y₃Al₅O₁₂ crystal (Tm:YAG) is currently considered as one of the most promising materials suited for coherent transient-based all-optical signal processing [11–14]. The point is that the wavelength of the optical transition ³H₆(1)–³H₄(1) of Tm ions doped into YAG is equal to 793 nm, where commercial diode lasers as well as solid state (Ti:Sapphire) lasers are available. Besides, a metastable electronic state ³F₄ of Tm, which has a lifetime of 10 ms [15], as well as hyperfine levels of the ground electronic state, which have a lifetime of 30 s under application of a small magnetic field [16], make this crystal suited for various long-time storage applications. Finally, Tm ions ex-

hibit a rather small homogeneous linewidth Γ_h of the optical transition, which leads to a significant memory capacity determined by the ratio Γ_{inh}/Γ_h , where Γ_{inh} is an inhomogeneous linewidth. Generally the homogeneous linewidth is determined from the Mims dephasing time T_M [17] of a two-pulse photon echo. In the absence of an applied magnetic field, $T_M = 75 \mu\text{s}$ [15] or even $116 \mu\text{s}$ [18] depending on Tm-doping level. The latter value corresponds to $\Gamma_h = 1/\pi T_M = 4 \text{ kHz}$ and, consequently, $\Gamma_{inh}/\Gamma_h \sim 10^6$ for a typical value of inhomogeneous linewidth, $\Gamma_{inh} = 17 \text{ GHz}$, in such a crystal.

It should be noted that such a long dephasing time is achieved only for dilute samples at liquid helium temperatures. In fact, all known low-temperature echo-experiments were performed with Tm:YAG doped with 0.1–0.5 at.% Tm³⁺. However, in specific cases, high doping levels may also be useful. For example, consider the

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code division multiple access in a photon-echo-based-multichannel optical storage [19–21]. In general, code channel division is based on the fact that each subscriber is given his own alphabet of noiselike signals (or code sequences), by means of which he transfers information. Division is possible as the signals from different subscribers differ in their forms, which allows one to organize simultaneous work of many subscribers in a collective frequency band. In the case of photon-echo-based storage, in order to organize the code division multiple access it is necessary to use the regime of accumulated photon echo [22]. Let the first (reference) pulse in each excitation pulse pair has its own phase modulation (code) and the reading pulse has one of these possible phase modulations. In this case, if the codes satisfy some orthogonality condition, the signal of accumulated echo reproduces only one of the object pulses provided that the phase modulation of the reading pulse coincides with that of the corresponding reference pulse. However, this is possible only if the time interval between the excitation pairs is longer than the phase memory time. Otherwise, all pulses that follow a reference pulse form a long object pulse which is reproduced upon read-out of information. In such a situation the phase memory time should only be longer than the duration of each excitation pulse pair. On the other hand, the lifetime of the excited state, T_1 , should remain as large as possible, since the ratio T_1/T_M determines the maximum number of channels. Otherwise, we are forced to use long-lived accumulated regime of photon echo, which is suited for long-storage applications but not for fast processing of information. In this work we demonstrate that highly doped Tm:YAG satisfies both conditions and may be used in optical echo-processors with code division multiple access.

2. Experimental setup

A Tm:YAG crystal with 10 at.% doping of Tm and with a thickness of 250 μm was used for the experimental investigations. The crystal was kept in liquid helium at 1.8 K. The structure of energy levels of Tm ions in YAG, which was investigated in [23,24], is shown in Fig. 1.

Signals of two-pulse, stimulated and accumulated photon echoes were excited and generated at 793.15 nm on the transition ${}^3\text{H}_6(1) - {}^3\text{H}_4(1)$. The corresponding profile of the absorption line which was measured in our highly doped crystal is also shown in Fig. 1. Since nuclear spin of Tm ions is equal to 1/2, there is no hyperfine splitting due to pseudo quadrupole interaction and the hyperfine levels are degenerate at zero magnetic field. Therefore, the long-lived stimulated photon echo (LLSPE) may be observed in the absence of an applied magnetic field due to metastable states ${}^3\text{F}_4$ (5556 cm^{-1}) and ${}^3\text{H}_5$ (8530 cm^{-1}) only.

The scheme of our experimental setup is shown in Fig. 2.

The basic element of the setup is a CW ring Ti:sapphire laser TIS-SF-07 (TEKHNO SCAN, Novosibirsk). The laser may be tuned in the range of 750–950 nm with

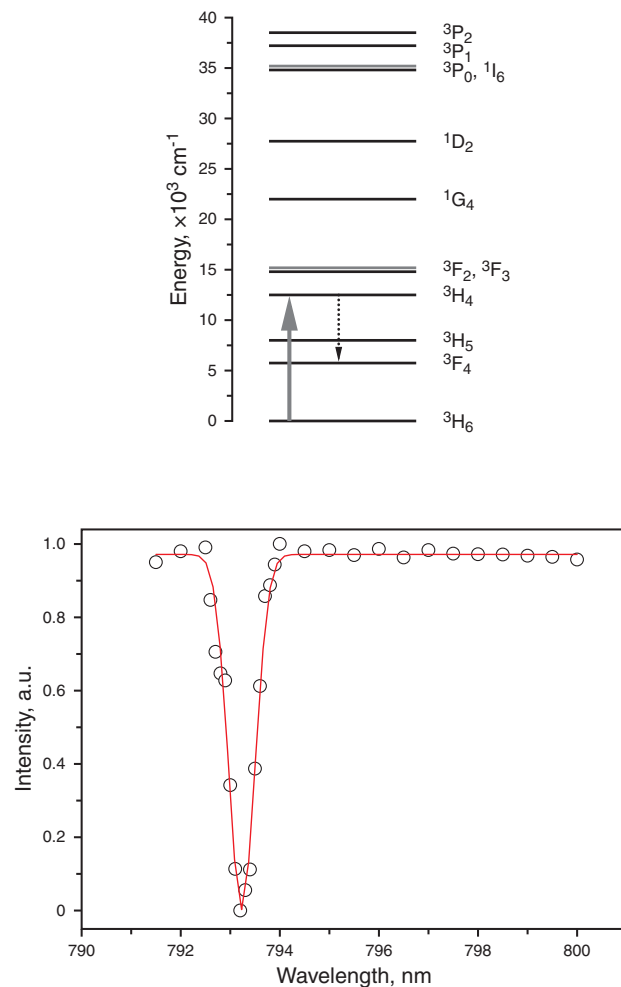


Figure 1 (online color at www.lphys.org) Energy level diagram for Tm in YAG [24] showing filling of metastable ${}^3\text{F}_4$ state after optical excitation (above) and absorption spectrum at 793.15 nm corresponding to the transition ${}^3\text{H}_6(1) - {}^3\text{H}_4(1)$, which is measured in the highly doped crystal at liquid helium temperature (below)

a linewidth less than 2 MHz and an output power of 1 W at 800 nm. A CW single-mode Ar laser Ar-5,5-150 (INVER-SIYA, Novosibirsk) is used as a pump. Excitation pulses are formed by an acousto-optical modulator AOM1. The laser power at the cryostat is 50 mW. The photon echo radiated in the forward direction is passed through a second acousto-optic modulator AOM2 to reject the excitation pulses and is detected with photomultipliers FEU-79 or FEU-143 depending on the spectral range. The photon counter is able to perform the measurements at a single-photon intensity level and to accumulate the signals with a repetition rate of 1 kHz. A control system which is combined with the photon counter [25] allows one to choose necessary durations of excitation pulses and time intervals between them as well as photon counting gate window and

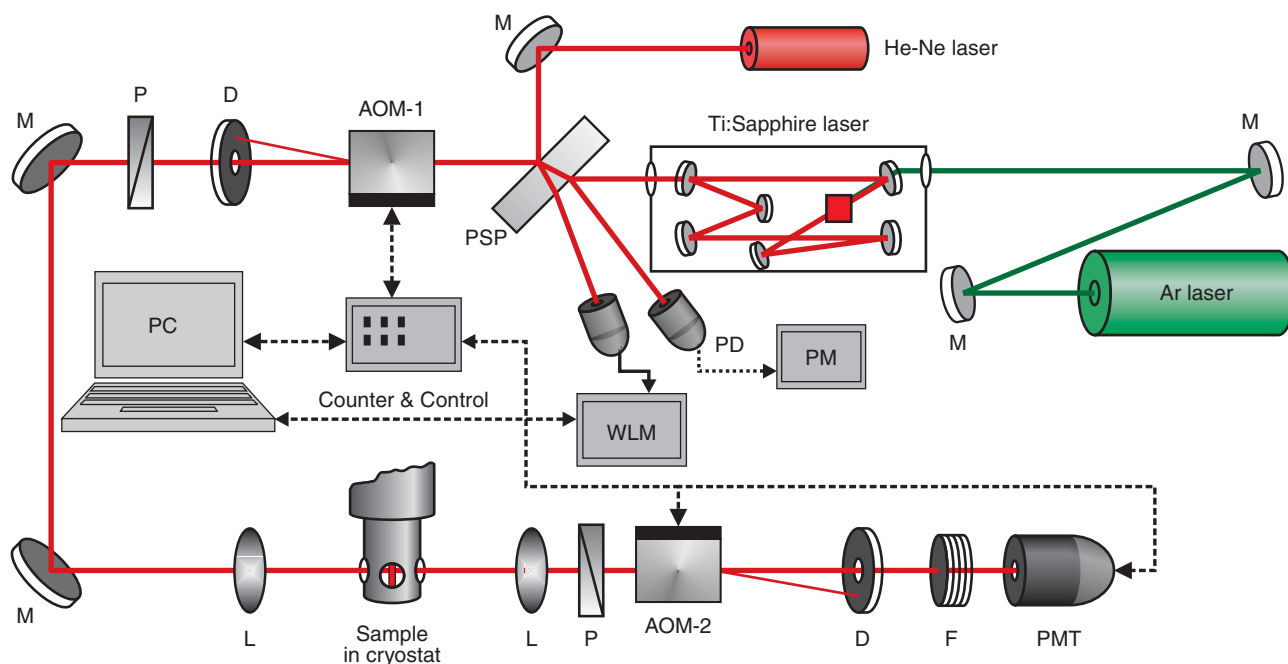


Figure 2 (online color at www.lphys.org) The scheme of experimental setup. M is a mirror, D – a diaphragm, F – a filter, L – a lens, P – a polarizer, PD – a photodiode, PM – a power meter, WLM – a wavelength meter, PSP – a parallel-sided plate, PMT – a photomultiplier

the number of accumulated signals. The time order of excitation pulses used in our experiment for observation of two-pulse, stimulated and accumulated photon echoes is shown in Fig. 3.

3. Experimental results and discussion

Two-pulse and stimulated photon echo decays at zero magnetic field, and the intensity of accumulated photon echo as a function of the number of excitation pulse pairs were investigated. The results are presented in Fig. 4 and Fig. 5.

First, consider the two-pulse decay curve (Fig. 4a). Analysis shows that the decay is nonexponential and can not be approximated by the usual decay law $I(\tau_{12}) \sim \exp(-4\tau_{12}/T_2)$, where T_2 is the phase relaxation time, I – the intensity of echo-signal and τ_{12} – the time interval between excitation pulses (see, for example, [26, 27]). A similar nonexponential decay was observed in a weakly doped Tm:YAG by R.M. Macfarlane [15] and interpreted in terms of a spectral diffusion model involving a distribution of nuclear-spin-flip rates of aluminum. The decay may be fitted to an expression of the form

$$I(\tau_{12}) \sim \exp \left[- \left(\frac{4\tau_{12}}{T_M} \right)^x \right], \quad (1)$$

first proposed by Mims [17] for the analogous situation in electron-spin resonance. Here T_M is a phase memory time and x is an exponent that depends on the details of the

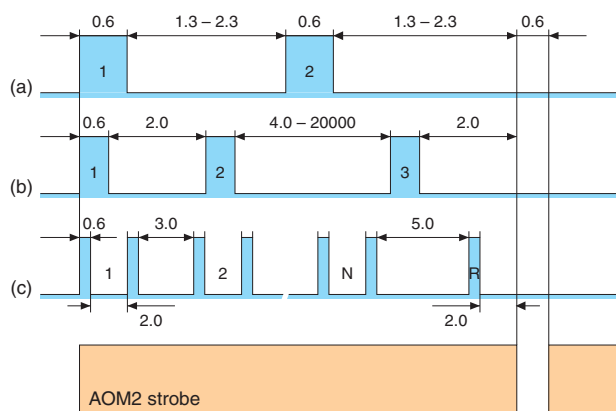


Figure 3 (online color at www.lphys.org) The scheme of excitation of two-pulse (a), stimulated (b), and accumulated (c) photon echoes. In the first two cases, the bold numbers 1, 2, and 3 enumerate excitation pulses. In the third case, the numbers 1, 2, ..., N enumerate excitation pulse pairs and R is the reading pulse. All time intervals are given in μs

nuclear-spin dynamics (T_M is equivalent to T_2 if $x=1$). In the case of YAG containing 0.1% Tm and zero magnetic field, $T_M=75 \mu\text{s}$ and $x=1.5$ [15]. Besides Tm-Al cross relaxation, there are mutual Tm-Tm spin flip-flops which become more significant with increasing concentration of Tm dopant ions. As a result, the phase memory

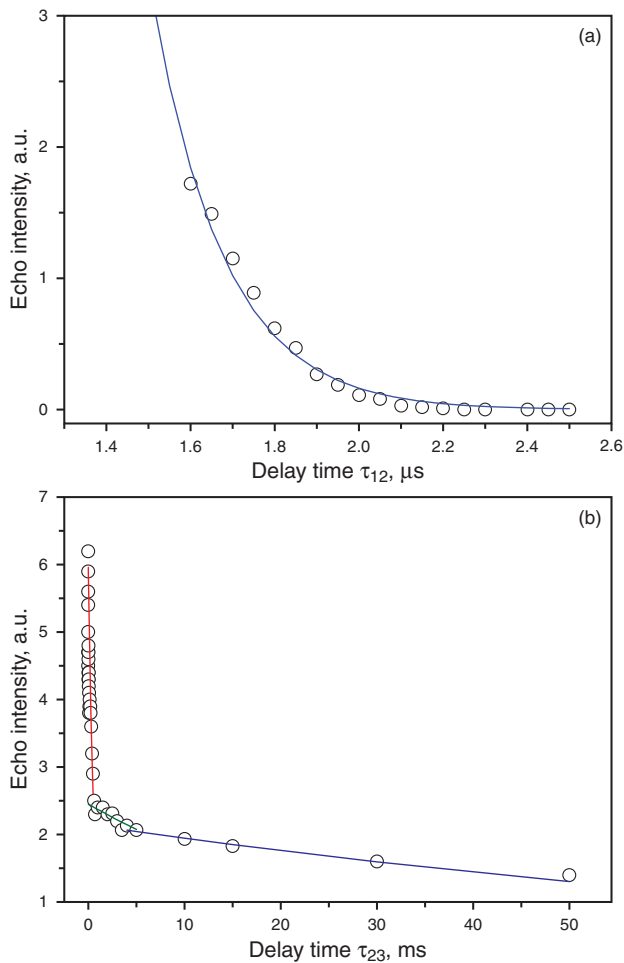


Figure 4 (online color at www.lphys.org) Two-pulse photon echo decay (a) and stimulated photon echo decay (b) in Tm:YAG (10 at.%) at 1.8 K and zero magnetic field. The second dependence is measured at a fixed time interval between the first two excitation pulses, $\tau_{12} = 2050$ ns; τ_{23} is the separation between the second and third pulses

time should be shorter at high dopant levels. In our crystal containing 10 at.% Tm, the decay is fitted to Eq. (1) with $T_M = 1.4$ μ s and $x = 1.3$ giving the solid theoretical line.

The stimulated echo decay curve which is depicted in Fig. 4b is now considered. The curve has characteristic knees that are typical for LLSPE [27]. The echo intensity is fitted to three exponentials giving the solid theoretical line, which correspond to decay times of 600 μ s (red section), 30 ms (green section), and 100 ms (blue section). The first decay time is simply the lifetime of the $^3H_4(1)$ excited state of the transition, in agreement with the value 590 μ s obtained in [18]. The second corresponds to the lifetime of the intermediate 3F_4 metastable state, which is populated due to the fast decay of the excited state. The value obtained in our experiment, 30 ms, is of the order of those

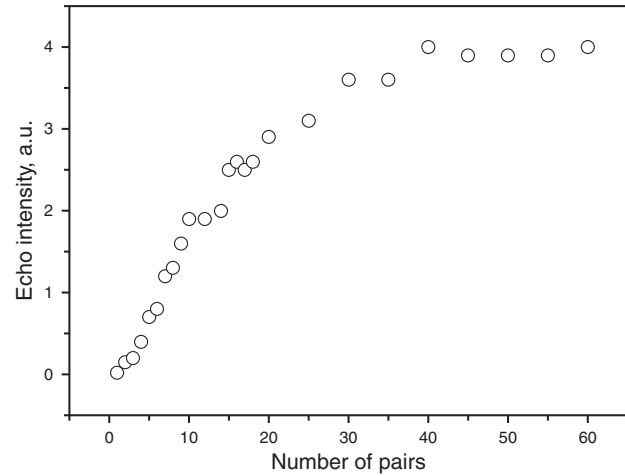


Figure 5 The intensity of accumulated photon echo as a function of the number of excitation pulse pairs in Tm:YAG (10 at.%) at 1.8 K and zero magnetic field

(10–12 ms) obtained in [15, 18]. The third, longest decay part corresponds to an energy shift arising from the coupling of Tm to the nuclear spins of lattice Al ions [18] and agrees well with the value 90 ms obtained in this work.

Finally, consider the intensity of accumulated photon echo as a function of the number of excitation pulse pairs (Fig. 5). The observed cumulative effect is about 4, which corresponds to incoherent sum of stimulated echo signals generated by different excitation pulse pairs due to the instability of the delay line [28]. This is not the optimal regime of accumulation from the viewpoint of increasing output signals [22, 29], but it is of no importance for the code division scheme mentioned in Introduction. Thus, in the case of highly doped Tm:YAG, the lifetime of the excited state, T_1 , remains approximately the same as in the case of dilute samples, while the phase memory time, T_M , proves to be fifty times shorter. Therefore, highly doped crystals are suited for multichannel coherent signal-processing with code division multiple access. By choosing appropriate doping level it is possible to optimize the phase memory time thereby minimizing the separation between excitation pulse pairs.

4. Conclusion

In summary, experimental setup which allows one to detect weak echo-signals in the regime of photon counting has been elaborated and the echo-experiments were performed with a highly doped Tm:YAG crystal (10 at.%) for the first time. Two-pulse and stimulated echo decays on the $^3H_6(1) - ^3H_4(1)$ transition were measured and accumulated photon echo was observed. It is shown that such a crystal may be useful for multichannel optical storage

and coherent optical signal-processing with code division multiple access.

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