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Nonlinear transmission of single-wall carbon nanotubes in heavy water at a wavelength of 1.54 μm and self-mode locking in a Er^{3+} : glass laser obtained using a passive nanotube switch

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***Abstract*. The nonlinear transmission of light by single-wall carbon nanotube suspensions in D_2O at a wavelength of 1.54 μm is studied. For the 250-ns probe pulse and the 40-MW cm^{-2} maximum intensity of radiation incident on a cell, the decrease in the absorption coefficient was 3.6 cm^{-1} while the absorption coefficient in a weak field was 17 cm^{-1}. *Q*-switching and self-mode locking were obtained in a pulsed Er^{3+} : glass laser by using a cell with the nanotube suspension as a passive filter.**

Keywords: *nanotubes, optical nonlinear effects, mode locking.*

1. Introduction

Single-wall carbon nanotubes (SWNTs) are now being extensively investigated. The studies are devoted to the improvement of the nanotube production techniques [1] and analysis of their electronic structure [2], which is revealed in the absorption, luminescence [1, 3 – 6] and resonance Raman spectra. Traditional synthesis technologies [7 – 9] provide the fabrication of nanotubes combined in bundles with different geometry and different conduction types. A great progress in the fabrication of single SWNTs was achieved by exposing nanotube suspensions in water with surfactants to intense ultrasonication followed by centrifuging allowing the mass-separation of fragments of a dispersed material [1]. This technology makes it possible to obtain well-reproducible nanotube suspensions in water containing a considerable percentage of individual nanotubes [1]. The presence of individual nanotubes is characterised by a set of well-resolved peaks observed in the absorption, luminescence [1, 4, 6], and Raman spectra.

Studies of the relaxation dynamics of electronic excitations in SWNTs [10, 11] revealed several electronic relaxation times lying in the range between 50 fs and 15 ps. This range is of interest for the development of various SWNT-based optical elements for applications in fast passive switches for lasers. Note here that, to our knowledge, such dye switches in the region of 1.5 μm are not available at present. Self-mode locking obtained in an erbium-doped fibre laser using an SWNT passive switch was first reported in Ref. [12].

In this paper, we study the nonlinear transmission of light by SWNTs and the possibilities of using an SWNT passive switch in a flashlamp-pumped 1.54-μm pulsed Er^{3+} : glass laser.

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2. Nonlinear transmission of light by the SWNT suspension in D_2O at a wavelength of 1.54 μm

We used in experiments unpurified SWNTs obtained by the HiPCO method (high-pressure catalytic CO decomposition) [13]. A nanotube suspension was prepared in the solution of a surfactant in heavy water. To disintegrate nanotube bundles, the suspension was exposed to ultrasound for an hour and subjected to centrifugal separation (170 000 *g*). The upper part of the solution containing a great percentage of single nanotubes was used in experiments. The preparation procedure and characteristics of the solution obtained in this way are described in more detail in Ref. [6].

Figure 1 shows the absorption spectrum of the SWNT mixture in D_2O. The absorption band at ~ 2 μm (Fig. 1a) belongs to heavy water. One can see from Fig. 1b that the wavelength 1.54 μm corresponds approximately to the absorption maximum of one of the types of carbon nanotubes.

We measured the dependence of the transmission of light in a cell with the SWNT suspension in D_2O on the 1.54-μm radiation intensity. The cell thickness was $L = 100$ μm, its transmission in a weak field was 78.5 % (the cell ends had no antireflection coatings and their transmission was 93 %), the transmission of a layer with SWNTs was 84.4 %. Measurements were performed using a Er^{3+} : glass laser operating in the active *Q*-switching regime obtained with the help of lithium niobate electro-optic switch. The laser emitted 3.0 – 3.5 mJ, 250-ns pulses in the TEM_{00} mode.

The transmission of light in a cell was measured as the ratio of the pulse energy transmitted through the cell to the incident pulse energy. The incident radiation was focused by a lens with a focal distance of 30 cm. The beam diameter in the cell and, hence, its intensity could be varied by placing the cell at different distances from the lens. The ratio of the beam cross section to that of the beam waist changed in our experiment by more than two orders of magnitude. We also measured the dependence of the radius of the Gaussian beam on the longitudinal coordinate. The method of

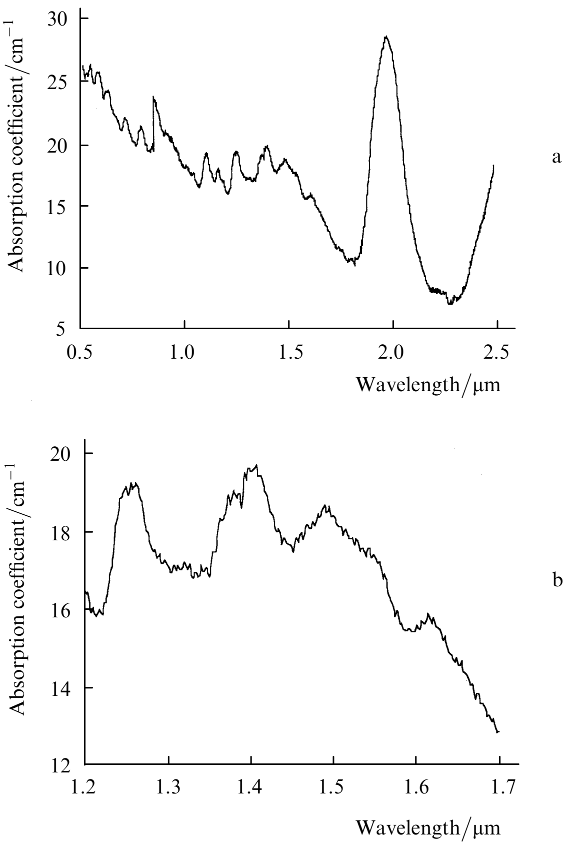

Figure 1. Absorption spectra of the SWNT suspension in D_2O in the 0.5–2.5-µm (a) and 1.2–1.7-µm (b) regions. The cell thickness is 100 µm.

variation of the radiation intensity used in our experiments reduced the influence of the probe beam energy instability on the accuracy of measurements.

Figure 2 shows the dependence of the transmission of light in the cell with the nanotube suspension on the peak intensity of incident radiation. Note that the absorption coefficient changed by 3.6 cm^{-1} when the radiation intensity incident on the cell was 40 MW cm^{-2}, while the absorption coefficient in a weak field was $\sim$ 17 cm^{-1}. The spread in the measured transmission considerably increased with increasing radiation intensity, and transmission decreased from 81.4 % (40 MW cm^{-2}) down to 79.5 % (80 MW cm^{-2}), which can be explained by the formation of a vapour-gas bubble in the cell caused by the laser beam.

The dependence of transmission on the peak intensity was calculated using the equation

$$\frac{\mathrm{d}I}{\mathrm{d}z} = -(\alpha_0 - \Delta\alpha)I - \Delta\alpha\,\frac{I}{1 + I/I_s} \qquad (1)$$

describing the pulse propagation in a medium. Here, I_s is the absorption saturation intensity; $\alpha_0 = 17$ cm^{-1} is the absorption coefficient of the suspension in a weak field; and $\Delta\alpha$ is the change in the absorption coefficient due to absorption saturation. Because the laser pulse duration was substantially longer than the excited-state relaxation time [10, 11], we assumed in the calculation that the absorption coefficient is determined by the instant radiation intensity. The second term in the right-hand side of expression (1) describes phenomenologically the known absorption saturation effect. Because in our case $\alpha_0 L \approx 0.15 \ll 1$, we can

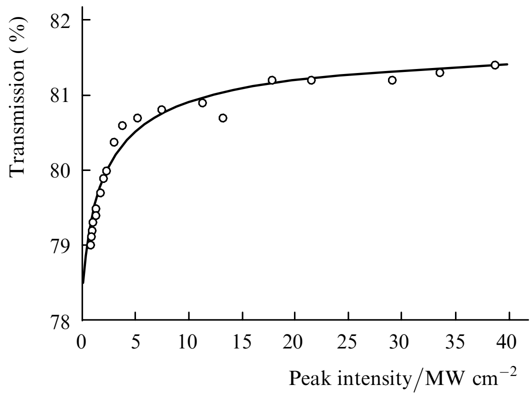

Figure 2. Dependence of the transmission coefficient of a cell with the SWNT suspension on the peak intensity of radiation incident on the cell. Circles are experimental data, the solid curve is calculation.

assume in the first approximation that the intensity I in the right-hand side of (1) is equal to the incident radiation intensity. In this case, we can easily obtain the required solution. Taking into account that the incident beam is Gaussian and assuming that the temporal profile of the beam is rectangular with the duration 250 ns, we can obtain the approximate dependence of the cell transmission T on the pulse peak intensity I_0:

$$T(I_0) = T_s^2 \times \exp\left\{-\alpha_0 L - \Delta\alpha L\left[-1 + \frac{I_s}{I_0 T_s}\ln\left(1 + \frac{I_0 T_s}{I_s}\right)\right]\right\}, \qquad (2)$$

where $T_s = 0.96$ is the transmission of one of the cell ends. For $I_s = 10^6$ W cm^{-2} and $\Delta a = 4.0$ cm^{-1} expression (2) describes the experimental data in the best way.

3. Passive *Q*-switching and self-mode locking in a Er^{3+} : glass laser obtained using a cell with SWNTs

A cell with SWNTs was used as a passive switch in a Er^{3+} : glass laser. The laser resonator of length 76 cm was formed by two mirrors with the radius of curvature of 1 m. The resonator contained an iris aperture to separate the TEM_{00} mode, an active element of diameter 4-mm and length 80 mm, and a cell with SWNTs located near one of the resonator mirrors. The cell thickness was $L = 100$ µm and the initial transmission of the SWNT layer at the 1.54-µm laser wavelength was 84.4 %. The temporal profile of the radiation pulse was recorded with an LFD-2a photodiode and a Tektronix 7104 oscilloscope with the time resolution of $\sim$ 1 ns.

Figure 3 shows the oscillograms of laser pulses at the sweep speeds 50 and 2 ns div^{-1}. One can see that the output radiation of the total duration $\sim$ 200 ns represents a train of short (shorter than 1 ns) single pulses with the pulse repetition period equal to the resonator round-trip transit time, demonstrating self-mode locking.

4. Conclusions

We have studied the dependence of the transmission of light in the SWNT suspension in D_2O on the radiation intensity at 1.54 µm. For the 250-ns laser pulse and the 40-MW cm^{-2}

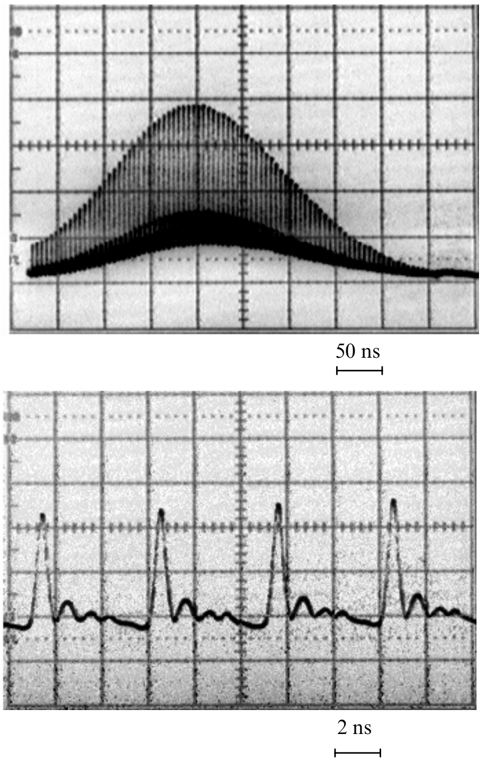

Figure 3. Output pulses of a Er^{3+} : glass laser with a SWNT passive switch.

maximum intensity of radiation incident on the cell, the absorption coefficient decreased by 3.6 cm^{-1}, while the absorption coefficient in a weak field was $\sim$ 17 cm^{-1}. These values can be compared with the absorption coefficients in Fig. 1b, where the height of a 'pedestal', which was not bleached at the radiation intensities used in experiments, can be estimated as 13 – 14 cm^{-1}, while the bleached part of absorption can be probably assigned to the 1.5-μm absorption peak. The absorption saturation intensity I_s was estimated as 10^6 W cm^{-2}.

We have obtained Q-switching and self-mode locking in a Er^{3+} : glass laser using a cell with the SWNT suspension in D_2O. The duration of mode-locked pulses was shorter than 1 ns, this value being limited by the time resolution of our detector.

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