LETTERS

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Self-mode-locking in erbium-doped fibre lasers with saturable polymer film absorbers containing single-wall carbon nanotubes synthesised by the arc discharge method

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***Abstract*. We studied the ring and linear schemes of erbium-doped fibre lasers in which passive mode locking was achieved with the help of saturable absorbers made of high-optical quality films based on cellulose derivatives with dispersed single-wall carbon nanotubes. The films were prepared by the original method with the use of nanotubes synthesised by the arc discharge method. The films exhibit nonlinear absorption at a wavelength of 1.5 μm. Pulses in the form of optical solitons of duration 1.17 ps at a wavelength of 1.56 μm were generated in the ring scheme of the erbium laser. The average output power was 1.1 mW at a pulse repetition rate of 20.5 MHz upon pumping by the 980-nm, 25-mW radiation from a laser diode. The pulse duration in the linear scheme was reduced to 466 fs for the output power up to 4 mW and a pulse repetition rate of 28.5 MHz. The specific feature of these lasers is a low pump threshold in the regime of generation of ultrashort pulses.**

Keywords: *self-mode-locking, erbium-doped fibre laser, solitons, carbon nanotubes.*

Single-wall carbon nanotubes (carbon SWNTs) are a new nanomaterial with remarkable physical properties. In particular, it was found recently that the absorption of near-IR laser radiation can be saturated in carbon SWNTs [1 – 3]. Self-mode-locking in solid-state lasers emitting at different wavelengths was demonstrated by using liquid [3 – 5] and film [1, 2, 6, 7] carbon SWNT elements. Composite carbon SWNT – polymer films [6] and carbon SWNT films synthesised or deposited on a quartz substrate [1, 2] were successfully used as saturable absorbers in fibre lasers operating in the passive mode-locking regime. Compared to other methods of self-mode-locking in lasers of this type such as the nonlinear rotation of the birefringence ellipse [8 – 10] or the use of a saturable semiconductor absorber [11, 12], the application of carbon SWNTs offer certain advantages, allowing the development of low-threshold fibre lasers by using comparatively simple and available technologies. In addition, carbon SWNTs can operate in a broad spectral range and have a high radiation resistance.

The optical parameters of films containing carbon SWNTs depend considerably on the method of synthesis of nanotubes. Therefore, the development of the manufacturing technology of carbon SWNTs with optical parameters required for the operation of ultrashort-pulse fibre lasers is an important problem of current interest.

In this paper, we proposed a new method for preparing carbon SWNT-containing films which were used to develop and study low-threshold subpicosecond erbium-doped fibre lasers.

The principal factor in the achievement of a high optical quality of media containing nanotubes is the disintegration of carbon SWNT bundles, which are most often formed during synthesis. Typically, a bundle contains 10 – 100 nanotubes of different diameters and, hence, with different bandgap widths (from 1.4 eV to 0 in metal nanotubes) [13]. Individual nanotubes in the bundle interact due to Van der Waals forces, and the bundle itself is a well-order structure demonstrating the characteristics reflexes of a one-dimensional crystal in X-ray diffraction patterns. The disintegration of bundles is efficiently produced by a high-power ultrasonic irradiation of carbon SWNTs in a solvent in the presence of surfactant molecules followed by ultracentrifugation and separation of the lightest upper fraction [14].

The type of interaction of a surfactant with a nanotube surface and its efficiency determine the concentration of single carbon SWNTs remaining in the solution. Different surfactants have been analysed earlier [15, 16] and the most efficient of them have been chosen. The use of concentrated aqueous carbon SWNT suspensions provided self-mode-locking in lasers with bulk elements [3 – 5]. We prepared films from water-soluble polymers (polyvinyl alcohol) by using also suspensions as solvents for the polymer component.

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In this paper, we used a principally new type of polymers – cellulose derivates, in particular, carboxymethyl cellulose (CMC). The advantage of this substance is that it represents simultaneously a surfactant with a high nanotube-dispersion activity [17] and a medium forming a film after the suspension drying. Thus, the number of components in the obtained material is reduced to two (carbon SWNTs and a polymer). Films based on other water-soluble polymers contain usually three components: carbon SWNTs, a surfactant, and a polymer. The presence of additional components in the film increases the level of losses caused by unsaturated optical absorption and reduces the operation efficiency of a nonlinear optical element.

When carbon SWNT films are used in fibre lasers requiring the minimisation of losses in a matrix with dispersed carbon SWNTs, another advantage of CMC becomes important – the possibility of preparing very thin (6 – 8 μm) homogeneous films, which are much thinner than, for example, polyvinyl alcohol films.

As far as we know, in this study we report for the first time the manufacturing of a saturable cellulose absorber. In addition, a new feature is the use carbon SWNTs synthesised by the arc discharge method. 'Arc' nanotubes have absorption bands shifted to the longer IR region (up to ~ 2.0 μm) than the absorption bands of nanotubes synthesised by other methods [high-pressure decomposition of the CO gas (HipCO), laser ablation method, etc.].

Single-wall carbon nanotubes were synthesised by the arc discharge method in the helium atmosphere by using the $Ni - Y_2O_3$ catalyst in the $C:Ni:Y_2O_3 = 2:1:1$ mixture filling a graphite anode. The arc current was 90 A [18].

Stable carbon SWNT suspensions were prepared by the dispersion of raw soot, containing 'arc' carbon SWNTs, in the CMC solution in ordinary or heavy water by ultrasonic irradiation followed by centrifugation at an acceleration of 150000 g for an hour (an Optima Max-E Beckman-Counter centrifuge). The homogeneous solution of the supernatant was a stable suspension of individual carbon SWNTs and was used to prepare films [15, 16].

Carboxymethyl cellulose films of the optical quality containing carbon SWNTs synthesised by the arc discharge method were prepared by pouring the carbon SWNT suspension on a immobile substrate in the 1 % (in mass) aqueous solution of the sodium salt of CMC (of the average viscosity) (Sigma), which was followed by a slow evaporation of the solvent in a vessel. The thickness of films was varied from 10 to 100 μm. Films of the required thickness with the needed carbon SWNT concentration were prepared by varying the volume ratio of the carbon SWNT suspension and pure polymer.

The optical properties of the films in the spectral range between 190 and 2200 nm were recorded with a Shimadzu UV-3600 spectrometer. The distribution of carbon SWNT diameters in the film was estimated from the characteristic frequencies of 'breathing' modes in the low-frequency region of Raman spectra. Raman spectra were excited by different wavelength of an argon laser and recorded with a triple Jobin-Yvon S-3000 spectrometer. We have found that the sample contains nanotubes of diameters from 1.2 to 1.7 nm.

Figure 1 presents the absorption spectrum of the CMC film with dispersed arc carbon SWNTs recorded in a broad spectral range. The spectrum exhibits distinct absorption bands corresponding to transitions between the first, second, etc. symmetric van Hove singularities in the density of the electronic states of an ensemble of nanotubes. The width of the absorption bands is determined by the width of the nanotube diameter distribution. Considerably narrower bands correspond to absorption of light by single nanotubes of different geometries [13]. Optical transitions are allowed between symmetric singularities. The operating wavelength of the erbium laser lies within the first absorption band of semiconductor nanotubes.

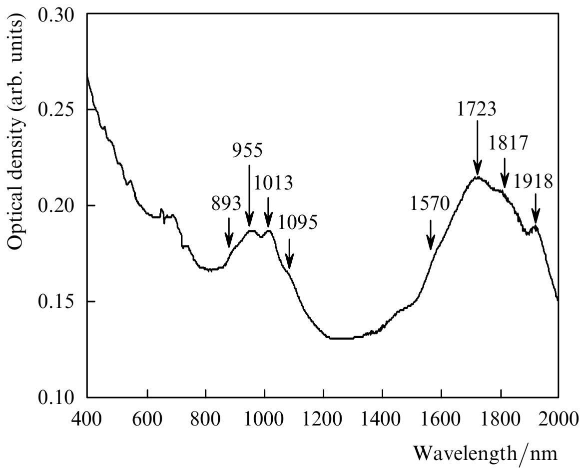

Figure 1. Absorption spectrum of the carboxymethyl cellulose film containing homogeneously distributed individual carbon SWNTs synthesised by the arc discharge method. The narrow absorption peaks denoted by arrows correspond to transitions between the first and second van Hove singularities in the density of one-electron states.

Note that the positions of absorption bands of carbon SWNTs dispersed in CMC differ slightly from their positions in the initial suspension. This property can be further used to tune the absorption spectrum to a particular wavelength of a fibre laser.

The saturable absorption in CMC films containing carbon SWNTs was measured at a wavelength of 1.560 μm by the z-scan method by using an Avesta-Project EFO-150 femtosecond erbium-doped fibre laser. The saturable absorption was 2.6 % for the peak radiation intensity of 2 MW cm^{-2}. Such films were used to obtain cw mode locking in erbium-doped fibre lasers. Figure 2 shows the schemes of lasers under study. In the ring scheme, a film was placed between the ends of fibres in a standard FC/APC connector. To prevent the influence of parasitic reflections on the mode-locking regime, the fibre ends were cut at an angle of $\sim 7°$. In the linear scheme, the film was pressed by the fibre end to a mirror with the 100 % reflection coefficient.

In the ring scheme, an erbium-doped fibre of length 1.25 m was used as the active medium. The fibre was pumped through a multiplexer by the 980-nm radiation from a laser diode in the propagation direction of laser radiation in the ring. The unidirectional lasing regime was provided by placing an isolator in the scheme. Radiation emerging from the erbium-doped fibre passed through a coupler, which provided the 50 % output of radiation; the rest of the radiation was incident on a saturable absorber. A module containing carbon SWNTs was placed immediately behind the output coupler to reduce the power of radiation incident on it. To obtain the negative group-velocity dispersion (GVD) required for producing soliton pulses, a SMF-28 fibre of length 8.5 m was used.

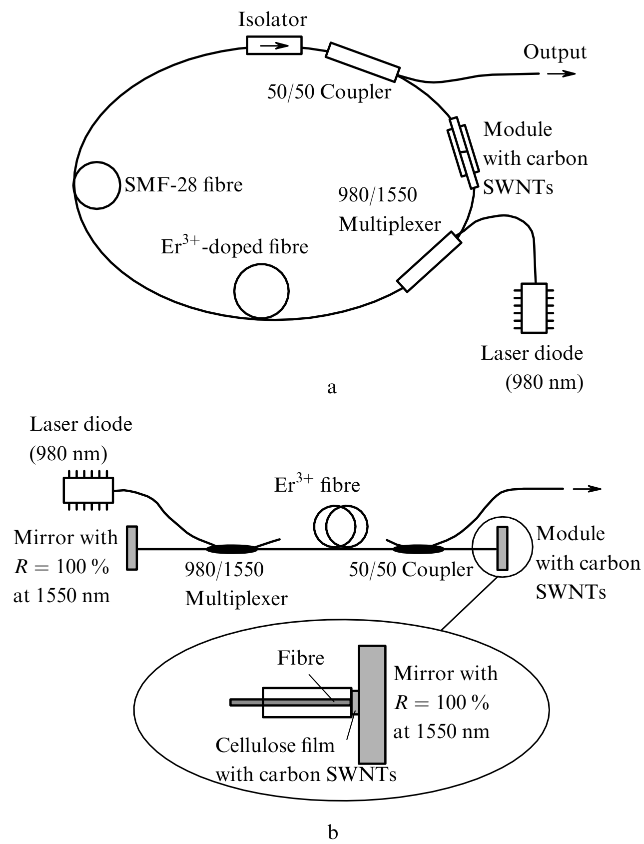

Figure 2. Schemes of ring (a) and linear (b) erbium-doped lasers.

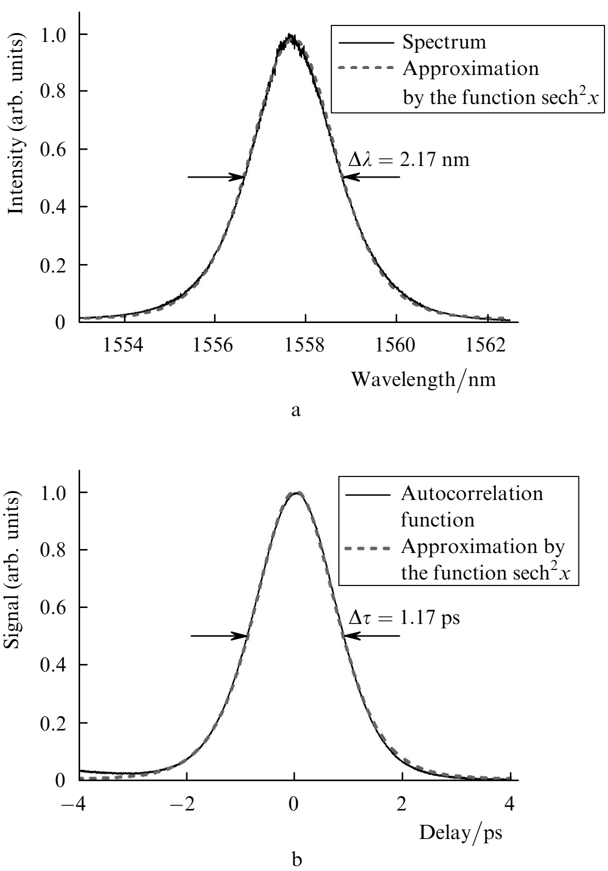

Figure 3. Spectrum (a) and the autocorrelation function of the ring laser radiation intensity (b).

Upon 25-mW pumping, stable lasing with a single pulse per the round-trip time in the resonator was achieved with the average output power of 1.1 mW. The pulse repetition rate was 20.5 MHz. Figure 3 shows the spectrum and autocorrelation function of the ring laser radiation intensity. One can see that they are well approximated by the function $\mathrm{sech}^2 x$. It follows from the results of measurements that $\Delta\tau\Delta\nu = 0.3159$, where $\Delta\tau$ is the pulse duration and $\Delta\nu$ is the width of the spectrum. For a transform-limited soliton pulse $\mathrm{sech}^2 x$, this value is 0.3148. Thus, we can assert that the laser emits transform-limited soliton-like pulses.

In the linear scheme, we used an erbium-doped fibre of length 90 cm and SMF-28 fibre pieces to compensate for the GVD. One end of the fibre was pressed to a mirror with the 100 % reflection at 1550 nm, while the other was pressed to the same mirror but through the CMC film containing carbon SWNTs. For the pump power of 40 mW, the output power achieved 4 mW at a pulse repetition rate of 28.5 MHz. Figure 4 shows the spectrum and autocorrelation function of the output radiation intensity. The pulse duration was 470 fs and its spectral width was 11 nm. It follows from these data that pulses have a chirp, which means that pulses of even shorter duration can be obtained after a complete compensation of the chirp.

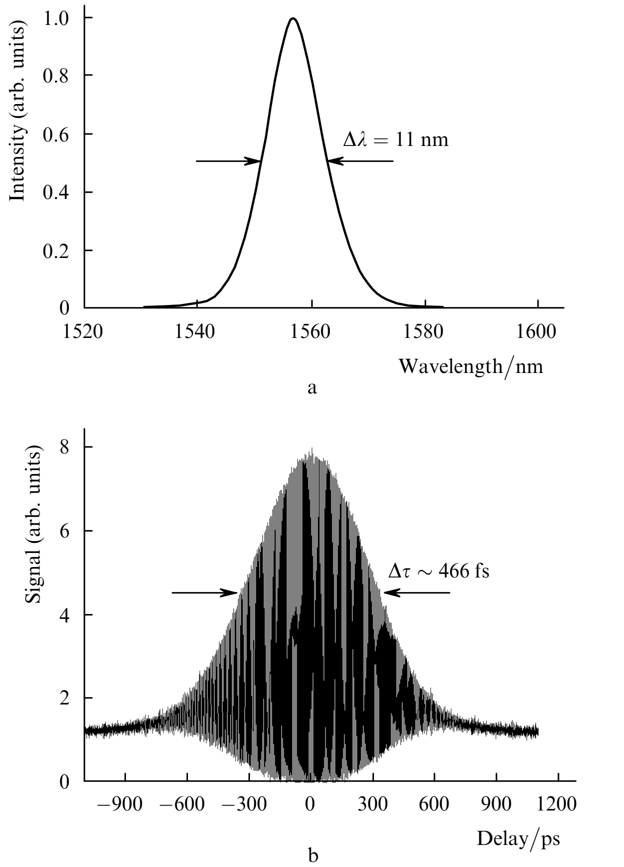

Figure 4. Spectrum (a) and the autocorrelation function of the linear laser radiation intensity (b).

Thus, we have fabricated an erbium-doped fibre laser with ring and linear resonators generating subpicosecond pulses at low pump powers due to the use of saturable absorbers of a new type – CMC films prepared by the method developed by us, which contain homogeneously dispersed carbon SWNTs synthesised by the arc discharge method.

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