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Effect of intracavity light-erosion plasma on the output parameters of a tunable laser

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***Abstract*. The possibility of using intracavity laser spectroscopy for studying the formation of carbon and metallocarbon fullerene molecular clusters synthesised in light-erosion heterogeneous plasma is analysed. The condensation of the emission spectrum of a tunable laser was used as an indicator of formation of a fullerene mass in the plasma.**

Keywords: intracavity laser spectroscopy, condensation of emission spectrum, fullerene molecular clusters.

The effect of so-called spectral condensation (SC) was observed [1–5] in the studies of absorption spectra of alkali metal vapours by the method of intracavity laser spectroscopy. This effect is manifested in an anomalous spectral amplification of emission from a tunable laser in the region of absorption lines of objects under study. The properties of SC caused by the presence of excited and ionised rare-earth metal atoms in the heterogeneous plasma were investigated in Refs [6–9].

In Ref. [10], this effect was observed for the first time in the presence of fullerene C$_{60}$ in an electric arc flame. It was also found in [10] that the character of the appearance of condensation lines upon additions of impurities of C$_{60}$ and cerium (or of other rare-earth metals) to the plasma was similar. This suggests that the fullerene or metallofullerene clusters produced in the electric-arc discharge plasma favour the appearance of SC. Therefore, it seems that the SC effect can be used for a quick qualitative monitoring of the formation of fullerenes, i.e., the features of SC can reflect to some extent the efficiency of the fullerene-mass formation (fullerenes and metallofullerene nanotubes) in a heterogeneous plasma.

Fullerenes are mainly produced by synthesis in the heterogeneous plasma of a high-current electric discharge or in a light-erosion torch appearing upon irradiation of a target by laser radiation. The parameters of a light-erosion plasma (concentrations of electrons and ions, their temperatures, etc.) vary in a broad range, which makes it possible to monitor the dynamics of the main stages and features of the development of SC during the existence of a plasma torch, which is the aim of this work.

This paper presents the results of the study of the appearance, development, and reproduction of SC in a tunable laser with an intracavity light-erosion torch. A heterogeneous plasma was produced by irradiating targets made of pure carbon or carbon with additions of cerium by a neodymium laser operating in repetitively pulsed, quasi-continuous, and free-running regimes.

A light-erosion plasma torch propagated from a target perpendicular to the optical axis of the resonator of a tunable Cr^{3+} : GSGG laser representing a spectrum analyser with a sensitivity of measuring the absorption coefficient $\chi \leqslant 10^{-7}$ cm^{-1}. The nonselective resonator of the spectrum analyser of length up to 50 cm was formed by two broadband mirrors with skewed rear faces and reflectivity $\sim 99\%$ at the lasing wavelength. The Cr^{3+} : GSGG crystal ($\varnothing 6 \times 65$ mm) placed inside a silver-plated reflector was excited by a 500-J xenon lamp and generated a `smooth' pulse with energy $E \leqslant 50$ mJ and duration $\tau \leqslant 150$ μs.

The neodymium laser had the energy $E \leqslant 30$ J and pulse duration $\tau \leqslant 1$ ms in the quasi-continuous and free-running regimes and $E \leqslant 14$ J and $\tau \leqslant 0.8$ ms (the energy and pulse duration were determined by averaging over ten measurements) in the repetitively pulsed regime, which was achieved by using intracavity LiF : F_2^- modulators. In the latter case, the duration of an individual pulse was varied from 60 to 100 ns, and the distance between the pulses was varied from 10 to 50 μs (by using modulators with different optical densities). The radiation power on the target was increased by factors of 50–240 by focusing an incident laser beam. The scheme of the experimental setup is shown in Fig. 1, and pulses emitted by the neodymium laser in different operating regimes are displayed in Fig. 2.

To obtain a complete picture of the development of SC, the method of `temporal sections' [8] was used, which allows

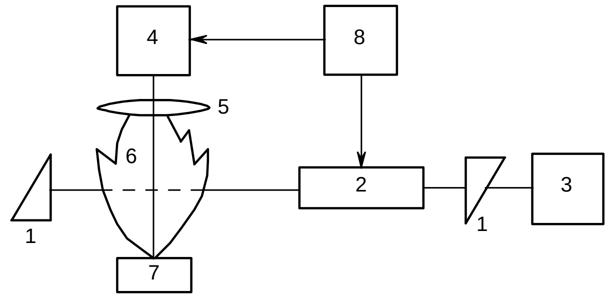

Figure 1. Scheme of the experimental setup: (1) cavity mirror; (2) Cr^{3+} : GSGG laser; (3) polychromator; (4) neodymium laser; (5) lens; (6) light-erosion torch; (7) target; (8) generator of paired pulses.

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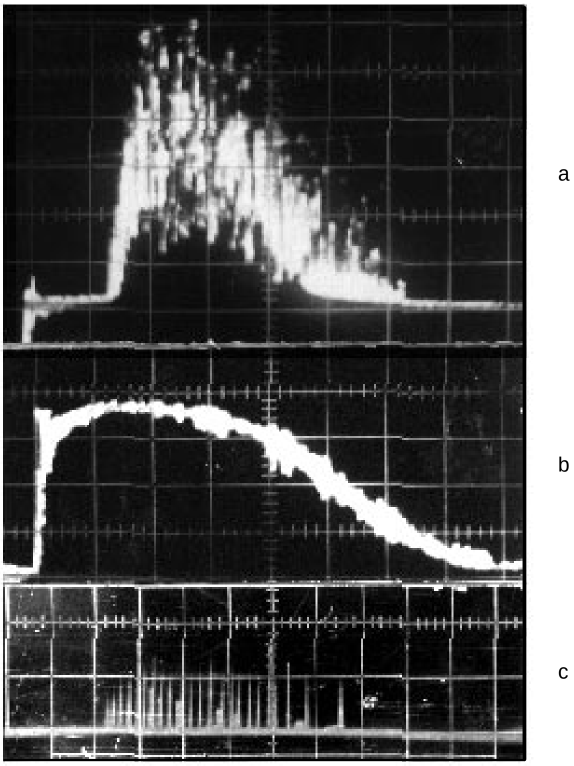

Figure 2. Radiation pulse from a neodymium laser operating in free-running (200 μs/div) (a), quasi-continuous (100 μs/div) (b), and repetitively pulsed (50 μs/div) regimes.

the detection of the dynamics of the emission spectrum of a Cr^{3+} : GSGG laser during its interaction with a light-erosion torch at fixed instants of time. The temporal and energy characteristics of laser pulses and the emission spectrum of a tunable laser were detected.

The appearance of SC was always accompanied by a strong change in the shape and duration of pulses from the tunable laser. This was earlier observed in Refs [6–9] in the form of regular spikes (or a single spike) of a short duration of a few nanoseconds or several tens of nanoseconds emitted by a Cr^{3+} : GSGG laser. In this case, the laser output decreased only weakly, no more than by an order of magnitude. Therefore, the peak output power of the tunable laser increased by a few orders of magnitude in the case of SC.

The emission spectrum of the tunable laser changed in the presence (or formation) of the fullerene mass (fullerenes and metallofullerenes) in the heterogeneous plasma. In this case, a broadband (several hundreds of Ångström) emission spectrum transformed to groups of very intense lines (line heads accompanied by satellites, Fig. 3) or single lines with the half-width $\delta\lambda \sim 10^{-1} - 10^{-3}$ nm. When a complete SC was achieved [7–9], lasing occurred only at a few lines whose intensity increased by several orders of magnitude. Therefore, the heterogeneous plasma acquired dispersion and phototropic properties when fullerene molecular clusters were synthesised in it. This was most distinctly manifested in the presence of rare-earth metals in the plasma.

Fig. 3 shows typical variations in the emission spectrum of the Cr^{3+} : GSGG laser upon its interaction with a light-erosion torch from different targets for the three operating regimes of the neodymium laser. It was found that the influence of the dynamics of variations in the pulse and power of the neodymium laser incident on the light-erosion torch on the appearance of condensation lines (Fig. 3, lines 24–30) depends on the off-duty ratio of short pulses from the neodymium laser (Fig. 2c). An increase in the time interval between the pulses, accompanied by an increase in the output power of the neodymium laser in an individual lasing spike, resulted in the red shift of the condensation lines (Fig. 3, lines 26–29).

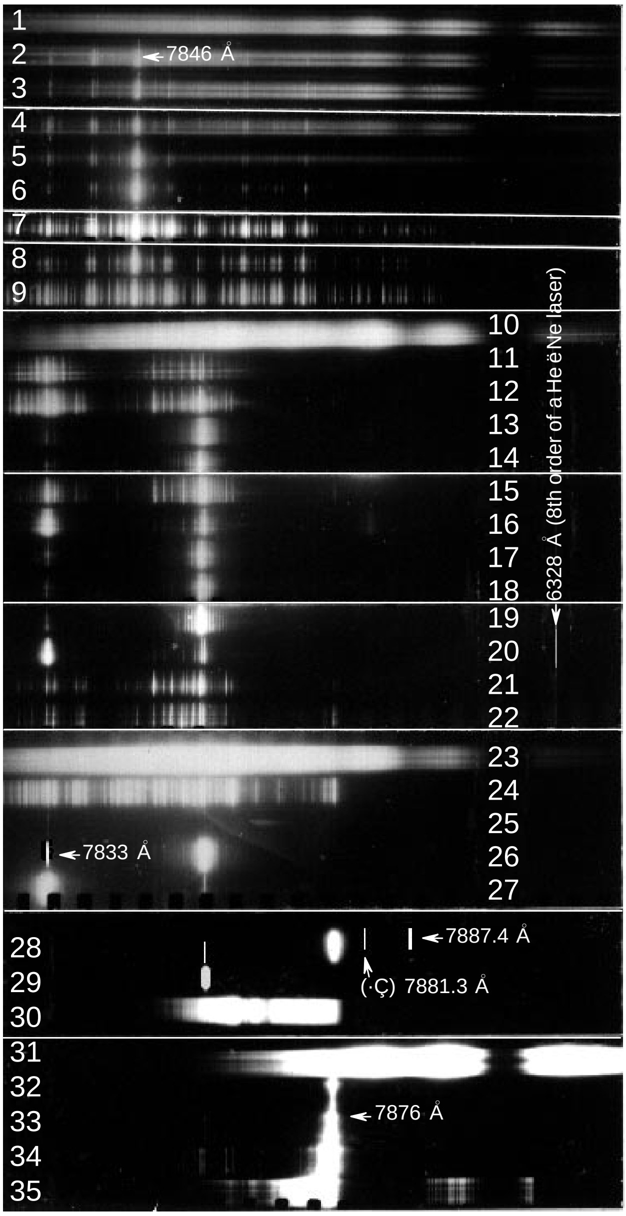

Figure 3. Emission spectra of the Cr^{3+} : GSGG laser with an intracavity light-erosion torch appearing after irradiation of target made of carbon (lines 2–9) or carbon with cerium (lines 11–22, 24–30, 32–35) by a pulsed neodymium laser operating in the repetitively pulsed regime with the distance between pulses equal to 10 (27), 20 (26), 35 (29), and 50 μs (28), in the quasi-continuous regime (32–35), and in the free-running regime (2–9, 11–22). ‘The temporal sections’ of emission spectra of the Cr^{3+} : GSGG laser were detected each 25 (2–9, 32–35) and 10 μs (11–22) (lines 1, 10, 23, 31 are the emission spectrum of the Cr^{3+} : GSGG laser in the absence of plasma in its cavity.

When a target was irradiated by the neodymium laser operating in the quasi-continuous regime, the parameters of a plasma torch (concentrations of electrons and ions, their temperatures, etc.) remained invariable, and only one condensation line was detected in the emission spectrum of the tunable laser (Fig. 3, lines 32–35). In this case, the intensity and width of the condensation line changed in time (`temporal sections' of the emission spectrum of the tunable laser were detected every ~ 25 μs). This made it possible to determine the temporal boundaries of a plasma layer (70–100 μs), which caused SC and affected the formation of condensation lines (it is assumed that the fullerene mass is produced most efficiently in this region of the plasma torch). The electron concentrations in this region, which varied from 3×10^{12} to 2×10^{13}, were measured with an interferometer.

The dependence of the appearance and type of the condensation lines on the chemical composition of targets made of carbon (Fig. 3, lines 2–9) and carbon with cerium impurities (lines 11–22) was studied. SC was detected upon the interaction of radiation from the $\mathrm{Cr}^{3+}:\mathrm{GSGG}$ laser with a light-erosion torch from a carbon target with cerium impurities for all the three operating regimes of the neodymium laser, whereas in the case of a pure carbon target, SC was detected only for free-running neodymium laser. The condensation lines caused by the torch from the carbon target and target with cerium impurities were detected in different regions. The torch from the carbon target with cerium impurities always produced intense condensation lines (cerium stimulates an increase in the fullerene mass in the heterogeneous plasma due to synthesis of metallofullerene nanotubes [11–14], whereas in the case of a purely carbon target, weak lines accompanied by satellite lines were observed (Fig. 3, lines 2–9). It seems that in the latter case, only fullerenes synthesised from evaporated pure carbon are present in the torch [11–13].

Thus, the main factors characterising the influence of the heterogeneous light-erosion plasma on the appearance and features of SC were determined in this paper. The dynamics of this phenomenon was studied by the method of `temporal sections' [8] and the main conditions for its reproduction were determined. The condensation lines were detected whose intensity was increased by several orders of magnitude. It was shown that impurities of rare-earth elements in a light-erosion torch cause the appearance of very intense condensation lines, which shift to the red with increasing radiation power of a neodymium laser on the target. The presence of rare-earth metal impurities (as fullerenes themselves) in the light-erosion torch leads to synthesis of metallofullerene nanotubes, resulting in SC. This suggests that SC can be used for studying the synthesis of molecular clusters produced in the heterogeneous plasma.

In further studies, an attempt will be made to obtain additional data on the possibility of using the SC effect for investigating the technology of synthesis of fullerenes or metallofullerenes at the stage of their formation in plasma objects. This will improve an understanding of the mechanism of formation of cluster systems in the heterogeneous plasma.

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