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Condensation of the emission spectrum of a wide-band laser in the case of intracavity emission scattering by an aerosol

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Abstract. It is found that inserting aerosol in a cavity of a multimode (wide-band) Co : MgF_2 laser can strongly affect its emission spectrum, provided the typical size of aerosol particles is comparable to the laser wavelength (~ 2 μm). In the presence of aerosol, intensity spikes are formed in the spectrum at the initial lasing stage, which develop in time. At moments of time ~ 100 μs, condensation of the spectrum was observed, which manifested itself as a sharp (more than two orders of magnitude) narrowing of the laser emission spectrum down to the value determined by the spectral resolution of a spectrograph (not better than 0.04 cm^{-1}), with an insignificant decrease in the output energy.

It is known that the emission spectrum of multimode (wide-band) lasers with a large homogeneous width of the gain profile is extremely sensitive to narrow-band frequency-dependent loss in a cavity, which was used for the development of intracavity laser spectroscopy (ICLS) [1]. For example, by placing a substance with a linear absorption spectrum in a dye laser cavity, one can detect absorption lines with coefficients as low as $\sim 10^{-11}$ cm^{-1} [2], which is inaccessible by using conventional methods of absorption spectroscopy. Using a Co : MgF_2 laser crystal in ICLS [3, 4], the authors managed to extend the method to the near-IR region, which considerably increased the number of substances that can be detected by the ICLS method.

In ICLS experiments, the condensation of the laser emission spectrum near an absorption line has been discovered [5 – 9]. The essence of this phenomenon is that an absorbing substance placed in a cavity not only causes usual intracavity absorption at the wavelength of the line being studied, which manifests itself as a dip in the emission spectrum of a wide-band laser, but, under certain conditions, causes the appearance of sharp intense spectral peaks near the frequency of the absorption band. Sometimes, the effect is so strong that the laser emission spectrum narrows and completely concentrates in such a peak. Although a common point of view on the mechanism of this phenomenon is unavailable, it is evident that condensation of the spectrum occurs near strong absorption lines.

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In the work reported here, we found that an aerosol inserted in a cavity of a wide-band Co : MgF_2 laser can very strongly affect its emission spectrum and, in particular, can cause condensation of the spectrum that is not associated with the presence of absorption lines.

The schematic of the experimental set-up is presented in Fig. 1. A Nd : $YAlO_3$ laser *1*, which operated in the free-running mode at a wavelength of 1.34 μm, was used for longitudinal pumping of a Co : MgF_2 crystal *2*, which was 32 mm long. The pump pulses were 150 μs long, and the pulse repetition rate was 1 Hz. Pump radiation was focused by a lens *3* to the crystal centre. The duration of Co : MgF_2 laser pulses was 130 μs. The laser operated at room temperature. Its cavity was formed by spherical mirrors *6* ($R = 20$ cm) and *7* ($R = 30$ cm) and was 40 cm long. The transmission of the output mirror *7* at the laser wavelength (2.25 μm) was 2%. To minimise the influence of interference effects caused by optical elements of the cavity on the shape of the emission spectrum of the Co : MgF_2 laser, the reflecting coatings were deposited onto substrates whose rear surfaces were tilted at an angle of 5°, and the faces of an active element were cut at the Brewster angle. In principle, the Co : MgF_2 laser is able to produce laser emission in the 1.6 – 2.5-μm region. The emission spectrum of our laser was determined by spectral characteristics of the mirrors. It was ~ 10 nm wide and had a centre at 2.25 μm.

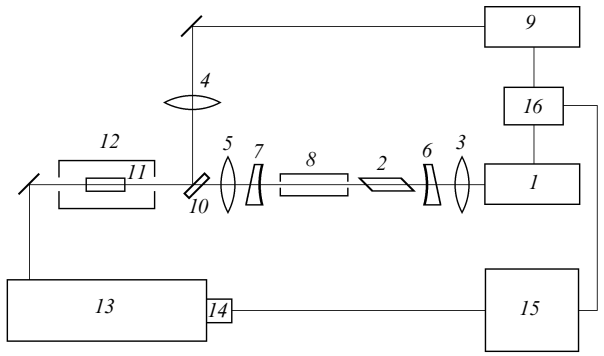

Figure 1. Schematic of the experimental set-up.
(*1*) Nd : $YAlO_3$ pump laser; (*2*) Co : MgF_2 crystal; (*3 – 5*) focusing lenses; (*6*, *7*) mirrors of the Co : MgF_2 laser cavity; (*8*) cell with an aerosol; (*9*) narrow-band Nd:YAG laser; (*10*) dichroic mirror; (*11*) nonlinear $LiNbO_3$ crystal; (*12*) oven; (*13*) diffraction spectrograph; (*14*) optical multichannel analyser on the basis of a linear CCD array; (*15*) personal computer; (*16*) synchronisation scheme.

To measure the spectral distribution of laser emission, we used a diffraction spectrograph *13* with 0.04-cm^{-1} resolution and an optical multichannel analyser on the basis of a linear CCD array *14* (a MORS-1/3648 analyser produced by the 'Multichannel Optical Recording Systems' Company), whose output signal entered a personal computer *15*. Because the range of spectral sensitivity of the CCD array was limited by a wavelength of 1.1 μm, wide-band IR emission of the Co : MgF_2 laser was preliminarily converted to the visible range. For this purpose, IR emission was mixed with narrow-band emission of a Nd : YAG laser *9* ($\lambda = 1.064$ μm, the linewidth not larger than 0.02 cm^{-1}, 5-μs pulses with energy of 1 mJ) in a nonlinear lithium niobate crystal *11*, which was heated in an oven *12* to obtain 90° phase-matching.

A dichroic mirror *10* and lenses *4* and *5* were used to bring two laser beams into spatial coincidence and to focus them into the nonlinear crystal. Radiation at the sum frequency was directed onto the entrance slit of the spectrograph *13* and was recorded. The synchronisation scheme 16 delayed a Nd : YAG laser pulse by any desired time with respect to the leading edge of a Co : MgF_2 laser pulse, which made it possible to measure the spectral distribution of radiation for the latter pulse at any moment of time using no additional instruments.

Aerosols were formed in a cell *8*, which was placed in the cavity of the Co : MgF_2 laser. The cell represented a quartz tube 80 mm long with a 32-mm inner diameter, which was closed from both sides with two Teflon covers. The Co : MgF_2 laser beam passed through two holes 4 mm in diameter that were drilled at the cover centres. Placing small amounts of different substances into the cell and heating them, we could produce aerosols. The aerosol formation was determined visually using scattering of an alignment He – Ne laser beam as a criterion. This cell was able to hold an aerosol in the laser cavity for several minutes after the termination of heating.

Fig. 2 presents spectral distributions of Co : MgF_2 laser emission at a moment of time 100 μs measured with respect to the leading edge of a laser pulse in the absence of an aerosol and in the presence of aerosol produced by heating colophony. The spectrum presented in Fig. 2a was obtained by averaging the results obtained for 100 pulses. The spectrum exhibits absorption lines caused by atmospheric methane and water vapour [10]. Fig. 2b presents a typical spectral distribution of a single laser pulse. In contrast to the averaged spectrum, it contains noise caused by quantum fluctuations of radiation intensity in separate laser modes [11, 12], but the envelope of the spectral distribution for a single pulse is similar to the averaged spectrum and contains no spikes.

The spectral distribution drastically changes in the case of the cavity containing an aerosol (Figs 2c – 2e). Figs 2c – 2e present typical spectral distributions obtained for separate laser pulses in the presence of an aerosol, which was produced by heating colophony, in the laser cavity. One can see that the aerosol causes sharp narrowing of the laser spectrum. In some cases, the spectrum was no wider than 0.04 cm^{-1} (the resolution of our spectrograph), and the spectral intensity in this case increased by approximately two orders of magnitude. We emphasise that the vapour produced during colophony heating had no effect on the laser spectrum before the appearance of aerosol, i. e., we did not observe the vapour absorption spectrum. The position of the condensed spectrum was not related to the position of absorption lines of atmospheric air and changed from one pulse to another.

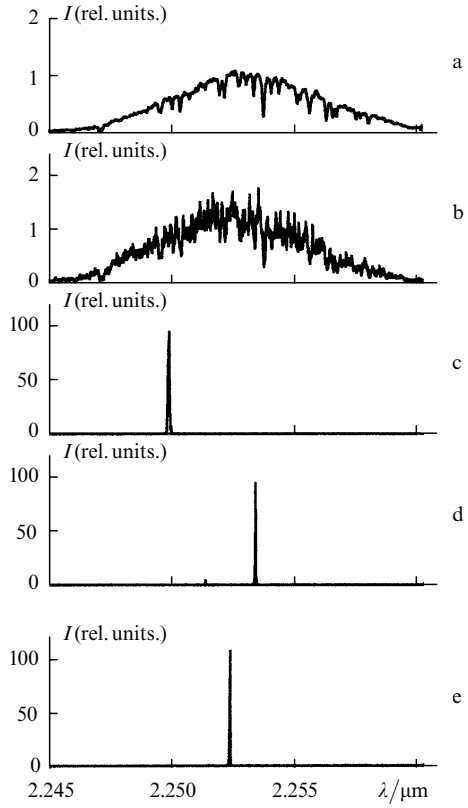

Figure 2. Spectral distributions of Co : MgF_2 laser emission at a moment of 100 μs, measured from the leading edge of a laser pulse, in the absence of aerosol obtained by averaging over 100 pulses (a) and for a separate pulse (b) and recorded in the presence of aerosol (c – e).

To estimate the loss introduced into the cavity by an aerosol, we compared the Co : MgF_2 laser energy in the presence and in the absence of aerosol. For an approximately threefold excess over the threshold, the insertion of aerosol into the cavity decreased the output laser energy by a factor of 1.2. Taking into account the transmission of the output mirror (2%) and the internal cavity loss (scattering by the mirrors, in the crystal, and from crystal faces) (1.6%), one can find that the additional loss introduced by the aerosol did not exceed 0.8% for the round cavity trip.

A similar condensation of the spectrum was also observed in the presence of aerosols produced by heating glycerol, stearic acid, and VM-1 oil for a diffusion vacuum pump. Aerosols produced by heating water and ethylene glycol did not cause the effect described above, which may be explained by the difference in the characteristic size of particles in these two groups of aerosols.

To estimate the size of particles forming an aerosol, we observed the angular intensity distribution in the diffraction pattern in the case of Fraunhofer diffraction of He – Ne laser radiation travelling through an aerosol cloud. Because the design of our experimental set-up did not allow us to perform these measurements directly in the cavity of the Co : MgF_2 laser, the quartz cell without covers was placed in a collimated beam of a He – Ne laser, and aerosol was produced in it. To observe the diffraction pattern, we used a screen positioned in the focal plane of a positive lens with focal distance of 330 mm, which was placed at the output of the cell.

Using the known expression $\sin\varphi = 0.61\lambda/r$ for diffraction by a circular screen, which relates the angular size φ

of the first diffraction maximum to the screen radius r, we estimated the size of aerosol particles. In all the cases, the diffraction pattern had no well pronounced minima and maxima, which is evidence of the polydispersion nature of the aerosols under study. Because of this, we used the angular size of the diffraction spot for estimates. Particles of water and ethylene glycol were found to have a typical radius of 5 – 10 μm, whereas the particles causing condensation of the spectrum were 1 – 2 μm in radius.

By recording the spectral distributions of Co : MgF_2 laser emission at different moments of lasing, we studied the dynamics of the development of laser spectrum in the presence of aerosol in the cavity. Unfortunately, our set-up did not allow us to record the time scan of the spectrum of a separate laser pulse. We recorded spectral distributions of emission at different moments of time for different laser pulses, which gives a qualitative description of condensation of the spectrum.

Examples of typical spectral distributions are shown in Fig. 3. The data presented there were obtained under identical conditions, i. e., when the laser spectral intensity increased due to condensation, we did not attenuate radiation incident on the entrance slit of the spectrograph. Fig. 3 shows that intensity spikes appear in the emission spectrum already at the initial lasing stage. These spikes increase in time to such an extent that the whole spectrum concentrates in a narrow spectral region, which is testified by saturation of the multichannel photodetector.

In Figs 3c – 3e, a sharp increase in the intensity due to condensation of the spectrum causes saturation of the CCD detector. In fact, we observe the development of the spectral distribution of the emission intensity of a multimode laser, which is typical of a laser with a cavity having frequency-dependent features (in particular, absorption lines in the case of measuring absorption spectra by the ICLS method). A small spectral width of spikes at the initial stage of lasing suggests that the scale of these specific features is small compared to the width of the unperturbed laser emission spectrum.

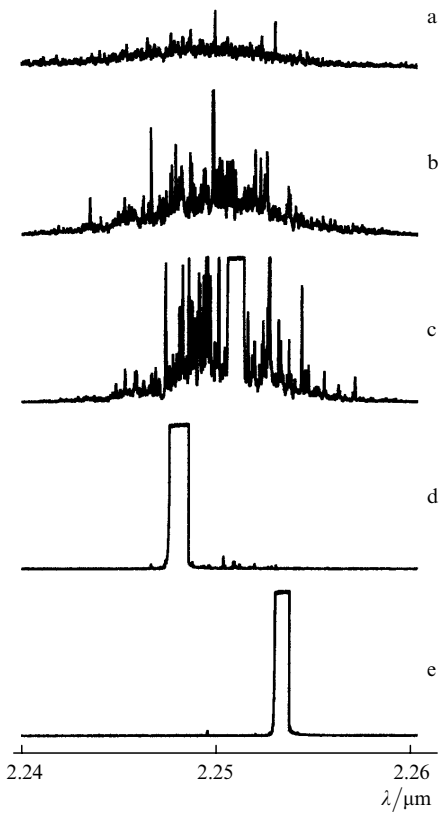

Figure 3. Spectral distributions of Co : MgF_2 laser emission obtained in the presence of aerosol in the laser cavity at moments of 25 (a), 50 (b), 75 (c), 100 (d), and 125μs (e) measured from the leading edge of a laser pulse.

In our opinion, the spectral condensation can be explained in the following way. It is known [13, 14] that the cross section for scattering of radiation with wavelength λ by spherical particles strongly depends on the parameter $2\pi\alpha/\lambda$, where a is the radius of scattering spheres. This means that an aerosol formed by such particles can act as a kind of frequency selector. Even in the case of weak selectivity, the role of such a selector in the cavity may be substantial. This is caused by the exponential development of spectral features during the laser pulse. For example, in the case of a cavity completely filled with an absorbing substance, the intensities of intracavity absorption lines increase according to the modified Lambert – Beer – Bouguer law [15]

$$\frac{I(t)}{I_0(t)} = \exp(-kct),$$

where $I(t)$ is the laser intensity at the absorption line centre at the moment t; $I_0(t)$ is the laser intensity near the absorption line; and k is the absorption coefficient at the line centre.

This means that spectral features will be observed in the spectrum for $k \sim (ct)^{-1}$, whose value for $t = 100$ μs is $\sim 3 \times 10^{-7}$ cm^{-1}. The attenuation of the signal caused by this absorption coefficient after a round trip through a cavity is equal to kL, which corresponds to $\sim 10^{-5}$ for the cavity length $L = 40$ cm. Therefore, when an aerosol produces a frequency selectivity exceeding 10^{-5} in a cavity, it leads to noticeable changes in the laser emission spectrum. It is likely that this condition was fulfilled for particles 1 – 2 μm in radius and was not fulfilled for particles of larger size. The nonreproducibility of the spectral position of the peak formed through condensation may be attributed to the nonstationary behaviour of an aerosol cloud on the one hand and the aerosol polydispersion on the other. It is likely that the picture of condensation for an aerosol consisting of identical particles will have a higher reproducibility.

We attempted to obtain condensation of the spectrum of a pulsed Rhodamine 6G dye laser (tunable within the 594 – 599-nm region with a pulse duration of 10 μs) under similar conditions, but no effect was observed for the aerosols mentioned above. This may be caused both by a increase in the parameter $2\pi\alpha/\lambda$ and by a shorten oscillation time, which may be insufficient for the condensation development.

In conclusion, we note that the specific features of spectral dynamics of multimode lasers in the presence of scattering particles in a cavity that were described here should be taken into account in the measurements by the ICLS method, especially in the IR range. Moreover, in our opinion, the phenomenon observed in the experiments can be used for narrowing, tuning, and stabilising laser radiation frequency.

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