Intracavity laser spectrometer based on a lamp pumped LiF:F_2^+ -color center laser

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An intracavity laser spectrometer at F_2^+ color centers in a lamp-pumped LiF crystal has been put in operation for the first time. It operates at a room temperature. Active centers are pumped with the help of a luminescent transformer at a Rhodamine 6G solution in ethyl alcohol. Quasicontinuous lasing has been obtained in a region 10450–10580 cm⁻¹ with a duration up to 20 µs, which has provided for a threshold absorption sensitivity of $5 \cdot 10^{-7}$ cm⁻¹.

One of the laser spectroscopy methods providing for the highest sensitivity is the intracavity (IC) absorption method. The method of broadband intracavity laser spectroscopy consists in a damping of laser radiation at absorption frequencies of the substance inside the cavity of a multimode laser.¹ The laser spectrum has sharp gaps at absorption frequencies and can be described by the equation

$$J(v, t) = J(v, t_0) \exp[-K(v)L_{\rm eff}],$$
 (1)

where $J(v, t_0)$ is the laser spectrum formed to the beginning of a lasing pulse; K(v) is the spectral absorptivity.

In IC-spectroscopy, the effective length of the absorption layer is determined by quasicontinuous lasing duration t:

$$L_{\rm eff} = ctL_{\rm a}/L_{\rm c}$$

where c is the light speed; L_a is the absorption layer length, and L_c is the cavity length.

The laser simulates a multipass absorption cell with much longer effective length L_{eff} . It equals to 300 km at a lasing duration t = 1 ms and $L_{\text{a}}/L_{\text{c}} = 1$.

The method has proved to be good in visible and near IR regions, due to multichannel photodetecting linear arrays of CCD-type well working there. In addition to a high sensitivity, the method has some other advantages: a wide spectrum (over 100 cm^{-1}) during a pulse, in which dozens of absorption lines can be simultaneously detectable; small sizes of the cell allowing a substance to be investigated under external excitation.

The color-center lasers are among the most promising ones for IC-spectroscopy. They have an extremely wide lasing band (up to 1000 cm^{-1}) and a wide uniform gain contour necessary for realization of high sensitivity of the IC-method. The first IC-spectrometer based on a LiF: F_2^+ -color center laser with a ruby laser as a pumping one was put up in operation by E.A. Sviridenkov with co-authors²; a continuous lasing duration of 1 µs was attained, which corresponded

to a threshold absorption sensitivity of $8 \cdot 10^{-5}$ cm⁻¹ in a 10100–11600 cm⁻¹ spectral range. At IAO SB RAS, an IC-spectrometer based on a LiF:F₂⁺-color center laser has been designed. For pumping, a ruby laser was used as well, but lasing duration attained 80 µs that provided for a threshold absorption sensitivity of 10^{-7} cm⁻¹ within the same spectral range.³ The use of xenon laser as a pumping one allowed designing a LiF:F₂⁺-color center spectrometer with lasing duration of 0.2 µs that restricted the threshold absorption sensitivity to 10^{-5} cm⁻¹ in the range 10430– 11200 cm⁻¹ (Ref. 4).

Lamp pumping of crystal color-center lasers makes it possible to obtain pulses with a smooth time profile and tens and even hundreds of microseconds in duration that opens up new prospects in design of IC-spectrometers. The lamp pumped LiF: F_2^+ -color center laser was first described in Ref. 5. Six ING12-7/120 lamps pumped active elements with pumping energy up to 1.5 kJ at a 60-µs light pulse. Maximal energy of the lasing pulse did not exceed 10 mJ at a lasing duration of 12 µs in the tuning range 9700–11200 cm⁻¹.

Transformation of the pumping lamp spectrum with a fluorescent transformer was reported in Ref. 6 as an effective method to improve laser characteristics. We applied this method in our work to design the IC-spectrometer.

Block-diagram of the spectrometer and its main elements are shown in Fig. 1.



Fig. 1. Block-diagram of the experimental setup: cavity mirrors (1 and 5), active element (2), flash-lamp (3), cell (4), MDR-23 diffraction monochromator (6), CCD photodetector (7), and PC (8).

The laser cavity is formed by two spherical mirrors with reflection indices of 99.5 and 92%. A cell of 35 cm length is placed into the cavity to fill in the investigated gases. An active element of $4.5 \times 4.5 \times 90$ mm in size made of LiF crystal is placed in a standard laser head of quantron type. Optical pumping is fulfilled by the luminescence of Rhodamine 6G solution in ethyl alcohol excited by INP5/90A lamp radiation.

Rhodamine 6G solution in ethanol is the most effective transformer for exciting F_2^+ -centers. Its luminescence bands coincide with F_2^+ -center absorption spectra. Besides, the solution cools the crystal and cuts the hard UV-radiation of the lamp, thereby protecting the crystal from thermochemical degradation.

A MDR-23 monochromator with a grating of 1200 grooves/mm records absorption spectra; it provides for inverse linear dispersion (in the 1st order) of about 10 Å/mm. An additional high-quality lens from the tool set for the CIN-1 interferometric table is installed at the monochromator output, which allows a manifold augmentation of dispersion and resolution. In the lens focal plane, Sony 1LX511 linear CCD array with 2048 cells and a length of 28 mm is mounted. The photodetector operation is described in Ref. 7 in more detail.

As was pointed out above, the effective length of the absorption layer $L_{\rm eff}$ is directly proportional to the lasing duration t. A steady-mode cavity lasing (free of mode jumps) during all the time t is supposed. In fact, when detecting temporal behavior of radiation intensity throughout a spectral range, a lot of modes are generated concurrently, hence, if to take the duration of the experimentally detected lasing pulse as a steady-mode lasing time, then the overestimated $L_{\rm eff}$ and, consequently, absorption sensitivity of the spectrometer is obtained.

In this work, to determine $L_{\rm eff}$, an optical slit and a FEU-28 (photomultiplier) were mounted one after another at the monochromator output instead of the diode array; the multiplier signal was recorded by a S9-8 digital oscillograph with a time resolution of 50 ns. This time was much less than the transient time at possible jumps of lasing modes. With the help of the optical slip, a very narrow frequency band (< 0.1 cm⁻¹) was isolated from the whole spectrum (100–150 cm⁻¹). The temporal behavior of intensity allowed a conclusion about the lasing character.

As is seen from Fig. 2, the laser generates at steady modes, i.e., without frequency jumps, hence, L_{eff} is completely determined by t.

In the 10450–10580 cm⁻¹ region, the lasing pulse duration makes 15–20 µs depending on the excess of the pumping energy threshold. Such duration provides for the 1000 m effective length of the absorption layer in the cell at the absorbing substance space factor $L_a/L_c = 0.3$, which corresponds to a threshold absorption sensitivity of $5 \cdot 10^{-6}$ cm⁻¹. The laser is able to operate with a pulse repetition rate up to 3 Hz, which permits the pulse accumulation mode to be realized.



Fig. 2. Temporal behavior of the laser radiation intensity. The time resolution is equal to 50 ns, spectral resolution is 0.1 cm^{-1} .

The absorption spectrum of the water vaporcontaining atmospheric air was registered by this spectrometer at a pressure of 4.5 Torr (Fig. 3).



Fig. 3. Absorption spectra of atmospheric air in the $10480-10520 \text{ cm}^{-1}$ region: calculated with the use of the information system from Ref. 8 (*a*) and detected in this work (*b*).

Comparison of the recorded spectrum and the spectrum calculated with the use of information system "Spectroscopy of Atmospheric Gases" (Ref. 8) shows that the spectrometer threshold sensitivity $(5 \cdot 10^{-7} \text{ cm}^{-1})$ corresponds to the spectrometer sensitivity obtained with the help of Eq. (1).

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