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EFFECT OF THE FREQUENCY TUNING RATE OF A DIODE LASER ON THE SHAPE OF AN ABSORPTION LINE PROFILE DURING MEASUREMENT OF LOW CONCENTRATIONS IN AN EXTERNAL CAVITY WITH OFF-AXIS RADIATION INPUT

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The asymmetry of the detected profiles of water vapor absorption lines in a quartz tube at room temperature and pressures of 0.03–1.00 Torr was studied. The measurements were made by the method of diode laser spectroscopy with an external optical cavity. An off-axis alignment of the cavity was used for quality recording of narrow absorption line profiles. The measurements were made with different rates, 0.1–0.8 cm^{-1}/ms, of tuning of the laser frequency with mirrors that have reflectivities of 99 and 99.98% and different directions of the laser frequency tuning. The asymmetry of the lines arises because of the time delay of the radiation inside the cavity. The results of modeling the measurements taking the filling of the cavity with laser light and its decay over time into account at the cavity output are in good agreement with experimental data.

Keywords: *laser spectroscopy, frequency tuning, optical cavity, absorption spectroscopy, water concentration measurement.*

Introduction. Sensitive measurements of the concentrations of gases based on laser spectroscopy are currently used in climatology [1], geology [2], medicine [3], nuclear power [4], and other areas of science and technology. Laser diode spectroscopy with an external multipass cell (cavity) occupies a special place. The advantages of this method include its high sensitivity and rate of determining concentrations, its noninvasive nature, and simplicity of adjustment in a compact configuration. The varieties include frequency-modulation spectroscopy [5], cavity ringdown spectroscopy (CRDS) [6], integrated cavity output spectroscopy (ICOS) [7], etc. ICOS makes it possible to combine narrow band cw lasers with external optical cavities in simple and effective configurations. An absorption signal is obtained by time integration of the laser light passing through the cavity. The concentration of the absorbent is determined from the output signal, which is a function of the reflections in the mirrors and the scattering and absorption losses of the radiation inside the cavity. A modification of this method with introduction of light into the cavity with a shift relative to its axis (off axis OA-ICOS) makes it possible to greatly increase the density of cavity modes and to obtain envelopes of the absorption lines with much higher resolution than in ordinary ICOS, which is particularly important for measurements in low-pressure media [8, 9].

As part of studies of the components of a glow discharge plasma [10–13], a number of measurements have been made of low densities of the isotopomers of water in a discharge. The measurements were made by a modified OA-ICOS method. In [10, 11] an asymmetry was observed in the detected absorption lines. In this paper the effect is systematically investigated experimentally for water absorption lines in the 7180–7182.5 cm^{-1} range. This phenomenon is analyzed by modeling under the measurement conditions.

Experiment. Figure 1 is a sketch of the apparatus. A cavity of length 45 cm with mirrors at the end of a water-cooled quartz tube with an inner diameter of 20 mm is used in the OA-ICOS method. The water vapor pressure in the cavity was monitored with a sensor (Thyracont VSM77DL) in a filling and pumpout system attached to the resonator through the valves I and O and was kept within limits of 0.1–1 Torr. The tube was pumped with a vacuum station (Pfeiffer TDS-022)

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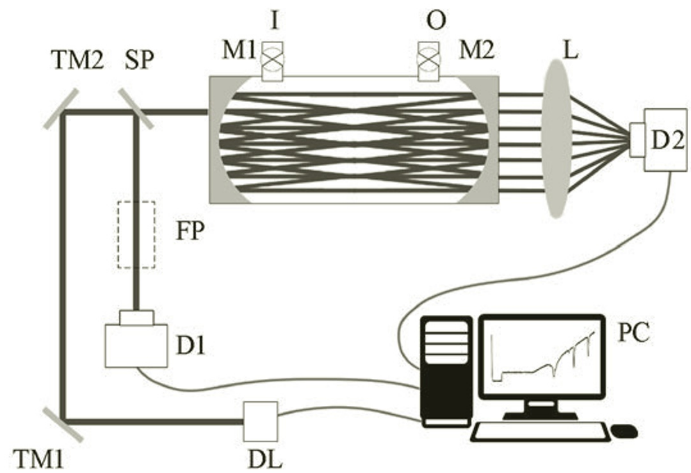

Fig. 1. A sketch of the experimental apparatus.

mounted in the filling and pumpout system to a base pressure of $5 \cdot 10^{-5}$ Torr. Two identical pairs of mirrors M1, M2 for the cavity with reflectivities of 99.98 and 99% and radii of 100 cm were used.

Light from a diode laser DL (Eblana Photonics DM-1392) was introduced into the cavity off axis using turning mirrors TM1 and TM2 and split by a beam splitter plate SP into two beams, one of which went into the cavity while the other was directed to the detector D1. The laser was tuned over the 7180–7182.4 cm^{-1} range, had a linewidth of 2 MHz (~10^{-4} cm^{-1}), and was thermally stabilized at a temperature of 303 K. The signal from detector D1 made it possible to account for atmospheric absorption of the radiation and to subtract it from the analytic signal. A Fabry–Perot (FP) etalon (quartz) was inserted in the channel to account for nonlinearity in the rate of change of the laser frequency during the linear change in the injection current and for frequency calibration, since the degree of nonlinearity varies for different tuning rates and a corresponding calibration was carried out when the spectra were calculated. The spectrum of the etalon was used to transform the spectra from a time to a frequency representation. After the cavity the radiation was collected by a lens L onto detector D2. Control of the laser, and collection and processing of the data were carried out by a computer PC with a universal input-output circuit board (National Instruments PCI-6120). To control the apparatus and collect the data a special program was developed in the LabVIEW medium. The source was a diode laser with a frequency tuning range of ≈2.4 cm^{-1} for the specified injection current. Tuning was done with current pulses. The duration of the current pulse was 5 ms for a digitization rate of 800 kHz; the current segment for tuning the frequency was 60% of the overall duration of the pulse, which corresponds to a tuning rate of 0.8cm^{-1}/ms. When an analog to digital converter (ADC) was used, the tuning current signal consists of a sequential graduated set of values at certain time intervals (1.25 μs for a digitization rate of 800 kHz). The spectral resolution of the system can be estimated as follows: the 2500 points in the spectrum correspond to a segment of ≈2.4 cm^{-1}, so the distance between neighboring points in the spectrum is ~10^{-3} cm^{-1}. Here the Doppler absorption profile covered ~100 points. The spectra were averaged over 2500 cycles of the injection current.

Results and Discussion. To clarify the character of the asymmetry, several experiments were done with different pairs of mirrors at 99.98 and 99%, which influence the damping time for the radiation in the cavity. For the more highly reflecting mirrors the decay time constant $\tau_0(\nu, R)$ (where ν is the laser frequency and R is the reflectivity of the mirrors) was measured experimentally. The calculations used the reflection coefficient derived from the damping time rather than from the conflicting manufacturer's data (>99.98%). For the 99% mirrors the damping time $\tau_0(\nu, R)$ was estimated to be 0.17 μs and not verified experimentally. Measurements with the 99% mirrors did not manifest any noticeable distortions in the line profile. The reasons for the absence of distortions in this type of reflection coefficient are discussed below.

Changing the sampling rate for the digital control system from 800 to 100 kHz in the time representation signifies a change in the duration of the unit current pulse from 1.25 to 10 μs; the duration of the total pump current pulse then increases proportionally, from 5 to 40 ms, which corresponds to tuning rates of 0.8 and 0.1 cm^{-1}/ms. Figure 2 shows absorption profiles that were obtained. It is clear that when the tuning rate is reduced the asymmetry decreases substantially. Measurements were also made with a change in the direction of the frequency tuning from mirrors with a reflectivity of 99.98% at a tuning rate of 0.8 cm^{-1}/ms. As the pump current is raised the laser frequency decreases, and as the current is

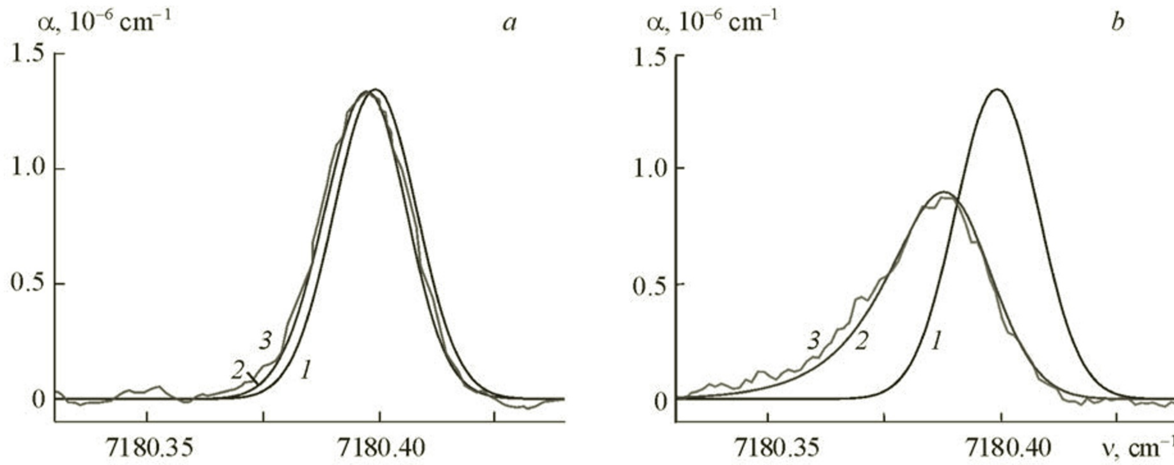

Fig. 2. Results of the experiment and calculations for laser tuning rates of 0.1 cm^{-1}/ms (a) and 0.8 cm^{-1}/ms (b): (1) Doppler profile, (2) theory, (3) experiment, mirrors 99.98%, p = 0.035 Torr; wave number changes in the direction of reduction.

lowered, it rises. With a change in the tuning direction, the shape of the asymmetry in time is retained, while on going from time to frequency it undergoes mirror reflection.

A mathematical model was developed to interpret the processes taking place in the system and leading to distortion of the line shape. Modeling and analysis of a nonstationary system for processing signals of arbitrary form, of which an optical cavity is an example, are based in a dynamic representation on expansion of the signals in terms of unit monochromatic pulses of the simplest shape. For a stepwise variation in the injection current the laser signal can be represented as a sequence of narrow rectangular monochromatic pulses of equal duration Δt with amplitudes determined by the magnitude of the signal.

In general the signal at the output of the cavity is determined by its response to a rectangular monochromatic pulse,

$$I_{\text{out}}(t, \Delta t) = C_p \sum_{i=0}^{t/\Delta t} I_{\text{in}}(t_i')h(t, t_i', \Delta t, R), \quad t_i' = i\Delta t \,, \tag{1}$$

where C_p is the parameter for introduction of radiation into the cavity; t is the time at which the measurement is made; Δt is the pulse duration; $h(t, t', \Delta t, R)$ is the response of the system to a unit rectangular pulse (a nonstationary transition function) at time t'; $I_{\text{in}}(t')$ is the intensity of the incident radiation at time t'; and $I_{\text{out}}(t, \Delta t)$ is the intensity of the radiation at the system output at time t.

The quantity R_{eff} accounts for the losses in the cavity owing to the transmission of the mirrors but also associated with absorption by the medium inside the cavity:

$$R_{\text{eff}}(t, R) = R(1 - a(t)) \,, \tag{2}$$

where $a(t)$ is the absorption loss during a single passage through the cavity with $a(t) << 1$ and R is the reflectivity of the mirrors.

The laser used in the experiment has a finite coherence length $l_c = c/\Delta\nu \approx 150$ m (c is the speed of light and $\Delta\nu$ is the line width); the effective optical path length in the cavity with mirrors that have reflectivity >99.98% is estimated from the damping time constant at 3.2 km, while the spectra are averaged over 2500 cycles so that interference can be neglected and the total intensities at the cavity output can be calculated directly.

In the case of off-axis radiation input the cavity has a response function to a rectangular pulse of monochromatic radiation that is given by expressions derived from the formulas of [9]:

$$h(t, t', \Delta t, R) \text{ before the beginning of the pulse,} \tag{3}$$

$$h(t, t', \Delta t, R) = \frac{(1 - \exp^{-(t-t')/\tau_0(t',R)})T^2}{2(1 - R_{\text{eff}}(t', R))} \text{ during the pulse,} \tag{4}$$

$$h(t, t', \Delta t, R) = \frac{(1 - \exp^{-\Delta t/\tau_0(t',R)})T^2}{2(1 - R_{\text{eff}}(t', R))} \exp^{-(t-t'-\Delta t)/\tau_0(t',R)} \text{ after the end of the pulse,} \tag{5}$$

where t' is the onset time of the pulse; $\tau_0(t', R)$ is the damping time constant in the cavity; and $T = 1 - R$ is the transmission of the mirrors neglecting losses.

In general, the damping time constant of the cavity depends, not only on the reflectivity of the mirrors, but also on the absorption of the medium in the cavity:

$$\tau_0(t', R) = \frac{l}{c((1 - R) + \alpha(t')l)}, \tag{6}$$

where $\alpha(t)$ is the absorption coefficient and l is the distance between the cavity mirrors.

The resulting signal at the output of the cavity is a sum (1), the components of which are given by Eqs. (2)–(6). The shape of the absorption spectrum line profile $\alpha(t)$ is determined by Doppler broadening,

$$\alpha(t) = \alpha_0 \exp\left[-4 \ln 2 \left(\frac{t - t_0}{\Delta\nu}\right)^2\right], \tag{7}$$

where α_0 is the absorption index at the line center, $\Delta\nu$ is the half width of the absorption line, and t_0 is the coordinate of the line center.

The contribution to the signal intensity at a given time on the frequencies at which the source emitted during the preceding moments of time is determined by Eqs. (1), (3), (4), and (5).

The absorption index for $\alpha(t)/l << 1$ is

$$\alpha_{\text{out}}(t) = \frac{I_{0\text{out}}(t) - I_{\text{out}}(t)}{I_{\text{out}}(t)} \frac{1 - R}{Rl}, \tag{8}$$

where $I_{0\text{out}}(t)$ is the intensity at the output from the empty cavity. Equation (8) is derived from Eq. (6) of [9].

The results of the experiment and calculated distortion of the line for different tuning rates (durations of the unit pulse) are shown in Fig. 2.

Thus, several factors affect the asymmetry of the absorption line. First of all, the asymmetry of the line is related to the fact that filling the cavity with radiation and the damping of the radiation at its output are not instantaneous. For certain durations of the unit tuning pulse and gas pressures (line widths), this time delay can have a significant influence on the shape of the absorption line. In addition, increasing the duration of the unit laser pump current pulse for a fixed photon lifetime in the cavity reduces the asymmetry. If the pulse duration is reduced to the level of the minimum possible in our system (1.25 μs) and the photon lifetime in the cavity is changed by replacing the mirrors, then for a shorter photon lifetime the distortion of the line shape will be smaller. Here the change in the direction of the frequency tuning is a mirror reflection of the asymmetry in the frequency representation. This is related to the fact that when the direction of the tuning is changed in the time representation the asymmetry remains the same, but on going from times to frequencies the calibration curves turn out to be mirror symmetric.

Conclusions. The asymmetry of the absorption lines in spectra recorded in a high-Q cavity with rapid tuning of the laser frequency has been studied experimentally and theoretically. It has been shown that the asymmetry is independent of the direction of the frequency tuning, but it depends on the cavity Q and the rate of frequency tuning, and agrees well with mathematical modeling. The asymmetry of an absorption line with off-axis feed of radiation into a high-Q cavity is related to the fact that tuning the frequency requires time to fill and empty the cavity with radiation at a given frequency. Because of this, during recording of the profile of an absorption line there is a superposition of the instantaneous intensity and the undamped intensities of previous frequencies. Thus, increasing both the tuning rate and the Q-factor of the cavity above a certain limit (0.1 cm^{-1}/ms with 99.98% mirrors) cause a significant distortion of the absorption lineshape.

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