USE OF PULSED DIODE LASERS IN LINE SHIFT MEASUREMENTS

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Spectrometer based on a pulsed diode lasers, operating in $1.8-2.4 \mu m$ region, is described. Automatic control of measurements and their processing has been executed with CAMAC and IBM PC/AT. A method based on summation of pulses which differ by the frequency change rate is proposed.

At present great interest is attracted to investigations of the line shift by pressure. It is attributed to the fact that in many applied problems, such as sounding, laser radiation propagation, and others, the neglect of the shift results in considerable errors.¹ Besides, the shift coefficients give new information about molecular interactions and can be used for determination of the intermolecular potential constants.²

Shift coefficients for main atmospheric gases are within $(0.001-0.015) \text{ cm}^{-1}/\text{atm}$ range. As a result, highly sophisticated spectral equipment is needed to measure shifts. Therefore, the development of such an equipment or its improvement with the use of modern computers is very important.

This paper describes a diode spectrometer of high resolution with automated system of data acquisition and processing and presents some results of the spectrometer evaluation at measurement of water vapor absorption line shift by nitrogen pressure.

Diode lasers on the basis of GAInAsSb are used in the spectrometer as radiation sources, generating in the wavelength range from 1.8 to 2.4 μ m and operating in a repetitively pulsed mode with the pulse repetition frequency up to 100 Hz and pulse duration of 1500 μ s, the range of continuous frequency tuning of a single mode is (0.3–1) cm⁻¹. Instantaneous radiation spectrum width of these lasers, measured earlier in Ref. 3, is less than 60 MHz. Figure 1 shows the typical shapes of the time sweep of an intensity and radiation spectrum.

Tuning of a laser emission wavelength in a more wide range has been carried out by temperature variation of a laser crystal. For this purpose the laser was mounted on a Peltjier microrefrigerator, which, in its turn, was installed on a cool-pipeline of a nitrogen cryostat. Such a structure allowed us to change quickly the laser crystal temperature in the range (80–300) K and tune the laser radiation frequency within 400 cm⁻¹ wide interval

For a stabilization of the laser crystal temperature at a given point, the scheme described in Ref. 4 was used, which provided the stabilization accurate to 0.01 K. As is shown below, when a technique of pulse accumulation is used, such an accuracy is quite sufficient for spectroscopic measurements.

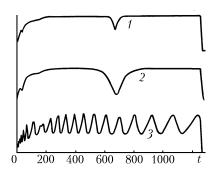


FIG. 1. Simultaneous record of the transmission function of the control cell with H_2O vapor (1), cell with water vapor broadened by nitrogen (2), and a Fabry-Perot interferometer (3). The interval of free dispersion of the interferometer is 0.00819 cm⁻¹.

The laser radiation wavelength measurements were conducted using a monochromator MDG-23 with the accuracy up to 1 cm^{-1} . For more accurate relative measurements and also in order to refer the laser wavelength to the reference absorption lines, Fabry-Perot interferometers were used, which provide the accuracy better than 0.001 cm⁻¹.

Single-pass (reference) and multipass (measuring) cells allowed one to model gaseous media under study at room temperature. The pressure in cells was measured with the mercury and oil manometers.

This spectrometer enabled simultaneous recording of all necessary spectroscopic information during one laser pulse. As our experience shows, a three-channel recording allows simultaneous measurements of the transmission function of a Fabry-Perot etalon, as well as signals before and after a cell with the gas under study, what makes it possible to obtain all parameters of the absorption line profile in one pulse.

Signals recorded with the spectrometer were entered in the automated system of data acquisition and processing. System was developed using electronic blocks in the CAMAC standard and an IBM PC/AT computer. Software for the recording system and data processing makes possible the simultaneous recording, in three channels, of necessary number of pulses, their averaging, restoration of the spectrometer frequency scale, and determination of the parameters of isolate and overlapping absorption line profiles with the use of different theoretical models for profile.

Analog-to-digital conversion of signals was done with the use of three ADC-10/1, which are controlled by crate controller from a computer. The maximum speed of ADC is 1 μ s, a word size is 10 bit, a capacity of buffer memory is 4 k. The read time of a digital code of signals from one pulse of 1500 μ s duration from the buffer memory of three ADCs into the computer memory is 60 ms. After the record of the necessary number of pulses, their processing has been conducted. Restoration of the frequency scale is one of the most important procedure in the signal processing, which determines the measurement accuracy a spectral line center and its shift.

Functions $f(\sqrt{x})$, f(1/x), $f(\ln(x))$, $f(e^x)$, $f(e^{x^2})$ were used for determination of frequency change law v (cm⁻¹) during a pulse. The best results of the frequency scale restoration were obtained with the function

$$v(cm^{-1}) = -[A/(x-B)] + C, \qquad (1)$$

where A, B, and C are constants, determined by a least square fit. The table presents the deviations of center position of interferometer transmission maxima from the theoretical dependence with the use of different frequency scale models. It can be expected that the accuracy of spectral line center determination will be greater than the deviation of the interferometer maximum centers from the theoretical curve, since the deviations in the table include inaccuracies of the scale in addition to the inaccuracy in interferometer maximum center determination due to noise.

The use of a computer in the spectrometer allows one to make accumulation and summation of pulses to increase the signal-to-noise ratio. In this paper in the pulse summation not only their relative displacement is taken into account, as it was done in Ref. 5, but also the pulse-to-pulse instability of the frequency change rate, which we have revealed in our study (see Fig. 2). This fact results in the necessity for pulse summation according to a nonlinear law.

As it was already said, a nonlinear behavior of the frequency is subject to the hyperbolic law most accurate. Therefore, for determination of the function (1) of two successive pulses it is sufficient to determine the coordinates of three corresponding transmission maxima of the interferometer in both pulses. In Fig. 2 x_1, x_2, x_3 are coordinates of three maxima of the first pulse, y_1 , y_2 , y_3 are coordinates of maxima of the second pulse. When we determine constants A, B, and C, we obtain the one-to-one correspondence of pulse points I and I^{T} . In the summation it is necessary to take into account the step-wise behavior (discreteness) of the pulse digitization, i.e., if the coordinate y of the second pulse, which is between integer readouts m and m+1, so that $m \le y < m+1$, conforms to an integer readout (coordinate) n of the first pulse, the result of summation of corresponding pulse points will be obtained by the following formula:

$$I_{\Sigma}^{n} = I_{n} + I_{m}^{T} + (I_{m+1}^{T} - I_{m}^{T}) (y - m),$$
(2)

where y = -a/(n - B) + c. With the help of the same function the corresponding signals of the rest two channels are summed.

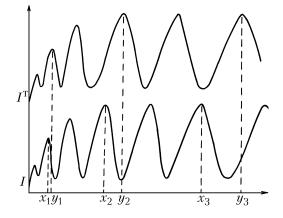


FIG. 2. The scheme of summing laser pulses having different rates of the frequency variation.

No.	$x^{1/2}$	$Exp(x^2)$	Ln(x)	Exp(x)	1/x
1	6.6·10 ⁻³	$2.6 \cdot 10^{-2}$	$4.8 \cdot 10^{-2}$	$-3.5 \cdot 10^{-3}$	$6.9 \cdot 10^{-4}$
2	$-1.3 \cdot 10^{-2}$	$-1.5 \cdot 10^{-3}$	$1.0 \cdot 10^{-2}$	$-7.5 \cdot 10^{-4}$	$-5.4 \cdot 10^{-4}$
3	$-4.9 \cdot 10^{-3}$	$-1.9 \cdot 10^{-2}$	$-1.1 \cdot 10^{-2}$	$1.9 \cdot 10^{-3}$	$-2.4 \cdot 10^{-4}$
4	$1.0.10^{-2}$	$-2.3 \cdot 10^{-2}$	$-2.2 \cdot 10^{-2}$	$2.6 \cdot 10^{-3}$	$-4.4 \cdot 10^{-4}$
5	$1.2 \cdot 10^{-2}$	$-1.0 \cdot 10^{-2}$	$-2.6 \cdot 10^{-2}$	$2.3 \cdot 10^{-3}$	$-4.3 \cdot 10^{-4}$
6	$-1.1 \cdot 10^{-2}$	$1.9 \cdot 10^{-2}$	$-2.5 \cdot 10^{-2}$	$1.7 \cdot 10^{-3}$	$8.8 \cdot 10^{-5}$
7	$1.8 \cdot 10^{-3}$	$6.4 \cdot 10^{-2}$	$-1.9 \cdot 10^{-2}$	$3.8 \cdot 10^{-4}$	$5.1 \cdot 10^{-4}$
8	$1.4 \cdot 10^{-2}$	$-1.6 \cdot 10^{-2}$	$-1.0 \cdot 10^{-2}$	$-3.3 \cdot 10^{-4}$	$1.5 \cdot 10^{-3}$
9	$6.8 \cdot 10^{-3}$	$-7.4 \cdot 10^{-3}$	$-1.7 \cdot 10^{-3}$	$-2.6 \cdot 10^{-3}$	$6.0 \cdot 10^{-4}$
10	$2.3 \cdot 10^{-3}$	$2.8 \cdot 10^{-2}$	$6.8 \cdot 10^{-3}$	$-5.0 \cdot 10^{-3}$	$-1.8 \cdot 10^{-3}$
11	$-2.5 \cdot 10^{-2}$	$1.5 \cdot 10^{-2}$	$1.8 \cdot 10^{-2}$	$-2.1 \cdot 10^{-3}$	$-1.1 \cdot 10^{-3}$
12	$1.3 \cdot 10^{-2}$	$-2.4 \cdot 10^{-2}$	$3.1 \cdot 10^{-2}$	$5.5 \cdot 10^{-3}$	$1.2 \cdot 10^{-3}$

TABLE I. Results of restoration with different models of the frequency scale.

With the help of the above-described spectrometer some test measurements of water vapor line center shift at $v = 5521.9039 \text{ cm}^{-1}$ by the nitrogen pressure were carried out. To do this, the multipass cell has been filled with the mixture of water vapor and nitrogen, while the reference cell was filled with only pure water vapor. Then we simultaneously recorded (with the accumulation of 20 pulses) interferometer transmission maxima and absorption line profiles of pure H₂O and that broadened by nitrogen vapors (see Fig. 1). Then the shift with respect to water vapor line center in the reference cell was determined with a computer.

We have obtained the shift value of 0.0089 ± 0.0004 cm⁻¹, which well coincides with $\delta v = 0.0092\pm0.0008$ cm⁻¹ measured with a Fourier-spectrometer in Ref. 6.

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