

# Role of noise in the diode-laser spectroscopy of the spectral line profile

A I Nadezhdinskiĭ, V V Plotnichenko, Ya Ya Ponurovskiĭ, M V Spiridonov

**Abstract.** Questions concerning precise measurements of the spectral-line-profile parameters by diode-laser spectroscopic methods were examined. The instrumental function of a distributed-feedback diode laser ( $\lambda = 1.53 \mu\text{m}$ ), consisting of the additive contributions of the noise due to spontaneous emission, frequency fluctuations, and intensity fluctuations, was investigated. An analytical formula was obtained for the spectrum of the diode-laser field formed by frequency fluctuations. The spectral density  $g_0$  of the frequency fluctuations, determining the width of the central part of the emission line profile of a diode laser, was found by two independent methods (by fitting to a Doppler-broadened absorption line profile and by finding the intensity of the residual radiation and the saturated-absorption line width). The parameters  $\Omega$  and  $\Gamma$  of the spectral density of the frequency fluctuations, coupled to the relaxation oscillations and determining the wing of the diode-laser emission line profile, were determined experimentally. By taking into account the instrumental function of the diode laser, involving successive convolution with the recorded emission spectra, it was possible to reproduce correctly the spectral line profile and to solve accurately the problem of the 'optical zero'. The role of the correlation between the intensity noise and the diode-laser frequency was considered.

## 1. Introduction

The progress in the development of the IR-spectroscopic techniques and particularly in diode-laser spectroscopy, demonstrated by an appreciable increase in the spectral resolution (to within better than  $10^{-5} \text{ cm}^{-1}$ ) and in the precision of the recorded spectra (signal/noise ratio greater than  $10^5$ ), has generated numerous new applications and significantly advanced the traditional applications of IR spectroscopy, especially in the physics of the Earth's atmosphere and of the atmospheres of other planets. By virtue of its high precision, the spectral line profile and especially the line width and shift, determined by the buffer-gas pressure, serve as an excellent analytical criterion in many problems of astrophysics and the physics of the atmosphere.

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The study of the spectral line profile by diode-laser spectroscopic methods makes it possible to observe the fine effects of the intermolecular interaction such as a reduction in the inhomogeneous line broadening (the Dicke effect), the dependence of the impact characteristics on the velocity of the absorbing molecule (the wind effect), the line splitting owing to the collision anisotropy, etc. [1]. Apart from this, it is possible to investigate indirectly, via the line-profile parameters, the characteristics of the diode-laser radiation itself: the intensity and frequency noise, as well as their correlation.

In the presence of the diode-laser radiation noise, the spectral line profile and the associated intermolecular interaction processes must be considered in a self-consistent manner: diode-laser radiation field + active medium + buffer-gas molecules. If the study is restricted to the uncorrelated fluctuations in the frequency and in the intensity of the diode-laser radiation, it is possible to introduce the diode-laser instrumental function (IF). Different components of the IF, associated with spontaneous emission and quantum frequency and intensity fluctuations, lead to different distortions of the profile in its precision treatment.

One of the manifestations of the laser IF in high-resolution molecular spectroscopy is the 'optical zero' problem. This effect is usually attributed to the inhomogeneously broadened diode-laser spectrum when part of the radiation is not absorbed by the spectral line, which leads to the appearance of a residual radiation at the centre of the saturated line. It has been assumed that this parasitic effect may be excluded by filling a cell with a gas at a fairly high pressure, determining the intensity  $\Delta I$  of the transmitted radiation at the centre of the saturated line, and using it as the true zero.

Observation of a dependence of  $\Delta I$  on the experimental parameters (the gas pressure  $p$  and the cell length  $L$ ) has become a factor complicating this approach [2]. In this case, diode lasers for the mid-IR range were used and the effect was attributed to the spontaneous emission by a diode laser. A similar and even more pronounced dependence was observed in a diode laser in the near-IR range [3]. In this case the effect was explained by taking into account two inhomogeneously broadened components of the diode-laser spontaneous emission spectrum and the intensity fluctuations. These factors have stimulated a detailed study of the influence of the laser IF on the spectral line profile, the preliminary results of which have already been published [4].

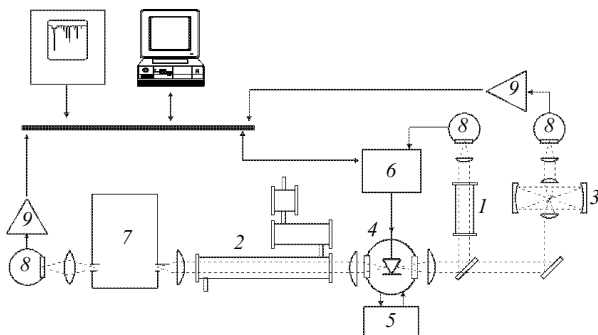
This communication describes a study of the IF of a distributed-feedback laser ( $\lambda = 1.53 \mu\text{m}$ ). Allowance for the laser IF, involving successive convolution with the recorded transmission spectra, makes it possible to reproduce correctly the spectral line profile and to solve correctly the problem of the 'optical zero'.

## 2. Experimental setup

The diode-laser spectrometer (DLS) setup, presented in Fig. 1, has already been described in detail [4]. We shall consider briefly the principal units and parameters of the DLS. The laser generated pulses of 4–10 ms duration at the  $\lambda = 1.53 \mu\text{m}$  wavelength at a repetition frequency of 40 Hz. A temperature-stabilisation system (5), utilising a thermoelectric Peltier cooling unit as the controlling component, ensured a temperature instability at the level of  $10^{-3}$  K in the temperature range from  $-15$  to  $+50$  °C.

The laser radiation, emitted from the opposite faces of the laser crystal, was collimated into two parallel beams and it formed three optical channels. The analytical channel (2) contained a cell with the test gas and an IKS-31 monochromator (7) for the selection of one radiation mode. The two auxiliary channels were designed to stabilise the tuning curve of the laser along the reference-gas absorption line (1) and the frequency calibration line (3). The stabilisation channel (1) consisted of a reference cell 20 cm long with the reference gas and a system for the stabilisation of the laser current along the gas-absorption line (6). The frequency calibration channel (3) consisted of a confocal cavity with the free spectral range  $D^* = 0.00849 \text{ cm}^{-1}$  and a Fabry–Perot interferometer with  $D^* = 0.049272 \text{ cm}^{-1}$ .

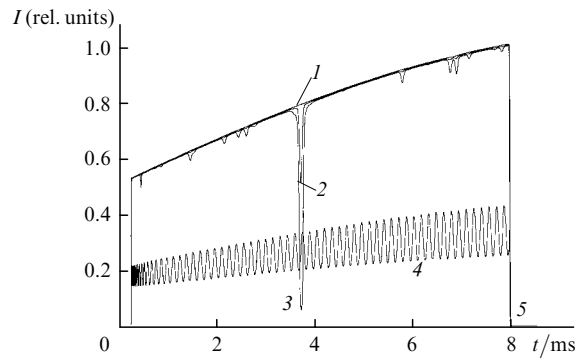
The frequency scale calibration was carried out with the aid of interferometers and the absolute frequency calibration was achieved on the basis of the gas absorption lines in the reference channel (1). After passing through a preamplifier (9), the signals from photodetectors (8) reached the analogue–digital converter (ADC) inputs. The two-stage temperature stabilisation system, (5) and (6), ensured a long-term instability of the diode-laser temperature not worse than  $4 \times 10^{-4}$  K. The circuit board of a 12-bit PCI-MIO-16E-1 ADC from the National Instruments Company was used to record the signal. The ADC sampling frequency was 1 MHz, the sampling length was 8700, and the number of accumulations ranged from 1 to 255. After the accumulation, the ADC ensured a signal/noise ratio better than  $5 \times 10^4$ . The spectra were recorded and processed in the Labview 5.0 system and they were also processed with the aid of the Matlab 5.2 system. The processing procedure has already been described in detail [4].



**Figure 1.** Diode-laser spectrometer for precise measurements of the line profiles: (1) reference channel; (2) analytical channel; (3) frequency-calibration channel; (4) diode laser in a cryostat; (5) system for the thermal stabilisation of a laser with a thermoelectric Peltier cooling unit as the controlling component; (6) thermal stabilisation system based on the reference-gas absorption line; (7) monochromator; (8) photo-detectors; (9) preamplifiers.

Acetylene (purity 99.9%) was used as the test and reference gas. The filling and pressure-measuring system consisted of a backing vacuum pump, a differential manometer (Digitron Instruments) with pressure-measuring ranges of 0–200 and 0–500 mbar, and a barometer (MKS Instruments Inc.) for the precise measurement of the pressure in the range 0–10 mbar.

Fig. 2 presents a typical example of the transmission spectra recorded with the three-channel DLS and needed for the investigation of the gas-absorption line profile.



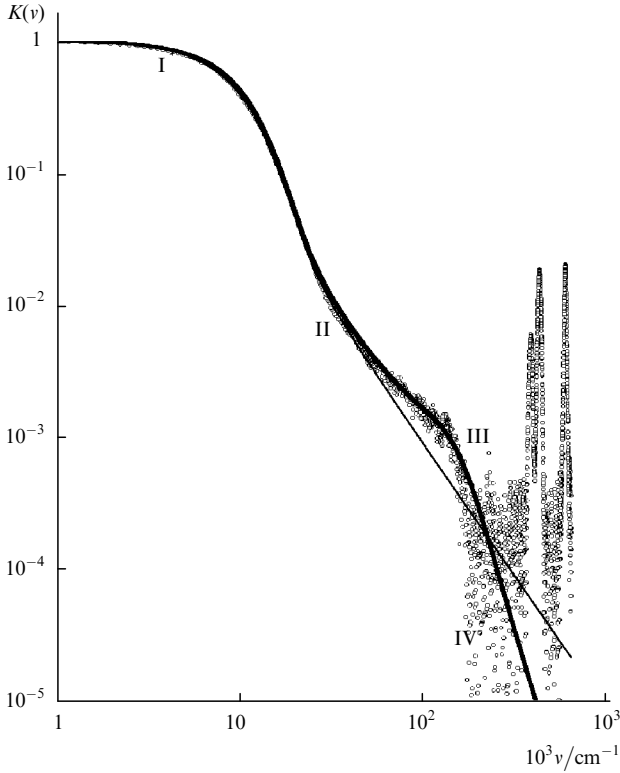
**Figure 2.** Example of recording the spectra (with the aid of a three-channel diode-laser spectrometer) needed in the investigation of the gas-absorption line profile: total transmission line (base line), i.e. the intensity of the radiation transmitted through an empty cell 200 cm long (1), the absorption line of acetylene used as the reference gas (cell length 20 cm, pressure 3 Torr) (2), the acetylene test line [ $R(13)$ ,  $\nu_1 + \nu_3$  band, pure-gas pressure 0.25 Torr, cell length 200 cm] (3), transmission signal of a Fabry–Perot interferometer ( $D^* = 0.049272 \text{ cm}^{-1}$ ) (4), and the total absorption line ('optical zero') (5).

## 3. Diode-laser instrumental function

With the aid of the DLS, we were able to observe clearly the influence of the laser IF, manifested by the distortion of the Doppler-broadened spectral line. Fig. 3 presents a normalised acetylene absorption spectrum [ $R(10)$  line,  $\nu_1 + \nu_3$  band] on the double logarithmic scale, obtained at a pressure of 0.3 Torr in a cell 200 cm long. For clarity, Fig. 3 shows only half the line profile and the line centre corresponds to the zero frequency. The result of fitting the Voigt curve to the line profile is included, as is the curve describing the line profile after correction for the laser IF. The dynamic range of variation of the absorption coefficient exceeded  $4 \times 10^4$  and the spectral resolution was  $10^{-4} \text{ cm}^{-1}$ . Such conditions during recording of the spectrum made it possible to observe the distortions in the Doppler-broadened line profile associated with the manifestation of the laser IF. The line profile (Fig. 3) can be conventionally divided into four regions:

I. The region described by a Gaussian profile with the FWHM  $\omega_G = 15.768 \times 10^{-3} \text{ cm}^{-1}$ . This inhomogeneously broadened part of the line profile is due to an ensemble of velocity-distributed absorbing noninteracting particles. The value of  $\omega_G$  obtained from line fitting corresponds to the theoretical width of the acetylene line calculated for this frequency range ( $\omega_G^{\text{theor}} = 15.821 \times 10^{-3} \text{ cm}^{-1}$ ).

II. The line profile distorted by the laser IF. The result of fitting a Voigt profile to the Doppler-broadened line yields a nonzero Lorentzian component  $\omega_L = 2.871 \times 10^{-3} \text{ cm}^{-1}$ .



**Figure 3.** Normalised absorption spectrum of acetylene [ $R(10)$  line,  $\nu_1 + \nu_3$  band] at a pressure of 0.3 Torr and for a cell 200 cm long. Half the line profile is shown and the line centre corresponds to the zero frequency. The thin line is the result of fitting a Voigt profile to the line profile [integral intensity  $A = (19.919 \pm 0.021) \times 10^{-3} \text{ cm}^{-1}$ ,  $\omega_G = (15.768 \pm 0.036) \times 10^{-3} \text{ cm}^{-1}$ ,  $\omega_L = (2.871 \pm 0.047) \times 10^{-3} \text{ cm}^{-1}$ ]. The thick line is the diode-laser radiation noise and the circles represent the experimental values.

This quantity determines the width of the central Lorentzian part of the diode-laser emission line arising from the frequency fluctuations and introduces a distortion into the spectral line profile in the form of a Lorentzian wing.

III. The region of the line wing modified by the frequency noise of the diode-laser radiation. The sharp decrease in the absorption coefficient in the wing (Fig. 3), where it is inversely proportionally to  $\nu^4$ , occurs in the  $\sim 0.15 \text{ cm}^{-1}$  frequency band. It is then lower by more than four orders of magnitude than the absorption coefficient at the maximum. The line-profile distortions are caused by another power component of the laser IF.

IV. The region of the remote line wing with  $\nu \gg 1 \text{ cm}^{-1}$  (not shown in Fig. 3). In this frequency range the influence of the laser IF on the line profile results from the broadband (band wider than  $100 \text{ cm}^{-1}$ ) continuous emission from the diode laser. The use of a monochromator, truncating the recorded spectral band to  $2.5 \text{ cm}^{-1}$ , almost completely eliminates the spontaneous emission. The peaks of the  $R(10)$  line wing are coupled to the resonant absorption of radiation by weaker hot and combination bands of acetylene.

The spectral line profile (Fig. 3) thus carries virtually the entire information about the radiation-field noise and the line broadening mechanisms. A correct reproduction of the profile therefore requires taking the influence of the laser IF into account.

## 4. Theory

We shall consider in greater detail the role of the emission spectrum of a diode laser in the precise recording of a spectral line. We shall assume that the diode-laser emission spectrum corresponds to the steady state and retains its shape during frequency scanning. Furthermore, the scanning is sufficiently slow to be able to describe correctly the diode-laser spectrum at each instant. By solving the Maxwell equations for the laser radiation-absorbing medium it can be shown that, in the case of noisy radiation, the  $\Delta I(\nu)$  spectrum recorded by a photodetector represents a convolution of the transmission spectrum of the medium  $T(\nu)$  with the laser IF  $S(\nu)$ :

$$\Delta I(\nu) = \int_{-\infty}^{\infty} T(x)S(\nu - x) dx. \quad (1)$$

The quantity  $S(\nu)$  can be expressed in terms of the Fourier integral of the field correlation function  $E$ :

$$S(\nu) = \text{Re} \int_{-\infty}^{\infty} \langle E(t)E^*(t + \Delta t) \rangle \exp(-i\nu\Delta t) d\Delta t. \quad (2)$$

In the presence of uncorrelated noise in the diode-laser radiation, the laser IF can be represented by the sum of several terms:

$$S(\nu) = S_{\text{sp}}(\nu) + S_{\text{qu}}(\nu) + S_E(\nu). \quad (3)$$

where  $S_E(\nu)$  is the spectrum of the diode-laser radiation field arising from the quantum frequency fluctuations;  $S_{\text{sp}}$  and  $S_{\text{qu}}$  are the IF components associated with the spontaneous emission and quantum fluctuations in the radiation intensity. It should be noted that the emission spectrum may be influenced also by other additional sources of noise, for example the pump-current and temperature fluctuations, the flicker noise, the mode-switching noise, etc.

We shall restrict our discussion to the fundamental sources of the diode-laser radiation noise included in expression (3). We shall assume that the sources of noise which are not fundamental may be eliminated by selection of the diode laser employed, by optimising the pump and thermal stabilisation systems, by selecting the diode-laser operating regime, etc. The correlation between the intensity and frequency noise is ignored in expression (3). When account is taken of this correlation, the problem of the line profile becomes significantly more complicated because it is then necessary to consider the open self-consistent ‘diode-laser radiation field – active medium – buffer-gas molecules’ system and the term laser IF is no longer quite correct.

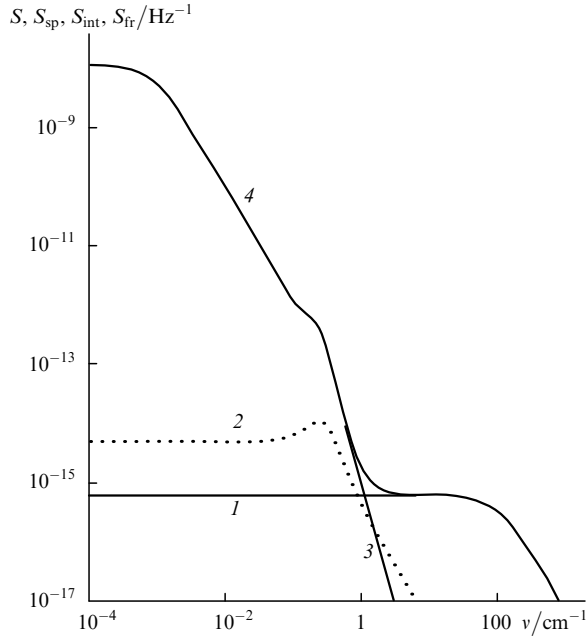
Fig. 4 (curve 4) gives the IF for one of the investigated distributed-feedback diode lasers ( $\lambda = 1.53 \mu\text{m}$ ). The different components of the IF lead to different distortions of the spectral line profile during its precise recording. We shall consider in greater detail the role of each term in expression (3).

### 4.1 Spontaneous emission

The spontaneous emission in the active region of the diode laser leads to the appearance of the noise component of the radiation together with the coherent component:

$$I \propto |E|^2 = E_0^2 + 2E_0 \cdot E_{\text{sp}} + E_{\text{sp}}^2, \quad (4)$$

where  $I$  is the diode-laser radiation field intensity;  $E = E_0 + E_{\text{sp}}$  is the field vector consisting of the sum of the coherent and spontaneous components.



**Figure 4.** Instrumental function for one of the distributed-feedback diode lasers investigated  $S(4)$  and the spectral densities  $S_{sp}(v)$  (1),  $S_{int}(v)$  (2), and  $S_{fr}(v)$  (3).

The spectral density of the relative diode-laser noise due to spontaneous emission  $S_{sp}(v)$  (Fig. 4, curve 1) was determined experimentally. It has the broadest spectrum (more than  $100 \text{ cm}^{-1}$ ). The spectral density  $S_{sp}(v)$  in this band amounts to  $\sim 5 \times 10^{-16} \text{ Hz}^{-1}$ . The spontaneous noise in the diode-laser radiation leads to the problem of the ‘optical zero’: part of the radiation passes through the test gas at the absorbing-gas concentration known to be higher a priori, whereas the entire radiation should be absorbed at the line centre. At the same time, the influence of the ‘optical zero’ caused by the spontaneous emission of the diode laser may be reduced appreciably by employing in the optical system a monochromator which truncates the recorded spectral band.

#### 4.2 Intensity fluctuations

The radiation of a diode laser, which includes the noise resulting from spontaneous emission, includes charge-carrier concentration fluctuations when this radiation is absorbed in the active region of the laser. This is accompanied by correlated quantum fluctuations in the frequency and intensity of the diode-laser radiation. The total spectral density of the intensity fluctuations  $S_{int}(v)$  (Fig. 4, curve 2) consists of two parts:

$$S_{int} = S_{sp} + S_{qu} = S_{sp} + \left( \frac{\partial I}{\partial n} \frac{\partial n}{\partial v} \right)^2 S_{fr}, \quad (5)$$

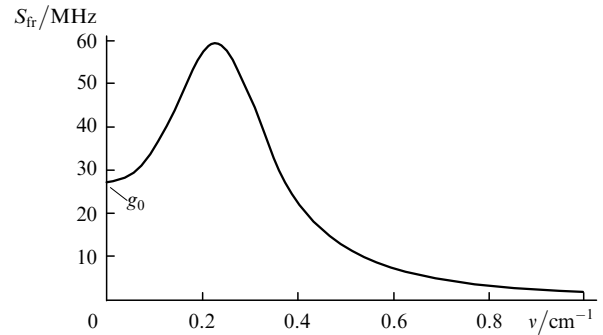
where  $S_{fr}$  is the spectral density of the quantum frequency fluctuations. The correlation of the frequency and intensity noise is taken into account on the right-hand side of the equation. The spectral noise density  $S_{int}(v)$  (Fig. 4, curve 2) has a characteristic maximum in the region  $\nu \approx 0.2 \text{ cm}^{-1}$ . This resonance is associated with the relaxation-oscillation frequency [5]. The maximum in the spectral density of the intensity fluctuations for the investigated laser does not exceed  $\sim 6 \times 10^{-15} \text{ Hz}^{-1}$ . The intensity noise affects the recording of collision-broadened lines.

#### 4.3 Frequency fluctuations

The spectral density of the quantum frequency fluctuations  $S_{fr}(v)$  is determined from the solution of the system of equations describing the quantum fluctuations in the carrier density in the active zone of the diode laser and the amplitudes and phases of the generated radiation field [5]:

$$S_{fr}(v) = g_0 \frac{\Omega^4}{(\Omega^2 - v^2)^2 + v^2 \Gamma^2}. \quad (6)$$

The parameters  $g_0$ ,  $\Omega$ , and  $\Gamma$  are expressed in terms of the average nonlinear susceptibility of the medium and the second moments of the noise operators, which occur in the quantum-mechanical Langevin equation;  $g_0$  is the spectral density in the region of zero frequency;  $S_{fr}(v)$  has a resonance near  $\nu = \Omega$ ;  $\Omega$  is the frequency of the relaxation oscillations. Fig. 5 shows the spectral density of the quantum fluctuations of the diode-laser frequency obtained for one of the investigated lasers.



**Figure 5.** Spectral density of the quantum frequency fluctuations  $S_{fr}(v)$  of one of the investigated lasers.

#### 4.4 Spectrum of the laser-radiation field resulting from the frequency noise

Neglecting the intensity fluctuations, the field correlation function in formula (2), determined by the frequency fluctuations, may be expressed in the form

$$\langle \mathbf{E}(t) \cdot \mathbf{E}^*(t + \Delta t) \rangle = E_0^2 \exp \left[ -\frac{1}{2} \left\langle \left( \int_0^{\Delta t} f(x) dx \right)^2 \right\rangle \right]. \quad (7)$$

The right-hand part of this equality may be expressed in terms of the inverse Fourier transformation of the spectral density of the quantum frequency fluctuations [formula (6)]. Expression (7) can be readily evaluated for the case of small rapid fluctuations when  $g_0 \ll \Omega$ ,  $\Gamma$ . Under these conditions, the spectral field density  $S_{fr}$  is the product of a Lorentzian profile with a FWHM  $g_0$  and the normalised spectrum of the frequency fluctuations:

$$S_{fr}(v) = \frac{g_0}{\pi(v^2 + g_0^2)} \frac{\Omega^4}{(\Omega^2 - v^2)^2 + v^2 \Gamma^2}. \quad (8)$$

Fig. 4 (curve 3) shows the contribution of the frequency fluctuations to the laser IF. The central part of the diode-laser emission line profile is described by a Lorentzian profile with a FWHM  $g_0$ . The spectral density introduced by the Lorentzian component of the IF is  $S_{field} \approx 10^{-8} \text{ Hz}^{-1}$ . The wing of the line profile (a spectral-band region  $\nu \geq 0.2 \text{ cm}^{-1}$ ) is described by the second multiplier in expression (8). The spectral density in this frequency band differs by more than

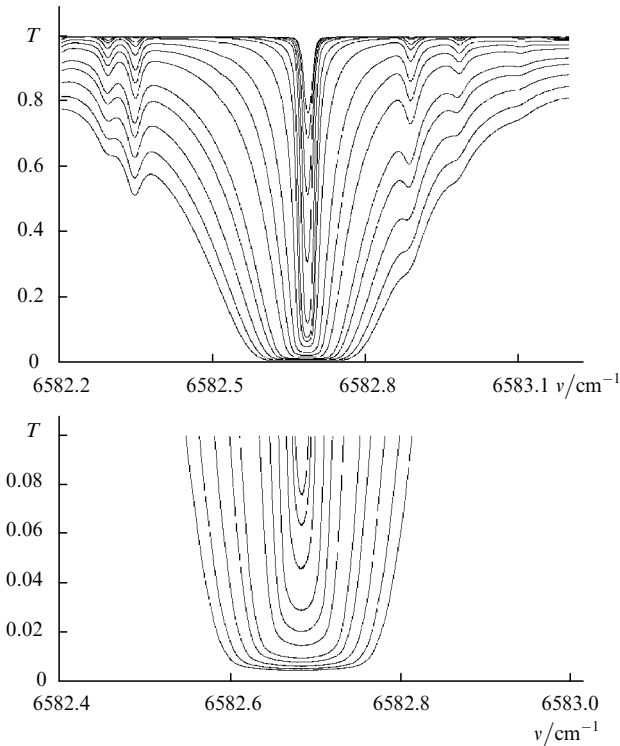
three orders of magnitude from  $S_{\text{field}}$  in the central part. In the interval between the central part and the wing, there is a characteristic peak associated with the frequency  $\Omega$  of the relaxation oscillations. The far wing of the emission line profile ( $\nu > 1 \text{ cm}^{-1}$ , Fig. 4) is due to spontaneous emission. This inhomogeneous component of the emission spectrum is ignored in expression (8) because spontaneous emission may be suppressed maximally with the aid of a monochromator.

Since the laser IF distorts the spectral line profile, a correct reconstruction of the profile must take into account the IF (with the parameters  $g_0$ ,  $\Omega$ , and  $\Gamma$ ) through the convolution described by formula (1). The part of the laser IF associated with  $g_0$  plays the main role in the study of the Doppler-broadened line profiles and of the line profiles with the half-widths  $\omega_{1/2} \ll \Omega$  and  $\Gamma$ . However, if the dynamic range of variation of the absorption coefficient is wide, the power dependence of the laser IF, manifested by a marked distortion of the profile, may also have an effect (Fig. 3). We shall consider the determination of  $g_0$  in greater detail.

## 5. Experiment

### 5.1 Determination of the parameter $g_0$

One of the manifestations of the influence of the diode-laser spectrum is the problem of the ‘optical zero’. Fig. 6 gives the transmission spectra of acetylene at different pressures ( $p \approx 0.02 - 94 \text{ Torr}$ ) in a cell  $L = 200 \text{ cm}$  long. Part of the radiation evidently passes through the test gas even at the centre of the saturated line (see the lower enlarged part of Fig. 6). It has to be noted that the total absorption in Fig. 6 corresponds to zero transmission. The saturated spectral line acts as a narrow spectral filter which fully absorbs the radiation of a diode laser tuned to the centre of this line.



**Figure 6.** Transmission spectra of acetylene in a cell 200 cm long at pressures of 0.02–92 Torr.

$\Delta I$  then characterises the fraction of the radiation in the wings of the diode-laser spectrum transmitted by the saturated spectral line. The possibility of varying the width of this filter  $\Delta\omega$  (the FWHM of the transmission spectrum) by a change in the experimental parameters ( $p$  and  $L$ ) makes it possible to reconstruct the laser IF.

The residual radiation intensity  $\Delta I/I_0$  at the centre of the saturated line, determined by the influence of the laser IF, can be expressed in the form

$$\frac{\Delta I(\nu - \nu_0)}{I_0} = \int_{-\infty}^{\infty} \exp[-K(\nu - \nu_0 - x)pL] S_{\text{field}}(x) dx, \quad (9)$$

where

$$K(\nu - \nu_0) = A f_{\text{prof}}(\nu - \nu_0)$$

is the absorption coefficient;  $I_0$  is the intensity of the incident radiation;  $\nu_0$  is the central line frequency;  $A$  is the integral intensity (line intensity);  $f_{\text{prof}}$  is the line profile. The saturated-line profile at the centre will be assumed to be Lorentzian:

$$f_{\text{prof}} = \frac{1}{\pi} \frac{\omega_{1/2}}{(\nu - \nu_0)^2 + \omega_{1/2}^2},$$

where  $\omega_{1/2}$  is the FWHM. It can be seen from Fig. 6 that the condition  $\Omega, \Gamma \gg \nu \gg g_0, \omega_{1/2}$  holds for lines recorded at high pressures ( $p > 15 - 90 \text{ Torr}$ ). We then have

$$f_{\text{prof}} \approx \frac{1}{\pi} \frac{\omega_{1/2}}{\nu^2}, \quad S_{\text{field}}(\nu) \approx \frac{g_0}{\pi \nu^2}.$$

Under these conditions, expression (9) can be readily evaluated:

$$\frac{\Delta I}{I_0} \approx \frac{g_0}{(\omega_{1/2} A p L)^{1/2}}. \quad (10)$$

The residual-radiation intensity  $\Delta I$  depends linearly on  $g_0$ . The ratio of the residual-radiation intensities for different spectral lines is inversely proportional to the square root of the ratio of their integral intensities:

$$\frac{\Delta I_1}{\Delta I_2} \approx \left( \frac{A_2}{A_1} \right)^{1/2}.$$

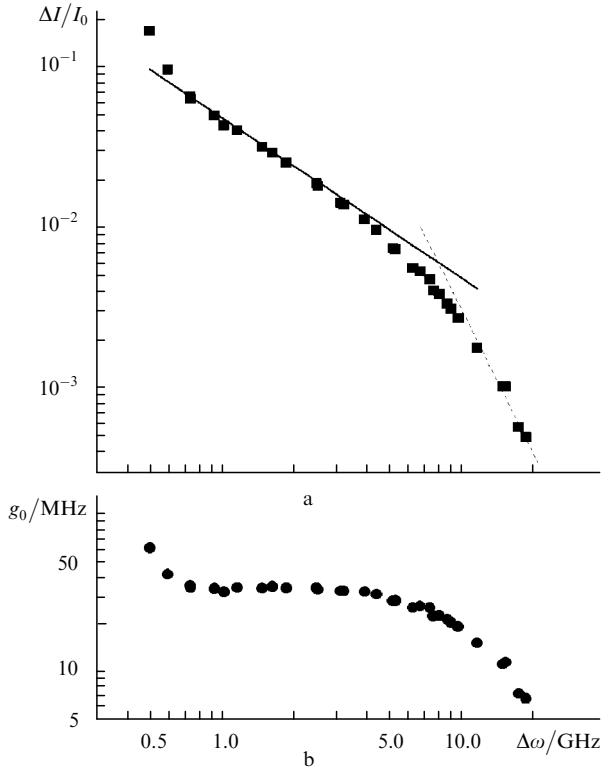
In order to find  $g_0$ , it is sufficient to determine the quantities  $\Delta I/I_0$  and  $\omega_{1/2}$  which occur in expression (10). Since the FWHM of the line is determined from half the signal intensity, one can write

$$\exp\left(-\frac{A p L \omega_{1/2}}{\pi \Delta\omega^2}\right) = \frac{1}{2}.$$

The final expression for  $g_0$  then assumes the form

$$g_0 \approx \frac{\Delta\omega \Delta I}{2 I_0} (\pi \ln 2)^{1/2}. \quad (11)$$

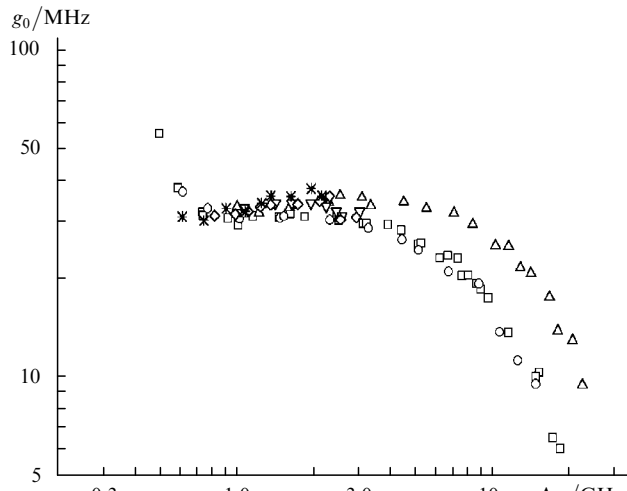
Fig. 7 shows the experimental dependences of the normalised residual-radiation intensity  $\Delta I/I_0$  on  $\Delta\omega$  together with the parameter  $g_0$  calculated from formula (11). In Fig. 7a, it is possible to select regions (fitting straight lines), which are described by different components of the IF. In the frequency range 500–5000 MHz (continuous straight line),  $\Delta I/I_0$  is inversely proportional to  $\Delta\omega$ . The Lorentzian central component of the laser IF with the FWHM  $g_0$  operates here (Fig. 7b). In the frequency range above 5000 MHz (dashed straight line), there is a sharp fall of  $\Delta I/I_0$  (proportional to  $1/\Delta\omega^3$ ). This section of the spectrum is determined by the wing of the laser IF (see region 4 in Fig. 3). The usual



**Figure 7.** Dependences of the normalised residual-radiation intensity  $\Delta I$  on the saturated-line width  $\Delta\omega$  (symbols) and the fitted straight lines (continuous and dashed straight lines describing different components of the laser IF) (a) and the FWHM of the Lorentzian component  $g_0$  of the IF calculated by formula (11) (b).

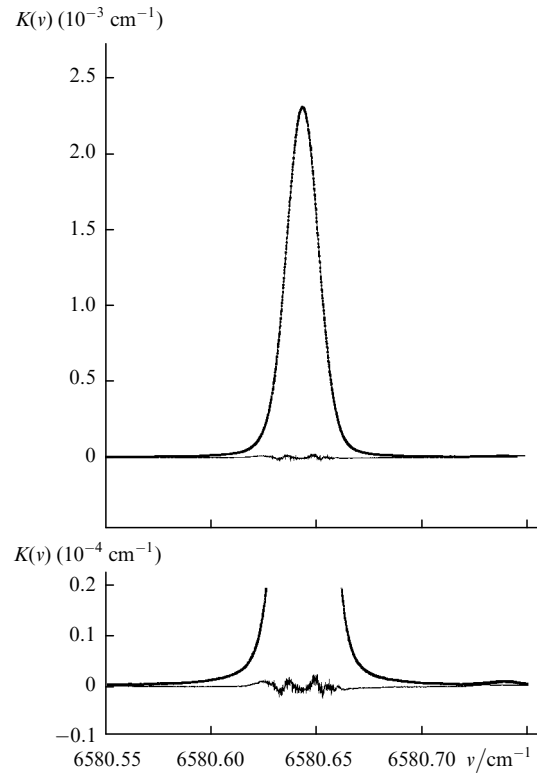
Bouguer law holds below 300 MHz (unsaturated absorption line,  $\Delta I \leq 20\%$  of the total signal).

A series of experiments was performed to determine  $g_0$  for a laser diode operating near  $\lambda = 1.53 \mu\text{m}$ , whereupon the frequency coincides with the intense lines of the  $R$  branch of the  $\nu_1 + \nu_3$  band of acetylene. Fig. 8 presents the values of  $g_0$  for different diode-laser tuning ranges [the  $R(10)$ – $R(15)$  absorption lines of acetylene]. The average value of  $g_0$  in the range  $\Delta\nu = 1000$ – $5000$  MHz is 35 MHz.



**Figure 8.** Dependence of  $g_0$  on  $\Delta\omega$  for different tuning ranges of a diode laser [absorption lines  $R(10)$  ( $\square$ ),  $R(12)$  ( $\Delta$ ),  $R(13)$  ( $\nabla$ ),  $R(14)$  ( $\diamond$ ), and  $R(15)$  ( $*$ ) of the  $\nu_1 + \nu_3$  band of acetylene].

An independent method was employed in order to confirm the validity of this approach to determination of the IF parameter  $g_0$ . It involved fitting of a Voigt profile to a Doppler-broadened line. The Lorentzian component of the profile should represent directly  $2g_0$ . The Gaussian component of the line width should serve as a criterion of the line fitting validity. It is calculated theoretically (for the lines of the  $R$  branch in the  $\nu_1 + \nu_3$  band of acetylene we have  $\Delta\omega_G \approx 0.0158 \text{ cm}^{-1}$ ). Fig. 9 gives the results of fitting a Voigt profile to the  $R(11)$  line of acetylene. The root-mean-square difference between the experimental and model profiles does not exceed 0.3%. Table 1 lists the values of  $g_0$  obtained for different laser-operation regions by independent methods.



**Figure 9.** Measured profile of the  $R(10)$  absorption line of acetylene (circles, cell length 2 m, pure-gas pressure  $p = 0.34$  Torr), the result of fitting a Voigt profile (continuous line) (a), and the difference between the calculated and measured profiles (b). The parameters of the calculation based on the Voigt model:  $\chi^2 = 3.591 \times 10^{-11}$ ,  $\nu_0 = 6580.64386(2) \text{ cm}^{-1}$ ,  $S = (5 \pm 0.2) \times 10^{-5} \text{ cm}^2$ ,  $\Delta\omega_G = (158.4 \pm 0.1) \times 10^{-4} \text{ cm}^{-1}$ ,  $\Delta\omega_L = (35.7 \pm 0.1) \times 10^{-4} \text{ cm}^{-1}$ .

**Table 1.**

Laser operating region (acetylene $R$ line)	$g_0/\text{cm}^{-1}$	
	obtained from line fitting	obtained from $\Delta I/I_0$ and $\Delta\omega$
$R(14)$	0.00239	0.00235
$R(13)$	0.00183	0.00179
$R(11)$	0.00357	0.00364
$R(10)$	0.00287	0.00278

## 6. Conclusions

Allowance for the laser IF, involving successive convolution with the recorded transmission spectra, thus makes it possible to reconstruct correctly the spectral line profile and to solve correctly the problem of the ‘optical zero’.

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