

A THERMOREFRACTIVE PHASE TECHNIQUE FOR ANALYZING GAS COMPOSITION OF THE ATMOSPHERE WITH A TWO-MODE GAS LASER

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A thermorefractive phase technique to measure minor atmospheric gas components is suggested. It was tested experimentally for estimating the ethylene (C₂H₄) content in the air. The effect of certain parameters upon the level of the recorded signal was assessed. The threshold sensitivity of the technique was experimentally estimated at $2 \cdot 10 \text{ cm}^{-1} \cdot \text{W}$.

Optical laser techniques are apparently the most sensitive and high-speed among modern methods available for analyzing gas composition of the atmosphere.¹ An important place among them is occupied by the thermorefractive phase absorption technique; it is classified as calorimetric.² Such techniques are based upon the effect of collisional non-radiative relaxation of molecules of the studied component into their ground state after absorbing resonant pumping radiation. Such absorption results in raising the air temperature within the beam path of the excitation beam. In particular, air refractivity in the heated volume changes, which can be recorded as a phase shift of the sensing lightwave. The source of such a wave is usually a continuously emitting He-Ne laser ($\lambda = 0.63 \text{ }\mu\text{m}$), which produces highly coherent radiation at a stable output power in studies published up to the present time this phase shift of the sensing wave was measured employing passive interferometers, such as the Michelson,³ Mach-Zehnder,⁴ Jamin⁵ instruments. Their sensitivity to changes in air refractivity is determined by the constant steepness of their spread functions. The Fabry-Perot multi-beam interferometer displays higher sensitivity to changes in the optical path length, and, consequently, its use might increase the sensitivity of the technique as a whole. However, this prospect is limited by the technological difficulties of manufacturing interference mirrors of high reflection and low absorption in their reflecting layers.

The two-mode laser interferometer based on a He-Ne laser ($\lambda = 3.3922 \text{ }\mu\text{m}$) with a phase-anisotropic resonance cavity⁶ is free of such drawbacks. Its operating principle is based on the linear dependence of the power $P_{1,2}$ in each of its two orthogonally polarized modes within the range of two-mode lasing $\Delta_{1,2}$ on the optical length L of the cavity. A typical dependence of $P_{1,2}$ on L is presented in Fig. 1 where regions of steep dependence of the output power on the optical length of the resonator (within the range $\Delta_{1,2}$) alternate with regions of single-mode lasing, where the power P_m depends only slightly on that length.



FIG. 1. A typical dependence of output power in one of the modes $P_{1,2}$ on the optical length L of the resonance cavity.

Apparently, changes of the optical length of a specimen, introduced into a two-mode laser interferometer, would produce respective changes in the output power in each mode, proportional to the steepness of the spread function $P_m/\Delta_{1,2}$

$$\Delta P_{1,2} = \pm P_m \frac{\Delta(nl)}{\Delta_{1,2}},$$

where l is the specimen length.⁶

It was demonstrated earlier that the steepness of the spread function of a given laser interferometer is widely adjustable by varying the mode frequency splitting $\omega_{1,2}$. For example, in this way a high sensitivity was achieved in recording the electron component of a pulsed plasma. Clearly the two-mode laser interferometer can be employed to record modulation of air refractivity due to absorption of modulated radiation from the heating laser by the minor component of interest. The technique suggested above was tested by experimentally estimating the content of ethylene C₂H₄, in the air.

We used a single-mode continuously emitting CO₂ laser (transition 00⁰1–10⁰0, line P(20), $\lambda = 10.59 \text{ }\mu\text{m}$), emitting out $P \approx 1 \text{ W}$ of power, as the source of the excitation radiation. The ethylene absorption coefficient at line 10 P(20) is comparatively low; $\kappa = 1.64 \text{ cm}^{-1} \cdot \text{atm}^{-1}$ (Ref. 7). However, it is

convenient for tuning and calibrating the measurement scheme: relatively high pressures (from approximately 10^{-1} to 10 mmHg) of the initial absorbing "C₂H₄-air" mixture can then be employed and measured comparatively accurately by diode mechanotrons during the experiment.

The optical scheme of the experimental setup is given in Fig. 2. The Ne-Ne laser ($\lambda = 3.39 \mu\text{m}$) resonance cavity was formed by two spherical mirrors 1 and 2 ($r = 2 \text{ m}$), one of them positioned within the evacuated cell 3. For the Ne-Ne laser to generate in two orthogonally polarized modes a phase-anisotropic wedge 4 made of crystal quartz was introduced. By shifting it across the laser axis the modal splitting frequency ω_{12} could be varied. To eliminate parasitic reflections from the facets and amplitude anisotropy both windows of the gas discharge tube 5 were oriented along two mutually orthogonal planes, slightly off the laser axis. Output emission in one of the modes was separated by the polarizer 6 made of calcite and was recorded by the photodiode 7.

The CO₂ laser 8 output, modulated by the mechanical chopper 9, was weakly focused by the lens 10 having a focal length of 33 cm. Entrance and exit

windows 11 of BaF₂ served to allow its radiation to pass through the cell. The sensing and heating laser beams intersected at a small angle. Such a paraxial geometry made it possible to exclude absorption of the heating CO₂ laser emission by optical elements of the sensing laser resonance cavity, which otherwise would have inevitably led to noise signals limiting the instrument sensitivity. Inside the cell the beams were aligned by flat aluminum mirrors 12 equipped with adjusting mechanisms. The heating beam was focused in such a manner that the beam neck coincided with the intersection of the axes of the He-Ne and CO₂ lasers beams. In the experiment the effective range of beam overlapping was about 15 cm. A pumping system served to evacuate the cell and fill it with the gas mixture being studied. It was capable of producing calibrated C₂H₄ mixtures. The initial absorbing mixture was diluted to atmospheric pressure by ambient air. Power output from the CO₂ laser was measured by the calorimeter 13.

The signal in the output He-Ne laser emission ($\lambda = 3.39 \mu\text{m}$) was measured employing the synchronous detection technique. The integration time in the experiment amounted to 10 seconds.

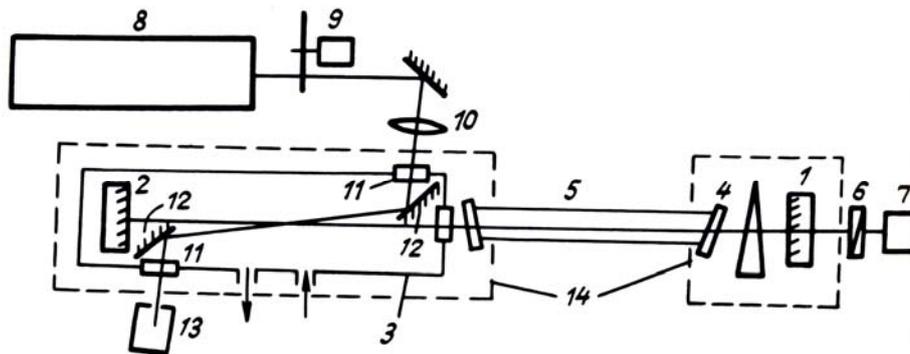


FIG. 2. Optical scheme of the experimental setup

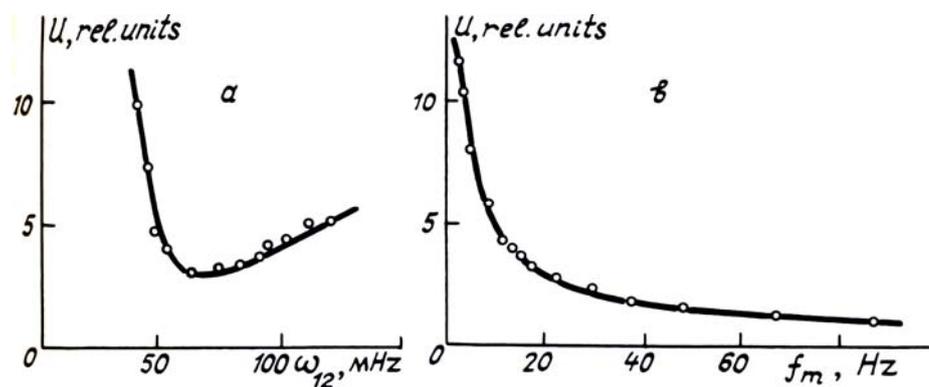


FIG. 3. Experimental dependence of the signal U on the mode-to-mode frequency splitting ω_{12} (a) and on the modulation frequency f_m (b).

a) $f_m = 28 \text{ Hz}$; $P \approx 0.8 \text{ W}$; $P_{\text{C}_2\text{H}_4} = 2 \cdot 10^3 \text{ ppm}$; b) $\omega_{12} = 105 \text{ MHz}$; $P \approx 0.9 \text{ W}$; $P_{\text{C}_2\text{H}_4} = 3.3 \cdot 10^2 \text{ ppm}$.

Using this experimental installation, we measured the dependence of the recorded signal U on the frequency splitting ω_{12} (see Fig. 3a) and on the

pumping radiation modulation frequency f_m (Fig. 3b). The shape of the curve in Fig. 3a for the two-mode laser interferometer reflects, as expected,

the dependence of its spread function steepness on ω_{12} : a slight decrease in steepness accompanies the diminishing frequency splitting down to $\omega_{12} \approx 70$ MHz, and an unlimited sharp surge in steepness occurs at $\omega_{12} \approx 40$ MHz. The stronger signal at higher steepness does not, however, automatically mean higher sensitivity, because the noise in each mode also increases then.

A rapid increase of the recorded signal also follows the decrease of the heating radiation modulation frequency (Fig. 3b). However, lowering this frequency is known to increase the spectral density of power fluctuations of technical origin. The modulation frequency optimal for this particular installation was found to be 17 Hz, providing a maximum signal-to-noise ratio.

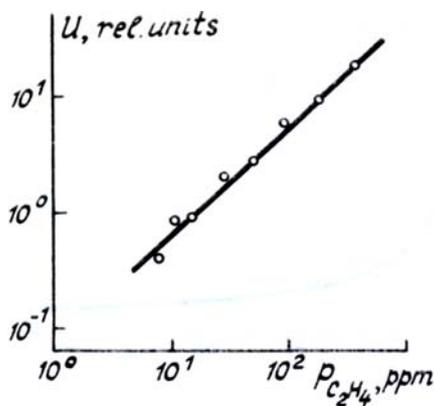


FIG. 4. The dependence of the signal U on the relative pressure of ethylene $P_{C_2H_4}$ ($\omega_{12} = 105$ MHz, $P = 0.8$ W, $f_m = 17$ Hz).

The signal vs. relative ethylene pressure in the air dependence is presented in Fig 4, within the investigated pressure range it displays a linear trend which is quite important from the metrological point of view. As seen from the presented dependence the dynamic range of measurable C_2H_4 concentrations spans at least four-to-five orders of magnitude. Considering the linearity of the dependence, this permits calibrating

the measuring system using certified mixtures containing $10^2 - 10^3$ ppm of pollutant.

Operating experience demonstrated this optical interferometer to be sensitive to air flows penetrating the resonator cavity and to acoustic perturbations of its mirrors.

During the present study we employed a two-layered (wood-porolone) acoustic casing 14 (see Fig. 2) to protect the interferometer from adverse effects of such a nature. It isolated the parts of the resonance cavity, from the ends of the gas-discharge tube to the mirrors. Such a design of the casing permitted the gas-discharge tube capillary to be freely cooled by air, so that no long-term temperature drift of the resonator length took place. Application of such acoustic isolation made it possible to lower the amplitude noise of the recorded signal by an order of magnitude.

From the dependence of the signal on the relative concentration of ethylene in the air (Fig. 4) one can assess the sensitivity of this technique. Extrapolation of the obtained linear dependence down to the noise level product taking account of the value of the C_2H_4 absorption coefficient ($\kappa = 1.64 \text{ cm}^{-1} \cdot \text{atm}^{-1}$), its minimum detectable recorded value: $\alpha_{\min} \approx 2 \cdot 10^{-8} \text{ cm}^{-1} \cdot \text{W}$.

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