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Reaching high sensitivity of radio-acoustic spectroscopy using "strong microwaves"

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Gas molecular spectroscopy is a powerful instrument for both fundamental studies and applications, such as qualitative and quantitative gas analysis, non-invasive medicine, atmospheric remote sensing, etc. Sensitivity is one of the key parameters of any spectrometer, which determines the range of its possible applications for resolving both fundamental and applied problems. The higher the sensitivity, the higher the accuracy of measurement of the spectral line parameters and the greater the number of lines that can be observed in the experiment (the smaller the number of molecules in a gas mixture needed for their lines to appear in the spectrum) and the higher the accuracy with which the properties of the molecules can be explored.

One can recall quite a large number of currently known wideband spectrometers used for a study of the spectra of various molecules in the mm/submm wave range. They can be divided into two types according to the principle of molecular spectra registering: from variations in the characteristics of either the probing radiation (first type) or the gas being studied (second type).

For most of the mm/submm spectrometers, in which either the radiation transmitted through a cell with gas or the radiation reradiated by the gas is detected, a sensitivity close to the limit determined by fundamental physical principles is achieved (see, e.g. [1], chapter 15, p. 414). The only method that permits one to advance in solving the problem of high sensitivity achievement is known as optoacoustic (photoacoustic or radioacoustic) detection of absorption [2].

The sensitivity of any spectrometer is determined by several factors. The most crucial ones are radiation power, detection system noise, and spectral purity of the radiation. In spectrometers of the first type (e.g., classical video-spectrometer), the sensitivity increases with increasing radiation power until the detection system noise becomes surpassed by the radiation noise. Typically for mm/submm video spectrometers with a liquid helium-cooled bolometer an upper limit of radiation power is about 1 mW. Further improvement of a spectrometer sensitivity can be achieved by increasing the optical path length and by data averaging (if ambient and gas conditions are stable enough).

The situation is radically different in spectrometers of the second type (e.g., in a spectrometer with radioacoustic detection of gas absorption – RAD spectrometer). The limiting sensitivity of the spectrometer is obtained when thermal fluctuations of the membrane are defined preferably by the Brownian motion of the gas. These fluctuations do not depend on the radiation power passing through the gas cell. Meanwhile, the useful signal amplitude is directly proportional to the radiation power absorbed by the gas and, therefore, linearly increases with the power. Thus, the sensitivity of RAD method is also linearly increasing.

In this report we present our recent results on reaching the high sensitivity of RAD spectrometer using a few approaches, such as increasing radiation power, modifying the cell and measurement parameters for the noise reduction and data averaging.

Backward-wave oscillators (BWOs) are used as radiation sources in the RAD spectrometers, a series of which covers a very wide range of 35 to 1500 GHz. High stability, low level of the phase noise, and exact knowledge of the frequency are provided by the use of a phase-locked loop system of the BWO radiation referenced by a microwave frequency synthesizer signal, which is synchronized with a frequency and time standard signal. The BWO power can vary from a fraction to tens of mW (reaching more than 100 mW in the best tubes) within the operating frequency range. Larger radiation power suitable for molecular spectroscopy was not available during long time because of the limited choice of mm/submm radiation sources.

Progress in the development of high-power mm/submm radiation sources, which was achieved in recent years, made it possible to test the “power” approach of increasing the RAD sensitivity [3]. In particular, an automated facility [4] based on a gyrotron operated at a frequency of about 263 GHz in the CW regime with a radiation power up to ~1 kW was developed in IAP RAS. This gyrotron permits smooth tuning of the radiation frequency, although within small (in terms of spectroscopy standards) limits of the order of 0.2 GHz due to varying the operating voltage and temperature of the gyrotron cavity.

Experimental spectra of the SO_2 and argon mixture obtained using the RAD spectrometer for different pressures of gas in the cell at two significantly different values of radiation power are shown in Fig. 1. It is clearly demonstrated that an increase of radiation power by about three orders of magnitude leads to a proportional increase of the RAD spectrometer sensitivity. The achieved sensitivity of the spectrometer is not a limit. The spectrometer sensitivity obtained in this study is determined only by the SO_2 line saturation effect, which can be substantially reduced by proper selection of the molecule, the transition, and experimental conditions.

For reducing the influence of external acoustic and mechanical noise the cell was weighted in about ten times by small lead balls 3-mm in diameter (initial cell weight was about 2.5 kg). Analysis of experimental spectra recorded using a cell with significantly different weights shows (Fig. 2) almost sevenfold increase of the SNR of the experimental spectra.

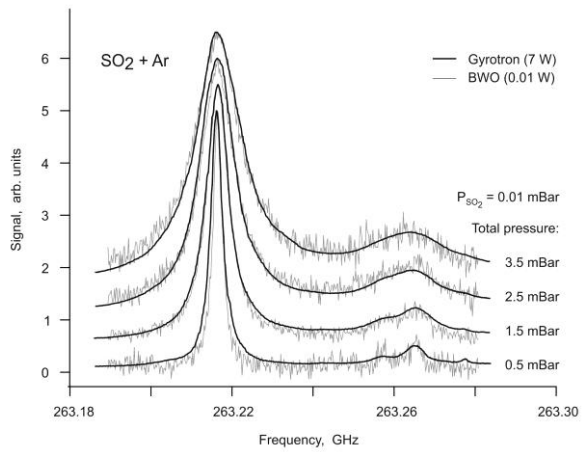

Fig. 1. An example of the experimental spectra of the SO_2 and argon mixture obtained using the RAD spectrometer for different pressures of gas in the cell at a constant level of radiation power. Partial pressure of SO_2 in the mixture is 0.01 mBar. The spectra obtained using a BWO with a radiation power of about 0.01 W (typical power of an OB-24 type tube) and using a gyrotron with output radiation power of about 7 W are shown by gray broken and black smooth lines, respectively. The synchronous detection time constant is 1 s.

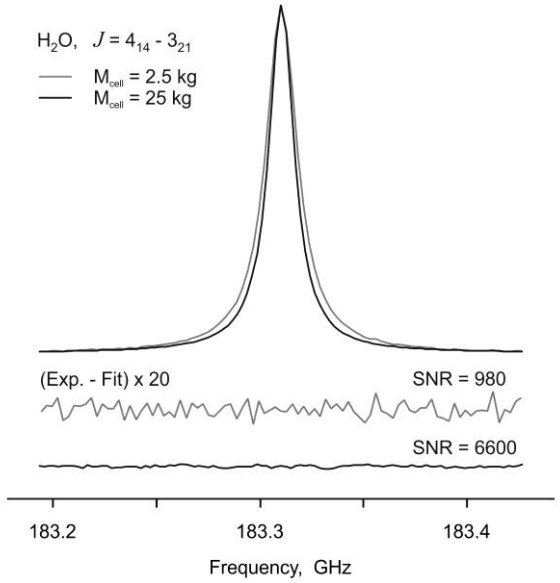

Fig. 2. Recordings of the 183 GHz water line obtained for two significantly different cell weights (2.5 kg and 25 kg). Residuals of the fit of model function to the experimental spectra zoomed in 20 times are shown in the lower part of the figure

Finally, high stability of radiation parameters (frequency and power) and experimental conditions (room and cell temperature, pressure in the cell) allowed averaging a large number of repeated experimental recordings for each chosen pressure for a multiple increasing of SNR for spectra recordings (see Fig. 3). The achieved sensitivity of the RAD spectrometer allowed the first observation of manifestation of the speed-dependence of the collision cross section of the 118-GHz oxygen fine structure line [5].

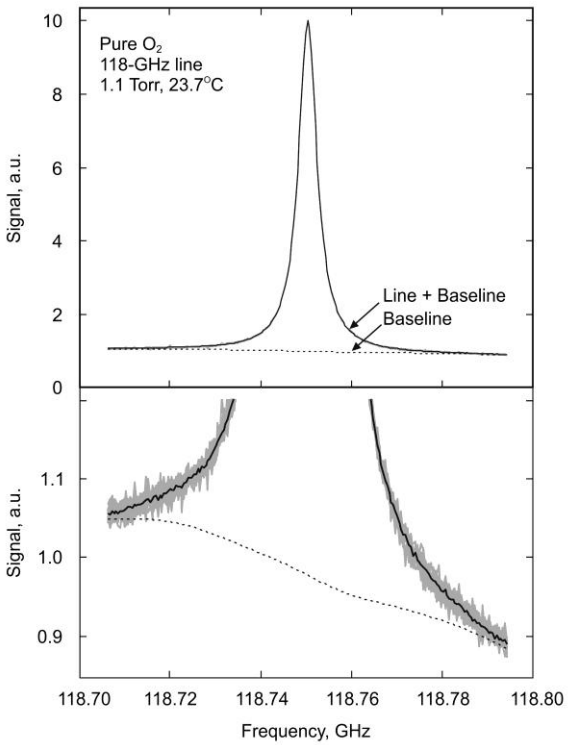

Fig. 3. Experimental spectra of pure oxygen near 118.75 GHz. The lower plot is a zoomed-in part of the upper plot. Twenty spectra recorded at 1.1 Torr of pure oxygen are shown by grey. The black curve is the result of their averaging. The dotted curve shows the baseline recorded for the same conditions with 1.1 Torr of pure nitrogen

It is worth noting that all the approaches were tested separately and being combined in one setup they can give the record sensitivity of the RAD spectrometer.

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