Recent results on THz gyrotron-based molecular spectroscopy

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Method of opto-acoustic (also called radio- or photo-acoustic) molecular spectroscopy is a powerful tool for both fundamental studies and applications. In this method the result of the interaction of the radiation with matter is detected by the change of the parameters of the matter and not of the radiation. It is likely the only method allowing reaching high sensitivity at reasonable recording time by increasing radiation power. This was particularly demonstrated in the teraherz (THz) region in our recent work [1] using spectrometer with radio-acoustic detection of absorption (RAD spectrometer) [2] and radiation of the free-running 263-GHz gyrotron operating in continuous wave regime with up to 1 kW output power [3].

Further improvement of the spectrometer sensitivity and its use for spectroscopy requires precise control of the gyrotron radiation frequency. A phase-locked loop (PLL) against a signal of the reference oscillator was used in order to control the gyrotron modulating anode voltage [4]. The PLL system, which block diagram is presented in Fig. 3, is similar to that used for backward wave oscillator (BWO) [5]. A specially designed fast voltage control unit can vary voltage within 0-1 kV with a speed of somewhat faster 1 kV/μs. Preliminary testing of the control system showed a modulation bandwidth of 150 kHz, defined by the time constant of the gyrotron anode circuit.

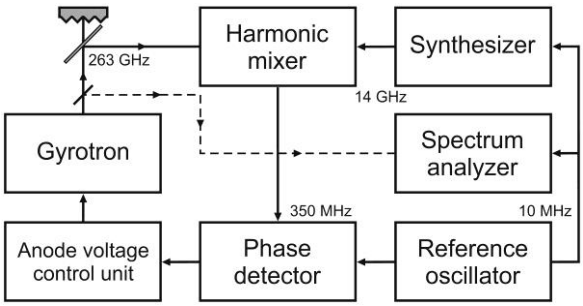

Fig. 1. Block diagram of the phase-locked loop units for a sub-THz gyrotron.

The principle of its operation is following. Part of the output radiation of the 263-GHz gyrotron is transmitted to the harmonic mixer, where it is mixed with the a harmonic of the signal from a microwave synthesizer with a frequency of about 14 GHz. The resulting signal at an intermediate frequency of 350 MHz is then directed to the phase detector for a frequency-phase comparison with the harmonic of a quartz clock serving as the reference oscillator. The error signal from the phase detector is then used as a control signal for the anode voltage control unit. In addition, the radiation spectrum was analized by Keysight spectrum analyzer N9010A equipped with an external mixer. Phase-locking the gyrotron frequency reduced the radiation spectrum width from 0.5 MHz for a free-running gyrotron down to about 1 Hz for the stabilized gyrotron (Fig. 2) which corresponds to relative frequency stability $\Delta f/f \sim 3 \cdot 10^{-12}$ for a measurement time of a few seconds.

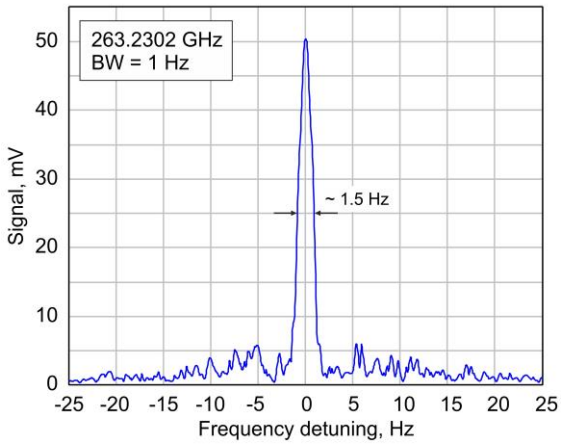

Fig. 2. Frequency spectrum of the gyrotron observed at 263.2302 GHz with phase-locked loop. Bandwidth of the spectrum analyzer was 1 Hz.

Our recent spectroscopic study using RAD method and a free running gyrotron facility [1] revealed the necessity for modulation of the gyrotron frequency aimed at reduction of the spectrometer baseline. It became possible by implementation of the PLL system of the gyrotron. Typical recordings of SO_2 line obtained using RAD spectrometer and gyrotron radiation at approximately 1 W power are shown in Fig. 3. Modulation of either radiation power (Fig. 3A) or its frequency along with synchronous signal detection at the first (Fig. 3B) and second (Fig. 3C) harmonic of the modulation frequency were applied for spectra recording. The line asymmetry due to baseline is clearly observed from the experimental spectra in case of power modulation in Fig. 3A. The situation is even worse if a weaker line is studied. Use of the frequency modulation decreases influence of the baseline on the line shape which is seen in Figs. 3B and 3C.

Experimental spectrum of a mixture of SO_2 (0.027 Torr) and Ar (0.26 Torr) was registered in the frequency range 263-264 GHz using RAD method and gyrotron radiation of about 0.5 W power (Fig. 4A). Calculated spectrum shown in Fig. 4B was modeled using Lorentz profile and line parameters from HITRAN and JPL catalogs.

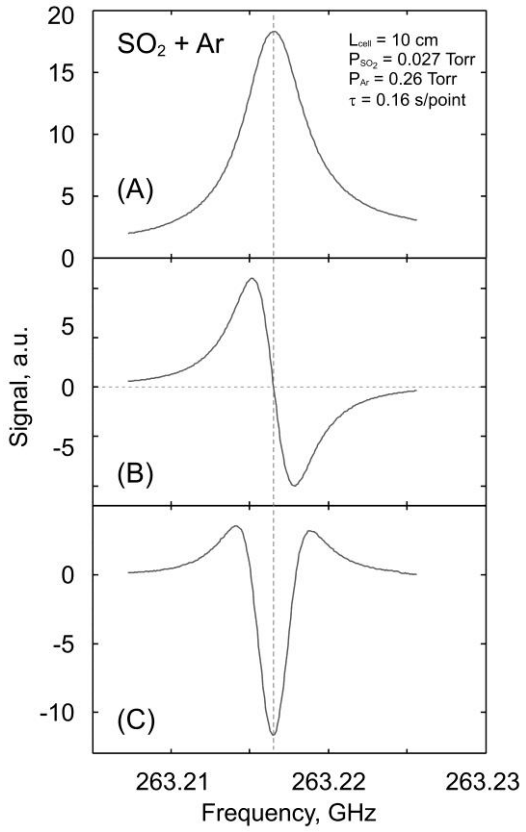

Fig. 3. Experimental recordings of the $44_{6,38} - 45_{5,41}$ line of $^{32}S^{16}O_2$ in a mixture with argon obtained using RAD spectrometer and gyrotron radiation (~ 1 W). (A) - modulation of the radiation power; (B) and (C) - modulation of the radiation frequency and signal detection at first (B) and second (C) harmonic of the modulation frequency (180 Hz).

It follows from the comparison of these two spectra that only some of the observed lines of the well-studied spectrum of SO_2 can be assigned to the tabulated ones. Futher analysis of the experimental spectra revealed that unassigned lines are narrower than the assigned ones. This may indicate that these lines arise from an interaction of absorbing molecules with radiation at second, third, etc. harmonics of the fundamental frequency. Calculated specta in the 526-528 GHz and 789-791 GHz ranges are shown in Fig. 4B and 4C, respectively (note frequency scales normalized by the corresponding harmonic number). Comparison of the calculated spectra with the experimental one confirmes presence of radiation at second and third harmonics of the gyrotron fundamental frequency. Using known parameters of SO_2 lines we estimated relative fraction of the radiation power coming to the gas cell at second harmonic as ~ 1% of the full power and ~ 0.1% for the third harmonic. Routine sensitivity of the spectrometer was determined from spectra analysis as ~ $8 \cdot 10^{-10}$ cm^{-1} (at 2-s time constant), which is limited by the power saturation effect of the molecular transition.

Results were confirmed by the high resolution spectra obtained using a BWO-based video spectrometer.

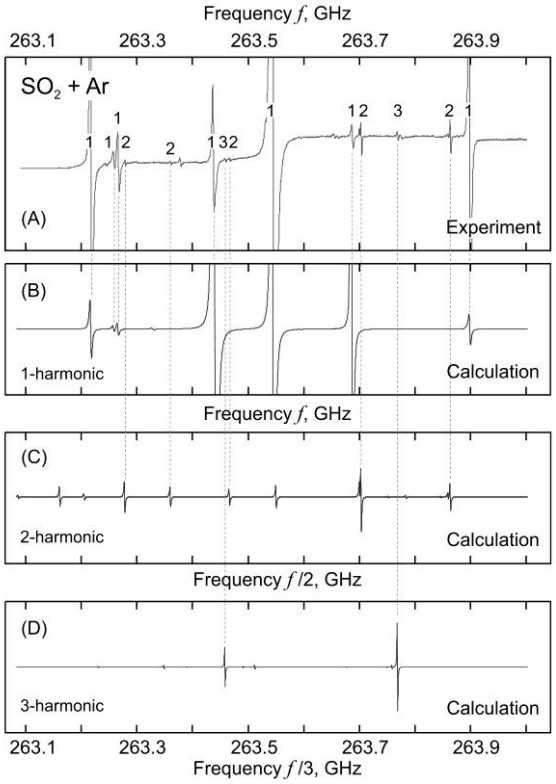

Fig. 4. (A) Experimental spectrum of a mixture of SO_2 and Ar obtained using RAD and gyrotron radiation of ~ 0.5 W power. Lines belonging to the 1st, 2nd and 3rd harmonics of radiation frequency are denoted by the corresponding numbers. (B,C and D) Calculated spectra of SO_2. Frequency scales are normalized by the corresponding harmonic number.

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References

1. *Koshelev M.A., et al.* Molecular Gas Spectroscopy Using Radioacoustic Detection and High-Power Coherent Subterahertz Radiation Sources // J. Molec. Spectrosc. 2017. V. 331, P. 9-16.

2. *Tretyakov M.Yu., et al.* Precise Measurements of Collision Parameters of Spectral Lines with a Spectrometer with Radioacoustic Detection of Absorption in the Millimeter and Submillimeter Ranges // Instrum. Exp. Tech. 2008. V. 51. P. 78–88.

3. *Glyavin M.Yu., et al.* Experimental tests of a 263 GHz gyrotron for spectroscopic applications and diagnostics of various media // Rev. Sci. Instr. 2015. V. 86 N. 5 P. 054705.

4. *Fokin A. et al.* High-power sub-terahertz source with a record frequency stability at up to 1 Hz // Scientific Reports. 2018. V. 8. P. 4317.

5. *Krupnov A.F.* Phase Lock-In of MM/SUBMM Backward Wave Oscillators: Development, Evolution, and Applications // Int. J. IR MM Waves. 2001. V. 22. P. 1-18.