International Journal of Infrared and Millimeter Waves, Vol. 20, No. 10, 1999

PRECISION RESONATOR MICROWAVE SPECTROSCOPY IN MILLIMETER AND SUBMILLIMETER RANGE

A. F. Krupnov, M. Yu. Tretyakov, V. V. Parshin, V. N. Shanin, and M. I. Kirillov

Applied Physics Institute of Russian Academy of Sciences
46 Uljanov St
603600 Nizhnii Novgorod GSP-120, Russia

Received August 7, 1999

1 Abstract

By the use of Fabry–Perot resonator with quality factor $\simeq 600\,000$ and fast precision (down to one Hertz) frequency control of coherent millimeterwave radiation source the 20 Hz accuracy in measurements of the width of resonance curve is obtained. This accuracy corresponds to the detection of 1.8×10^{-3} dB/km absorption in the sample filling the resonator and exceeds the before known sensitivity more than by an order of magnitude. The example of precise measurement of 60 GHz oxygen absorption band in the real atmosphere is presented. The new possibilities of application of precision resonator microwave spectroscopy to the atmospheric problems as well as to the ultra-low absorptions measurements in dielectrics and metal surfaces up to Terahertz frequencies are discussed.

Keywords: millimeterwave, submillimeterwave, Fabry – Perot, resonator, spectroscopy.

0195-9271/99/1000-1731$16.00/0

2 Introduction

Resonator spectroscopy is one of the main directions of progress of microwave spectroscopy [1]. The sample absorption measurement by resonator spectroscopy comes after all to the measurements of quality factors of the empty and loaded resonator. The resonator quality factor Q is defined as the ratio of the frequency of the resonance f_0 to the width of the resonance Δf. The sensitivity of resonator spectroscopy increases with increase of the resonator quality factor and increase of accuracy of measurement of the width of the resonance. In the present work we used Fabry–Perot resonator with quality factor nearing the limit for non–cooled mirrors [2], and developed the methodics of measurement of the resonance width with highest at the present time accuracy. Combination of the Fabry–Perot resonator with quality factor $\simeq$ 600 000 and fast, precisely controlled and powerful enough synthesized radiation source permitted us to reach accuracy of resonance width measurement 20 Hz at $\simeq$150 kHz resonance width in millimeter wave range and to obtain absorption coefficient sensitivity 0.0018 dB/km, exceeding the sensitivity previously known [3] more than by an order of magnitude.

3 Experimental setup

The most important parts of the experimental setup are the Fabry–Perot resonator and the system of fast precise control of the radiation frequency.

Quality factor of Fabry–Perot resonator having 26.5 cm length, spherical silver–plated mirrors 12 cm in diameter and 24 cm curvature radius, coupled with radiation source and detector by 6 micrometers Teflon film placed under 45 degrees to the resonator axis, was defined by unavoidable reflection losses (silver is the best reflecting material in millimeter wave range). The approach to the construction of high quality factor Fabry–Perot resonators is outlined in [2].

The Backward Wave Oscillator (BWO) [4] served as radiation source. Its frequency was stabilized by phase lock–in system against harmonic of microwave frequency synthesizer as described in [5]. The important improvement of the phase–lock system should be noted. The main source of error in the resonance width measurement is drift

of the resonance central frequency. Millimeterwave frequency synthesizers [6] have 10 to 40 millisecond switching time, thus preventing fast scanning of the resonance curve. Response time of the resonator itself $\tau \sim 1/\Delta f \sim 5$ microseconds. For precision measurement the observation time should be increased, say, in factor of ten i.e. up to $\geq$50 microseconds. In the present work the phase lock–in system was completed by the fast radiofrequency synthesizer with switching time ~200 nanoseconds and time between switching $\geq$50 microseconds, which provided the reference signal for phase detector in the lock–in loop, thus permitting precision and fast scanning of the BWO radiation frequency within ~200 MHz around the frequency defined by microwave frequency synthesizer. Results of each fast scan were recorded and processed separately. Radiation passed through the resonator was received by Shottky diode detector. The precision frequency control, signal aquisition and processing were done by computer.

4 Results

The example of experimental resonance curve of Fabry–Perot resonator record at $f_0 \simeq 85.139$ GHz and result of its fitting to Lorentzian curve (coinciding in the figure scale), and multiplied by 100 residual of the fit are presented in Fig. 1. Frequency scanning was chosen to be faster than characteristic times of changes of experimental conditions (air movements, temperature drifts etc), but slow enough for not to distort resonance curve. In the example scanning of 500 frequency points was done in 2 kHz steps, 60 microseconds/step and total time 15 seconds (500 scans). Increase of noise on the slopes of resonance curve visible in the figure is produced by transformation of phase noise of the radiation source to the amplitude one. Accuracy of measurement of resonance width equals to 20 Hz with the resonance width $\Delta f \simeq 164\ 728$ Hz.

As an example of application of the precision resonator spectroscopy we measured *in situ* atmospheric oxygen absorption band around 60 GHz produced by the clustering of magnetodipole transitions of fine structure of the rotational spectrum of this paramagnetic molecule (see, e.g., [7]). Air inside the open resonator served as a sample. Parameters of atmosphere were registered. We studied frequency

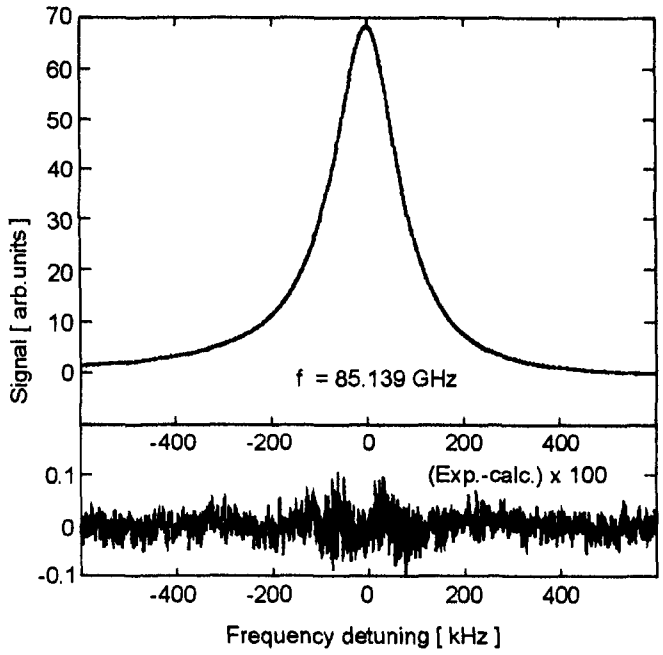

Figure 1: **Resonance curve of Fabry–Perot resonator record (500 centered scans). Residuals of the fit multiplied by 100 are presented below. Measured width of the resonance (FWHM) is equal to $\Delta f = 164\ 728(20)$ Hz.**

region from 46 to 85 GHz. The resonator quality factor was measured through each 560 MHz and the obtained values served for calculation of absorption.

In a broad band (as compared with measurements in one frequency point) some uncontrollable changes of quality factor were observed by the variation of coupling coefficient with frequency, produced, as it seems, by the changes of the BWO radiation beam exciting the resonator. This somewhat reduced absolute sensitivity in the studies of absorption dependence in a frequency range.

The dependence of atmosphere absorption vs. frequency is presented in Fig. 2. The experimental results obtained in different days at somewhat different parameters of atmosphere are presented by cir-

cles and crosses. Solid line corresponds to the absorption calculated in accordance with [3]. Multiplied by 10 residual of the fit is shown in the lower part of the figure. In the work [3] uncertainty of the results was estimated as $\sim \pm 2\%$. Our results approximately confirm estimation [3] (rather as $\pm 3\%$). But much higher sensitivity obtained in the present work in comparison with [3] permitted to find out inside the $\pm 3\%$ residuum the regular deviations from the band profile visible in Fig. 2 with signal-to-noise ratio ~ 10. The recommended in [3] variations of broadening and mixing parameters even in broad ranges did not change the regular appearance and did not lessen of the order of magnitude of residuum, what led to conclusion that the model accepted in [3] and widely used in atmosphere absorption calculations describes the oxygen 60 GHz band only within $\sim \pm 3\%$, what is less than accuracy of the modern experiment. More detailed discussion of this falls out of scope of the present paper and will be done elsewhere.

5 Discussion

Development of precision resonator microwave spectroscopy with sensitivity improved by more than order of magnitude and significantly fast opens the new possibilities in many fields of study.

In the atmosphere investigations new possibilities are opened not only in atmospheric lines studies, but also in real time dynamics of the atmospheric processes studies (atmosphere monitoring), separation of the effects defining the atmosphere radiation absorption and study of their dependencies on parameters. Direct measurement of low humidities in higher atmosphere from the board of flying apparatus, solution of the question of existence of non-resonant atmospheric absorption in the atmospheric windows, etc become possible.

The technique also gives new possibilities for ultra-low absorption studies in dielectric and metal surfaces. The reflection coefficient measurements of the metals, as it is known, gives opportunity to study Fermi surfaces and structure of the metals. It is possible now to measure the difference of reflection coefficient of silver from unity with accuracy 10^{-4}. Obtained in the present work loss tangent sensitivity 4×10^{-10} (for fully filled cavity) permits to measure absorption in thin films of all known dielectrics (including best CVD diamond films [8])

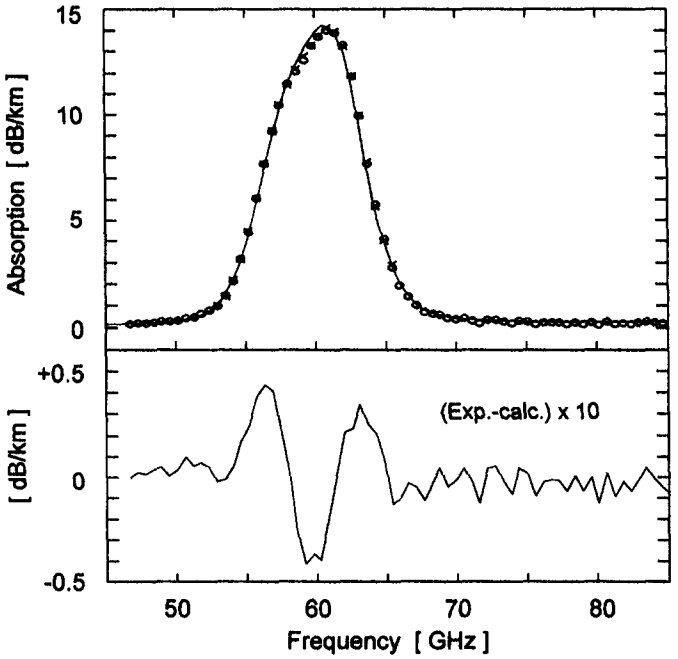

Figure 2: **Atmosphere oxygen O_2 absorption band around 60 GHz. Circles and crosses – this work experiment, curve – calculation according to [3]. Residual multiplied by 10 is shown below.**

with the large reserve of sensitivity. Silver and diamond have smallest (i.e., most difficult to measure) losses in metals and dielectrics correspondingly.

Existence of the technique able for such measurements in the continuous band up to Terahertz frequencies [5, 6, 9] gives possibility of broad variations of the studied objects.

6 Acknowledgements

Authors express their gratitude to B.A. Sakharov for valuable discussion of the construction of fast frequency synthesizer, V.V. Dorovskikh for the help in data processing, and A.B. Mazur for the help

in calculations. The work was supported by the Grants of RFBR N 97-02-16593 and DFG - RFBR N 98-03-04072 and State Programs "Fundamental Metrology" and "Physics of the Microwaves", to which authors express their gratitude.

References

[1] C.H. Townes, A.L. Shawlow, Microwave Spectroscopy, Dover, N.Y, (1975).

[2] A.F. Krupnov, V.V. Parshin, G.Yu. Golubyatnikov, I.I. Leonov, Yu.N. Konoplev, IEEE Trans. on Microwave Theory Tech, **47**, 284 - 289 (1999).

[3] H.J. Liebe, Int. J. Infrared and Millimeter Waves, **10**, 631 - 635 (1989); H.J. Liebe, P.W. Rosenkranz, G.A. Hufford, J. Quant. Spectr. Radiat. Transfer, **48**, 629 - 638 (1992).

[4] 4. M.B. Golant, R.L. Vilenskaya, E.A. Zyulina, Z.F. Kaplun, A.A. Negirev, V.A. Parilov, E.B. Rebrova, V.S. Saveliev, Pribory i Technika Eksperimenta (in Russian; available in English as Sovjet Physics - Pribory) N 4, 136 - 139 (1965).

[5] 5. A.F. Krupnov, Izv. VUZov, Radiofizika (in Russian; available in English as Radiophysics and Electronics), **41**, 1361 - 1374 (1998); A.F. Krupnov, Spectrochimica Acta A, **52**, 967 -991 (1996).

[6] A.F. Krupnov, O.P. Pavlovsky, Int. J. Infrared and Millimeter Waves, **15**, 1611 - 1624 (1994).

[7] W. Gordy, R.L. Cook, Microwave Molecular Spectra, Interscience (Wiley), N.Y, (1984).

[8] V.V. Parshin, V.G. Ralchenko, V.I. Konov, Proc. 23rd Int. Conf. Infrared Millim. Waves, Colchester, UK, Sept. 7-11 (1998), p. 232.

[9] G. Winnewisser, A.F. Krupnov, M.Yu. Tretyakov, M.Liedtke, F. Lewen, A.H. Saleck, R. Schieder, A.P. Shkaev, S.A. Volokhov, J. Mol. Spectrosc, **165**, 294 - 300 (1994).