

Use of high resonant-radiation powers to increase the sensitivity of microwave spectroscopes

I. I. Antakov, S. P. Belov, L. I. Gershtein, V. A. Gintsburg, A. F. Krupnov, and G. S. Parshin

Radiophysics Research Institute

(Submitted April 8, 1974)

ZhETF Pis. Red. 19, 634-637 (May 20, 1974)

We report the first use of sources of high-power coherent radiation (up to 10^3 W) at the resonant frequency of the observed spectral line of a gas in a microwave spectroscope with an acoustic detector. A sensitivity $(1-2) \times 10^{-11}$ cm $^{-1}$ is obtained in the 34-GHz band. The method can be used in the submillimeter region.

1. The possibility of greatly improving the sensitivity of microwave spectroscopes (including those operating in the millimeter and submillimeter bands) by using sources with large radiation powers at resonant frequencies of the observed lines and employing acoustic detection was pointed out earlier.^[1,2] The maximum signal power that can be received in observing a spectral transition in matter, i.e., the maximum power absorbed by the gas, is

$$P_c \sim \frac{1}{2} n h \nu \Delta \nu, \quad (1)$$

where n is the difference of the level populations, ν is the transition frequency, and $\Delta \nu$ is the homogeneous line width. This signal power is reached by passing through the sample a resonant transition-inducing radiation with intensity sufficient to saturate the transition

$$P_0 \sim \frac{3A^2(\Delta \nu)^2 c}{8\pi |\mu_{ij}|^2} S, \quad (2)$$

where $|\mu_{ij}|^2$ is the square of the matrix element of the dipole moment of the transition and S is the cross section of the sample. A natural way of increasing the signal from the lines is to increase P_0 and $\Delta \nu$. To increase the sensitivity of the spectroscope it is necessary that the increase of the radiation power not be accompanied by a corresponding increase of the noise of the receiver for the signals from the lines. This is precisely what is attainable in principle in a microwave spectroscope with acoustic detector (MAD), which belongs, according to the classification introduced in^[2], to the group of spectral instruments in which 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. We describe in this paper a practical investigation of the possibilities of increasing the sensitivity of spectroscopes by increasing the power of the resonant radiation. The source of the powerful coherent radiation is the MTsR generator,^[3] which is a maser operating at cyclotron resonance (CRM) in the 34 GHz band.

2. The absorption line in the gas was observed with amplitude modulation of the CRM radiation power passing through the MAD cell with thin mica windows, and then directed to a calorimeter comprising a matched load. The modulation frequency was 180 Hz. To protect the MAD microphone from the action of the microwave power, the channel connecting the cell with the microphone was constructed in the form of a packet of wave-

guides operating beyond cutoff. In view of the restricted tuning range of the available CRM, the choice of the investigated objects was considerably limited. In particular, we observed the $6_{15}-6_{16}$ line of formic acid HCOOH at the frequency 34378.86 MHz, the absorption coefficient of which in pure HCOOH vapor was $\gamma_0 = 5.4 \times 10^{-6}$ cm $^{-1}$, and the matrix element of the transition was close to 0.1 Debye unit. The line width was chosen to be large enough to exclude saturation up to powers on the order of 10^3 W, and according to the measurements it amounted to 70 MHz at half-intensity level. The broadening was effected by adding an extraneous gas, N $_2$ O in this case.¹⁾ The decrease of the absorption coefficient of the HCOOH lines as a result of the dilution with N $_2$ O was determined from the ratio of the output signals from known lines of N $_2$ O and HCOOH in the 300 GHz band, observed in the same cell before and after the experiments with the CRM. The attenuation coefficient was found to be 1.54×10^{-2} , i.e., the absorption coefficient of the observed $6_{15}-6_{16}$ line after dilution was $\gamma = 8.4 \times 10^{-8}$ cm $^{-1}$.

Figure 1 shows the MAD signal at the center of the $6_{15}-6_{16}$ line of dilute HCOOH at a CRM radiation power 10^3 W, and also the noise of the receiving part of the MAD with the gain increased 100 times (and with the modulation turned off). The noise did not depend on the presence or absence of CRM radiation passing through the cell; the signal from the line, however, increased in proportion to the radiation power (Fig. 2). In addition to the resonant signal from the spectral line, we observed also a weaker signal of nonresonant character, the nature of which is not yet clear. The separation of only the resonant part of the signal can be easily realized by using frequency resonant amplitude modulation of the radiation.^[1-3] The ratio of the signal to the rms deviation from the noise, determined from Fig. 1, is ≈ 6000 , i.e., the sensitivity of the MAD in our experiments, at a CRM power 10^3 W, was only $(1-2) \times 10^{-11}$



FIG. 1. Signal from the $6_{15}-6_{16}$ line of HCOOH, observed with the aid of an MAD and a CRM. The line absorption coefficient was 8.4×10^{-8} cm $^{-1}$ (HCOOH diluted with N $_2$ O), the radiation power was 10^3 W, the noise was plotted with the gain increased 100 times, and the time constant used in plotting was 1 sec.

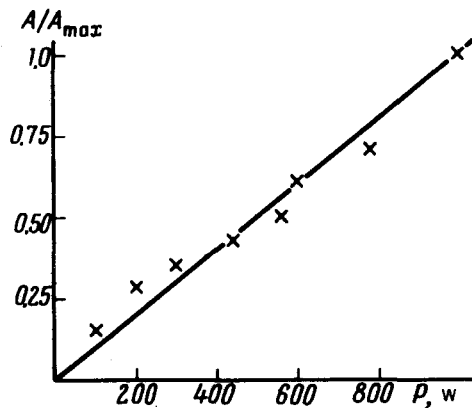


FIG. 2. Signal of the $6_{15}-6_{16}$ HCOOH line vs. the CRM radiation power in relative units.

cm^{-1} , which exceeds by more than one order of magnitude the sensitivity microwave spectroscopes of the ordinary type in this band.^[4]

3. Thus, we have experimentally confirmed the previously indicated possibility of exceeding, with the aid of MAD and powerful radiation forces, the sensitivity of ordinary microwave spectroscopes. In practice, this method of increasing the sensitivity is applicable not to all spectra; it is best applicable to the case of rare spectral lines with very small transition dipole-moment matrix elements. On the other hand, the use, for the first time, of high radiation powers at the frequency of the observed spectral lines opens up a new field of research in microwave spectroscopy, the investigation of nonlinear effects such as, for example, the observation of rotational spectra of nonpolar molecules that acquire a dipole moment as a result of polarization by an elec-

tric field (the corresponding calculation will be published separately), etc. A good object of the investigation may be the "forbidden" spectra.^[5] The described "force" method of increasing the sensitivity appears to be at present the only method of obtaining such a high sensitivity in the submillimeter and far infrared regions of the spectrum inasmuch as the sensitivity of the MAD does not depend on the frequency, and there exist powerful (albeit not yet tunable) radiation sources (see, e.g.,^[6]). We note that the obtained sensitivity is one third the theoretical value,^[2] thus indicating that further development in this direction is promising.

The authors thank A.V. Gaponov, V.A. Flyagin, and V.K. Yulpatov for constant interest in the work.

¹Large HCOOH concentrations affect adversely the lavsan membrane of the MAD microphone; in addition, the decrease of the absorption coefficient prevented the receiving apparatus from becoming overloaded.

¹A. F. Krupnov, L. I. Gershtein, V. G. Shustrov, and S. P. Belov, *Izu. VUZov Radiofizika* 13, 1403 (1970). S. P. Belov, A. V. Burenin, L. I. Gershtein, V. P. Kazakov, E. N. Karyakin, and A. F. Krupnov, *ZhETF Pis. Red.* 18, 285 (1973) [*JETP Lett.* 18, 167 (1973)].

²S. P. Belov, A. V. Burenin, L. I. Gershtein, V. V. Korolikhin, and A. F. Krupnov, *Opt. Spekt.* 35, 295 (1973).

³A. W. Gaponov, M. I. Petelin, and V. K. Yulpatov, *Izu. VUZov Radiofizika* 10, 1414 (1967).

⁴C. Townes and A. Schawlow, *Microwave Spectroscopy*, McGraw Hill; *MRR Spectrometer*, Hewlett Packard, Jan. 1970; H. W. Harrington, I. R. Hearn, and R. F. Kanskolb, *Hewlett-Packard Journal* 22, 2 (1971).

⁵K. Fox, *Phys. Rev. Lett.* 27, 233 (1971); J. K. A. Watson, *J. Mol. Spectr.* 40, 536 (1971).

⁶T. A. De Temples, T. K. Plant, and P. D. Coleman, *Appl. Phys. Lett.* 22, 644 (1973).