# SENSITIVITY OF MICROWAVE SPECTROMETERS USING MASER TECHNIQUES

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The minimum detectable absorption coefficient for an absorbing medium in a microwave waveguide has been written[1]

$$\alpha_{\min}=2e\alpha_0(2k_e T_e\Delta\nu/P_0)^{1/2}, \tag{1}$$

where $\alpha_0$ is the absorption coefficient per unit length of the waveguide, $e$ the Napierian base, $k$ Boltzmann's constant, $P_0$ the microwave power used, $T_e$ the effective noise temperature of the amplifying system, and $\Delta\nu$ its bandwidth. Most microwave spectrometers have involved noise temperatures $T_e$ of 10 000°K or greater. It is obvious that use of maser amplifiers can very much decrease $T_e$ in expression (1) and thus improve spectrometer sensitivity. It will be shown here that additional large gains even beyond the reduction of $T_e$ in expression (1) can be obtained by suitable use of maser-like techniques, assuming that sensitivity is limited only by random noise.

If the microwave power is sent into a perfect maser amplifier following partial absorption in a waveguide, one might expect that the noise temperature $T_e$ would be as low as $h\nu/k$.[2] But this is not true, since the waveguide is normally at a temperature above $h\nu/k$ and radiates noise into the amplifier, making $T_e$ comparable with the waveguide temperature $T_g$. Consider now the addition along the length of the waveguide of molecules in an excited state, which can undergo stimulated emission to a lower state. The noise radiated by the waveguide, including the excited molecules, is characterized by the temperature[2]

$$T_g'=\frac{h\nu}{k}\,\frac{a}{b-a}, \tag{2}$$

where $a$ is the total probability of stimulated emission per unit time per photon and $b$ is the total probability of absorption per unit time per photon. $b$ includes all losses in the waveguide. As excited molecules are added, $a$ approaches $b$ and $T_g$ becomes very large. However, the net absorption in the waveguide decreases since $\alpha_0=(b-a)/C$, where $C$ is the velocity of light, and hence $\alpha_{\min}$ appears to decrease without limit even with $\Delta\nu$ and $P_0$ fixed. A more complete analysis can be easily obtained with the help of expressions (14) and (15) of reference 2 for the waveguide case or of expressions (58) for the resonant cavity spectrometer.

If $n_0$ photons are introduced into a waveguide, the noise fluctuation when $b>a$ is

$$\Delta n=\left[n_0\left(\frac{a+b}{b-a}\right)\right]^{1/2} e^{(a-b)t/2}. \tag{3}$$

The signal due to a small change $\epsilon$ in $(b-a)$ associated with gas absorption is

$$S=n_0\, e^{(a-b)t}\epsilon t. \tag{4}$$

If this is regarded as a peak signal, and the rms signal is equated to $\Delta n$, then $\Delta n=S/\sqrt{2}$. After optimization with respect to the time $t$, one has from (3) and (4)

$$\epsilon=e[2(b-a)(b+a)/n_0]^{1/2}. \tag{5}$$

A similar expression is obtained when $a>b$, except that $(b-a)$ becomes $(a-b)$. If $b$ is due to normal absorption, then

$$b=\alpha_0(CkT_g/h\nu), \tag{6}$$

where $T_g$ is the temperature of the waveguide walls. Furthermore $a\approx b$ so that (5) gives for the minimum detectable absorption coefficient

$$\alpha_{\min}=2e(\alpha_0\alpha_0')^{1/2}(2kT_g\Delta\nu/P_0)^{1/2}. \tag{7}$$

Here $\alpha_0'=(b-a)/C$ can in principle be made as small as desired by adding excited molecules to increase $a$. The power level introduced into the waveguide is $P_0=n_0h\nu\Delta\nu$. Expression (7) should be compared with (1). When $T_e=T_g$, (1) gives approximately the best sensitivity obtainable by a perfect amplifier following the waveguide. (7) gives further increased sensitivity when excited atoms are added within the absorption cell. (7) also assumes that the following amplifier used is sufficiently good so that its effective noise temperature is much less than $T_g'$, which is not impractical to achieve.

Consider now a cavity spectrometer, for which the minimum detectable absorption coefficient is normally given by[3]

$$\alpha_{\min} \approx \frac{2\pi}{Q_c\lambda}\left(\frac{4kT_e\,\Delta\nu}{P_0}\right)^{1/2}$$

$$\approx \frac{2}{\lambda}\left(\frac{2\pi kT_e\,\Delta\nu}{WQ_c\,\nu}\right)^{1/2}, \tag{8}$$

where $Q_c$ is the cavity quality factor, $\lambda$ the wavelength, and $W$ the microwave energy stored in the cavity. Following Shimoda, Takahasi, and Townes,[2] the average number of photons in a cavity when $b>a$ is

$$\bar{n}=c/(b-a), \tag{9}$$

and the noise fluctuation is

$$\Delta n=(bc)^{1/2}/(b-a). \tag{10}$$

Here $c$ is the probability per unit time of introduction of a photon by any process (including spontaneous emission) other than stimulated emission. The signal due to a small change $\epsilon$ in $b-a$ is from (9)

$$S=c\epsilon/(b-a)^2. \tag{11}$$

For signal equal to noise, from (10) and (11),

$$\epsilon_{\min}=(b/c)^{1/2}(b-a)=(bc)^{1/2}/\bar{n}=[(b-a)b/\bar{n}]^{1/2}. \tag{12}$$

The last form of (12) shows the similarity with expression (5) for the waveguide. Sensitivity is best when $c$ is as small as possible, i.e., when $c=a$. Since also $a\approx b$, and $\bar{n}=W/h\nu$,

$$\epsilon_{\min}=b(n\Delta\nu/W)^{1/2}, \tag{13}$$

or

$$\alpha_{\min}=\frac{4\pi}{\lambda Q_c}\,\frac{kT_c}{h\nu}\left(\frac{h\Delta\nu}{W}\right)^{1/2}, \tag{14}$$

where $T_c$ is the wall temperature of the cavity, and $b=2kT_c/h\nu$.

Expression (14) is to be compared with (8). If a perfect amplifier is used outside the cavity, $T_e\approx T_c$ and (8) gives

$$\alpha_{\min}=\frac{2}{\lambda}\left(\frac{2\pi kT_c}{WQ_c}\,\frac{\Delta\nu}{\nu}\right)^{1/2}. \tag{15}$$

If the cavity is cooled so that $kT_c<h\nu$, then (8) may be written

$$\alpha_{\min}\approx\frac{2}{\lambda}\left(\frac{2\pi h\Delta\nu}{WQ}\right)^{1/2}. \tag{16}$$

The minimum detectable absorption coefficient given by (14), which applies when excited molecules are appropriately introduced directly into the cavity where absorption is to be detected, can hence be markedly less than the best achievable with external amplification alone, as given by either (15) or (16). It is interesting to express (14) in terms of the minimum detectable number of absorbing atoms or molecules. If $a$ approaches $b$, spontaneous emission is amplified more and more and the bandwidth $\Delta\nu$ to which the cavity system will respond becomes smaller, since $\Delta\nu=\nu(b-a)/a$.[2] Let $a$ approach $b$ until $W$ builds up to the point of saturation of the molecules to be detected. Then if $\alpha$ is expressed in terms of the total number of molecules $N$ (in upper and lower states) producing the absorption, one can show that (14) gives for the minimum number $N$ which can be detected in a time $1/\Delta\nu$

$$N_{\min}\approx\frac{16Q_L}{Q_c}\,\frac{kT_c}{h\nu}\,\frac{kT_N}{h\nu}, \tag{17}$$

where $Q_c$ is the quality factor for the cavity before insertion of molecules in the excited state, and $Q_L$ is $\nu/\delta\nu$ for the absorption line. Here the linewidth $\delta\nu$ is assumed due to relaxation processes alone; otherwise expression (17) is somewhat modified. $T_N$ is the temperature of the absorbing material, which would usually be the same as $T_c$ for the cavity walls, and we assume $kT_N\gg h\nu$. It is evident that some conditions allow detection of only a few molecules, if limitations are due to random noise only. Note also that (17) is independent of the matrix element for the absorption. The time required for detection of the number of atoms given by (17) is

$$t=1/\Delta\nu=3hV/(8\pi\mu^2Q_L^{\,2}), \tag{18}$$

where $\mu^2$ is the square of the matrix element for the transition being detected, and $V$ the cavity volume. Thus the matrix element does affect the time required for such measurement. For $|\mu|=10^{-18}$ esu, $V=1$ cm$^3$, and $Q_L=10^6$, (18) gives $t\approx10^{-3}$ sec. Hence the time required for sensitivities indicated by (17) is not excessively long.

A spectrometer involving some of the above principles, utilizing excited molecules directly in the cavity and a change in $b-a$, has actually already been demonstrated without perhaps its potentiality and general applicability being fully realized. This was the experiment of Shimoda and Wang[4] on an oscillating beam-type maser. There the sensitivity was found to be much greater than that for an ordinary cavity-type spectrometer; the factor of improvement obtained in practice was not precisely measured but was of the order of 50.

Various alternative situations can be worked out theoretically, and a number of types of arrangements to take advantage of the possibilities indicated above for increasing sensitivity can be rather easily seen. One such would be to insert paramagnetic material, for which a resonance is to be detected, into a microwave cavity at very low temperature and also containing ruby. The ruby would be "pumped" so that for it $a > b$, and for the entire cavity $a$ approaches $b$ until the system is approximately at the point of oscillation and the noise output is greater than that produced by any following amplifier. The desired resonance should be then detectable by sweeping it through the center of response of the cavity and excited ruby.

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[1] C. H. Townes and A. L. Schawlow, Microwave Spectroscopy (McGraw-Hill Book Company, New York, 1955), p. 414.

[2] K. Shimoda, H. Takahasi, and C. H. Townes, J. Phys. Soc. Japan 12, 686 (1957).

[3] See reference 1, p. 437.

[4] K. Shimoda and T. C. Wang, Rev. Sci. Instr. 26, 1148 (1955).

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# FLUCTUATIONS OF NUCLEAR CROSS SECTIONS IN THE "CONTINUUM" REGION*

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The assumption that the matrix elements of the compound nucleus are randomly distributed with respect to phase and magnitude has in recent years been very successful for the understanding of the fluctuations of the partial widths and the distribution of the level spacings of slow-neutron resonances.[1-3]

The purpose of this Letter is to examine the nontrivial consequences of the statistical assumption in the so-called "continuum" region, in which a large number of compound states overlap owing to the short lifetime of the compound nucleus. It is generally concluded that the excitation of a large number of intermediate compound states automatically implies that cross sections are smooth functions of the energy and that angular distributions are symmetrical around 90° to the beam direction. It will be shown that the proper statistical prediction for experiments performed with good energy resolution in the incident beam is that (1) partial cross sections and angular distributions fluctuate even though a large number of intermediate states are excited, (2) the formation and decay of the compound nucleus are independent only on the average, and (3) the fluctuations can be used to determine the lifetime of the compound nucleus in the "continuum" region.

Consider the compound-nucleus reaction proceeding from the initial state $|\alpha\rangle-$, i.e., the target nucleus and the incident wave—to the final state $|\alpha'\rangle$, a particular state of the final nucleus and the corresponding emitted wave. We assume the experiment to be performed with infinitely good energy resolution in the incident beam. The scattering matrix $S_{\alpha\alpha'}$ can be divided into two parts, one, $S_\alpha$, leading into the compound nucleus, and one, $S_{\alpha'}$, leading out of the compound nucleus. The intermediate compound states $|i\rangle$ of energy $E_i$ are excited with probability amplitudes $f(E, E_i)$ which are approximately of the form

$$f(E, E_i) \propto \frac{1}{(E - E_i) + i\Gamma/2}. \tag{1}$$

The "width" $\Gamma$ depends in principle on the intermediate state $|i\rangle$. At high excitation this dependence will be weak, because $\Gamma$ will be a sum of a large number of partial widths, of which no single one dominates. This situation should be realized at some 3-4 Mev above neutron binding energy in heavy nuclei, but is not very well realized at lower excitation. As we discuss the case of high excitation, we will neglect the fluctuations in $\Gamma$ and take it to be a constant. The "width" $\Gamma$ is related to the lifetime $\tau$ of the compound nucleus by $\Gamma = \hbar/\tau$ by the uncertainty principle. Equation (1) expresses that the compound states within a region of the order of $\Gamma$ are excited simultaneously and must be treated coher-