The Ammonia Beam Maser as a Standard of Frequency*

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Introduction

The ammonia beam maser has presented certain problems when considered as a primary standard of frequency. These problems come about because the maser's frequency of oscillation is quite dependent upon a variety of parameters. Also, with several of these parameters there is no unambiguous way of selecting a particular value for the parameter. In the past these parameters have been difficult to control and caused undesirable drift rates.

Recently a servomechanism has been installed which has eliminated cavity-tuning effects;[1] *i.e.*, the cavity is continuously tuned to be at the resonant frequency of the molecule. The method will be shown later. The elimination of this parameter has also reduced the effects of the other parameters.

In the case of ammonia beam pressure and focusing voltage, there is no clear-cut way of selecting values in order to get the maser to oscillate at the "proper Bohr frequency."

In part, some of the trouble arises from the fact that the quadrupole moment of N^{14} in ordinary ammonia causes an asymmetric splitting of the $J=3$, $K=3$ inversion line. This trouble can be avoided by using $N^{15}H_3$ instead of the ordinary ammonia. The present experimental work has been confined to $N^{14}H_3$; however, as soon as a system capable of recirculating the ammonia is completed, data will be taken on $N^{15}H_3$. A considerable improvement in reproducibility when using $N^{15}H_3$ has been reported by others.[2]

With all of its problems, the ammonia beam maser using $N^{14}H_3$ has demonstrated a resettable frequency of better than $\pm 3\times 10^{-11}$.

Zeeman Modulation

It has been shown[3] that the cavity-pulling of the maser frequency is given approximately by

$$\nu_0 - \nu = \frac{\Delta\nu_l}{\Delta\nu_c}(\nu_0 - \nu_c) \tag{1}$$

where ν_0 is the natural resonance frequency of the ammonia molecule; ν is the frequency of oscillation of the maser; ν_c is the resonant frequency of the cavity; $\Delta\nu_l$ is the natural line width of the ammonia transition in the maser; and $\Delta\nu_c$ is the cavity bandwidth. It has been suggested by Shimoda, *et al.*[1,4] that a small magnetic field will cause a Zeeman splitting of the ammonia line and thus cause an effective broadening of the ammonia line width. If the cavity is tuned to a frequency other than ν_0, a frequency shift of the maser is observed with the application of the magnetic field.

Under the influence of a magnetic field, the line width is given approximately by (see Appendix)

$$\Delta\nu_l = \sqrt{(\Delta\nu_0)^2 + (\Delta\nu_H)^2} \tag{2}$$

where $\Delta\nu_0$ is the unperturbed line width for the ammonia transition and $\Delta\nu_H$ is the amount the lines are split by the magnetic field. (See Fig. 1.) It has been found[5] that $\Delta\nu_H$ is given by

$$\Delta\nu_H = \alpha H$$

where the constant α is approximately 718 cps/oersted.

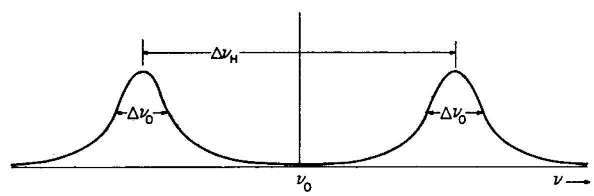

Fig. 1—Zeeman splitting of $J=3$, $K=3$ line of $N^{14}H_3$.

In order to obtain a continuous correction to the cavity tuning, the authors have found it desirable to apply a sinusoidally varying magnetic field to the maser; *i.e.*,

$$H = H_0 \sin \omega_m t.$$

Thus the line width will be given by

$$\Delta\nu_l = \sqrt{(\Delta\nu_0)^2 + (\alpha H_0)^2 \sin^2 \omega_m t}$$
$$= \sqrt{(\Delta\nu_0)^2 + \tfrac{1}{2}(\alpha H_0)^2 - \tfrac{1}{2}(\alpha H_0{}^2) \cos 2\omega_m t}.$$

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[1] J. C. Helmer, "Maser oscillators," *J. Appl. Phys.*, vol. 28, pp. 212–215; February, 1957.

[2] J. De Prins and P. Kartashoff, "Applications of Hertzian Spectroscopy to the Measurement of Frequency and Time," publication of Laboratoire suisse de recherches horlogeres, Neuchatel, Switzerland. (Topics on Radiofrequency Spectroscopy, August 1–17, 1960, International School of Physics, "Enrico Fermi," Varenna—Villa Monastero.)

[3] The reader is referred to J. P. Gordon, H. J. Zeiger, and C. H. Townes, "The maser," *Phys. Rev.*, vol. 99, pp. 1264–1274; August, 1955. While this is an approximate formula, it is sufficient to indicate the functional dependence of the maser frequency. Also a more elaborate theory[1] still suggests that $\partial\nu/\partial H=0$ determines the natural resonant frequency which, incidentally, differs from other possible means of obtaining this frequency.

[4] K. Shimoda, T. C. Wang, and C. H. Townes, "Further aspects of the maser theory," *Phys. Rev.*, vol. 102, pp. 1308–1321; June, 1956.

[5] C. K. Jen, "Zeeman effect in microwave spectra," *Phys. Rev.*, vol. 74, pp. 1396–1406; November, 1948.

Applying the binomial theorem one obtains

$$\Delta\nu_l \approx \Delta\nu_0 \sqrt{1+\frac{x^2}{2}} - \tfrac{1}{2}\Delta\nu_0 \sqrt{1+\frac{x^2}{2}}\left(\frac{x^2}{2+x^2}\right)\cos 2\omega_m t + \cdots \quad (3)$$

where $x \equiv \alpha H_0/\Delta\nu_0$ and the approximation is valid to the degree that

$$\frac{x^2}{2+x^2} \ll 1.$$

Substitution of (3) into (1) gives

$$\nu = \nu_0 - \frac{\Delta\nu_0}{\Delta\nu_c}\sqrt{1+\frac{x^2}{2}}\,(\nu_0-\nu_c) + \frac{\Delta\nu_0}{\Delta\nu_c}\,\frac{1}{2\sqrt{2}}\,\frac{x^2}{\sqrt{2+x^2}}\,(\nu_0-\nu_c)\cos 2\omega_m t. \quad (4)$$

There are two things of interest in (4); first, the frequency of the modulation term is twice the frequency of the Zeeman field, as one would expect and, second, this modulation term is proportional to $\nu_0-\nu_c$. It is this latter property which enables one to simply construct a servosystem that will continually control the cavity tuning.

The authors have found that the most convenient and most noise-free method of demodulation is that of phase demodulation. The block diagram of the system used is shown in Fig. 2 and the equivalent servodiagram of the maser-crystal-oscillator phase-lock system is shown in Fig. 3. In Fig. 3 the transfer functions of the various components are indicated below the components.

From the theory of servosystems, the total transfer function for Fig. 3 is given by

$$\frac{\phi_0(\omega)}{\phi_m(\omega)} = \frac{K_1K_2K_3}{1+K_1K_2K_3}\cdot \quad (5)$$

From this it follows that

$$\alpha(\phi_m(\omega)-\phi_0(\omega)) = V_1 = \alpha\phi_m(\omega)\left(1-\frac{K_1K_2K_3}{1+K_1K_2K_3}\right) = \alpha\phi_m(\omega)\left(\frac{1}{1+K_1K_2K_3}\right). \quad (6)$$

Substitution of the transfer functions into (6) yields

$$V_1(\omega) = \alpha\phi_m(\omega)\left[\frac{\omega^2\tau - j\omega}{\omega^2\tau+\alpha\beta-j\omega}\right]. \quad (7)$$

It is apparent that the bracketed expression in (7) has its maximum value when $\omega^2\tau \gg \alpha\beta$ which makes (7) have the value

$$V_1(\omega) \approx \alpha\phi_m(\omega). \quad (8)$$

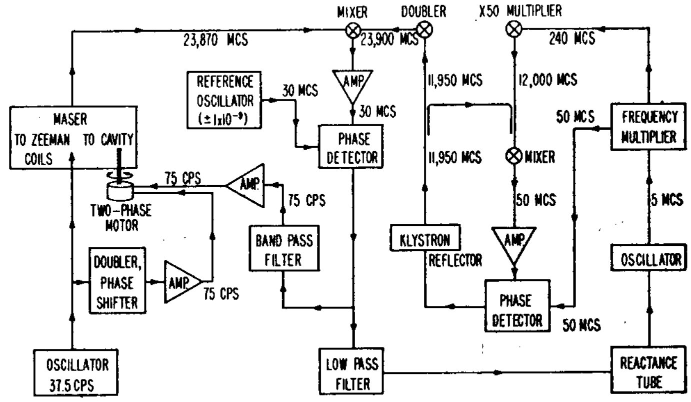

Fig. 2—Block diagram of complete system.

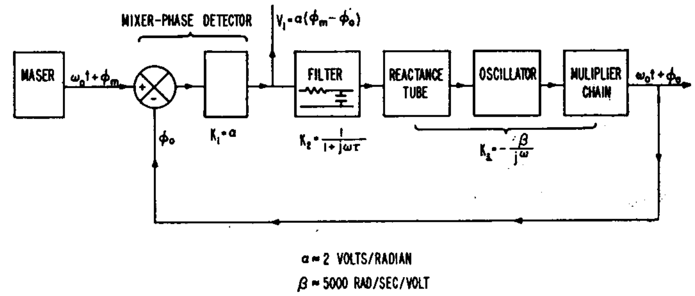

Fig. 3—Equivalent servodiagram for oscillator-maser phase-lock system.

As is customary in dealing with transfer functions, the quantities $\phi_m(\omega)$, $\phi_0(\omega)$ and $V_1(\omega)$ are the Fourier transforms of the time-dependent functions. Thus the voltage $V_1(t)$ can be obtained by taking the inverse Fourier transform of (8)

$$V_1(t) = \alpha\phi_m(t), \quad (9)$$

since α is a constant, and provided all important frequency components satisfy the relation $\omega^2\tau \gg \alpha\beta$.

Returning to (4), which can be written in terms of the total phase $\Phi(t)$,

$$\Phi(t) = \omega t + \phi_m(t) = 2\pi\left[\nu_0 - \frac{\Delta\nu_0}{\Delta\nu_c}\sqrt{1+\frac{x^2}{2}}\,(\nu_0-\nu_c)\right]t + \frac{\Delta\nu_0}{\Delta\nu_c}\,\frac{1}{2\sqrt{2}}\,\frac{x^2}{\sqrt{2+x^2}}\left(\frac{\nu_0-\nu_c}{2\nu_m}\right)\sin 2\omega_m t. \quad (10)$$

Comparison of (9) and (10) gives

$$V_1(t) = \gamma(\nu_0-\nu_c)\sin 2\omega_m t \quad (11)$$

where γ is the constant

$$\gamma = \left(\frac{\alpha}{4\sqrt{2}\nu_m}\right)\left(\frac{\Delta\nu_0}{\Delta\nu_c}\right)\left(\frac{x^2}{\sqrt{x^2+2}}\right) \quad (12)$$

and $(2\omega_m)^2\tau \gg \alpha\beta$.

Eq. (11), then, represents the demodulated signal from the maser. It should be noted here that the phase-locked crystal oscillator's frequency is determined by the maser frequency and thus this servosystem serves a double purpose: 1) as a low-noise phase demodulator, and 2) as a precise frequency divider to facilitate comparison with other systems, *e.g.*, the cesium beam.

For the NBS maser system, γ has the value of approximately 3.5 mv per part in 10^{10} of $N^{14}H_3$ maser frequency. While this seems like a sufficiently sensitive detection system, it must be remarked that there still exist significant noise sources which will tend to limit the precision of balance. The most important of these sources is the crystal oscillator used in the maser phase-lock system. Any phase jitter of this oscillator signal after multiplication to K band will be detected as noise in the phase demodulator. It is interesting to note that if the maser is detuned one part in 10^{10}, the peak phase modulation on the maser is only about 0.14° at K band!

The oscillator which is used in the NBS maser system has demonstrated the most nearly pure spectrum of any oscillator analyzed to date.[6] In fact, except for a very small white-noise pedestal, it is difficult to be sure whether the maser or this oscillator has the more nearly monochromatic signal. Although some noise may be eliminated by narrow banding the demodulator, this process cannot be carried too far or the Nyquist conditions for stability of the servosystem will be violated.

The servoloop for the cavity tuning is completed by applying the amplified signal from the phase demodulator to one phase of a two-phase motor which in turn controls the depth of a small plunger in the maser's resonant cavity. The reference for the other phase of the motor is obtained by doubling the frequency of part of the signal from the Zeeman modulating field supply.

Due to inherent noise in the system, it has been found that a better time-averaged frequency is obtained from the maser if the gain is advanced to the point where small oscillations about the null modulation point just begin. Under these conditions, the servomotor is oscillating back and forth at a rate of about 2 cps, and at an amplitude of about ± 5 parts in 10^{10} in terms of maser frequency. An 8-sec average of oscillator frequency shows a standard deviation of about 3 parts in 10^{11}.

Returning to the condition on (11) that

$$(2\omega_m)^2\tau \gg \alpha\beta,$$

it is possible to determine the minimum time constant of the phase-lock filter. For the NBS system, the product $(\alpha\beta)$ is about 10^4 sec^{-1} and thus $\tau \gg 0.05$ sec. Typically, τ is chosen to be about 0.5 sec to 1.0 sec. Since this time constant must be taken this long, again the stability of the crystal oscillator must be very good or deterioration of the maser stability will result.

[6] J. A. Barnes and L. E. Heim, "A high-resolution ammonia-maser-spectrum analyzer," IRE TRANS. ON INSTRUMENTATION, vol. I-10, pp. 4–8; June, 1961.

Basic Measuring Scheme

Quite an elaborate system is involved in detecting and dividing down the maser frequency. It is the purpose of this section to outline this system. See Fig. 2.

A 5-Mc crystal oscillator drives a multiplier chain. From this chain, 240 Mc is fed into a crystal multiplier; the 50th harmonic (12,000 Mc) of this and an 11,950-Mc signal from a klystron are fed into a crystal mixer. The 50-Mc beat note resulting is sent through an IF amplifier and then phase compared with 50 Mc from the multiplier chain. The phase difference is used as an error voltage to correct the klystron's frequency; hence, the klystron is phase locked to the 5-Mc crystal oscillator.

The klystron signal is sent to a crystal doubler giving 23,900 Mc. This signal goes in one side of a balanced crystal mixer. The maser's signal at approximately 23,870 Mc provides the reference into the other side of the above mixer. The 30-Mc beat note resulting goes to an IF amplifier and then to a phase detector. A frequency synthesizer, stable to better than a part in 10^8, is used as the reference into the other side of the phase detector. (One part in 10^8 at this reference gives a stability of about 1 part in 10^{11} at maser frequency.) The phase error resulting is then sent to a reactance tube which in turn corrects the frequency of the above-mentioned 5-Mc crystal oscillator. Therefore, the 5-Mc signal is phase locked to the maser's frequency, and hence the maser's stability can be analyzed by looking at the 5-Mc crystal oscillator or any multiple of it as derived from the multiplier chain.

Experimental Results

Maser Frequency Dependence on Operating Conditions

It has been the intent of the authors to find most, if not all, parameters that give instability and that cause frequency shifts; also, the control of critical parameters has been of concern. Some of the frequency-dependent parameters are beam pressure, electrode focusing voltage, Zeeman modulation voltage, fluctuations in the magnetic field of the earth, background pressure in the maser's vacuum system, alignment of beam nozzles and electrodes with respect to the resonant cavity, temperature effects, and a few other influences, most of which are quite minor.

In connection with the beam pressure a variety of nozzles have been tried including klystron grid material, crimped foil, and special drilled single-hole nozzles. Most of the data have been taken with 0.02-inch single-hole nozzles; this is by no means the optimal nozzle to use, and this nozzle is used mainly for symmetry reasons in the NBS double-beam maser. Beam pressures typically used are in the vicinity of 6 mm of mercury; such a pressure gives a molecular mean free path (λ) of 8×10^{-4} cm. Since λ is smaller than the nozzle hole diameter, this disallows a Maxwellian velocity distribution, but rather gives cloud diffusion which has a radial molecular intensity distribution. This system gives very uniform flow as can be illustrated from the frequency-time statistics, but quite a large consumption of ammonia (about 1 gr/hr).

Data were taken to determine the optimal beam pressure to use for frequency stability, and a typical family of curves is shown in Fig. 4. In another experiment the beam pressure was run up to about 14 mm of mercury to observe the continuation of the curves shown. It was found that the curves continued to approach each other and the second derivative became negative. This change in slope is attributed to a significant increase in the background pressure caused by the high flux of molecules.

By analyzing Fig. 4 one can quite easily observe the behavior of the focusing voltage V_F vs maser frequency since there is a 540-v difference between each of the curves in the family of curves plotted. The slope of maser frequency as a function of focusing voltage is positive with a positive second derivative also; the magnitude of the slope is in the vicinity of 1 part in 10^{10} of maser frequency per 200 v. This is rather severe for frequency stability, and the voltage has to be read to about 0.2 per cent in order to get resettability to 1 part in 10^{11}. This, of course, puts stringent requirements on the high-voltage power supply. One can further observe by analyzing Fig. 4 that the slope of frequency vs focusing voltage decreases for increasing beam pressures.

Another curve of interest is maser frequency vs the Zeeman frequency modulation voltage (see Fig. 5). If the Zeeman splitting coefficients are linear (the theory states they are to a first approximation—second-order effects are of the order[4] of 2×10^{-15} H^2—for all molecular frequency components within the range of the resonant cavity) then the slope of frequency vs modulation voltage should be zero. But the contrary result is that the curve has a negative slope of about 5 parts in 10^9 at maser frequency per oersted (rms); 2 volts rms applied to the magnetic-field coils of the NBS maser corresponds very closely to 1 oersted (rms) in the cavity. A similar effect has been observed on $N^{15}H_3$.[2]

With a slope as previously stated for maser frequency vs Zeeman modulation voltage, one might wonder if fluctuations in the earth's magnetic field would cause frequency shifts. From experiments performed it was found that the slope of this curve is essentially the same as for an ac field. Since the earth's magnetic field is about 0.56 oersted, a net shift of the maser's frequency of about 3 parts in 10^9 would be expected. Also, it was predicted that during fairly severe magnetic storms a frequency shift in the vicinity of 1 part in 10^{11} could be observed; an experiment was performed and an actual correlation was shown to exist. Since the shift due to magnetic storms is so small, it could be easily eliminated with a μ-metal shield around the maser's resonant cavity.

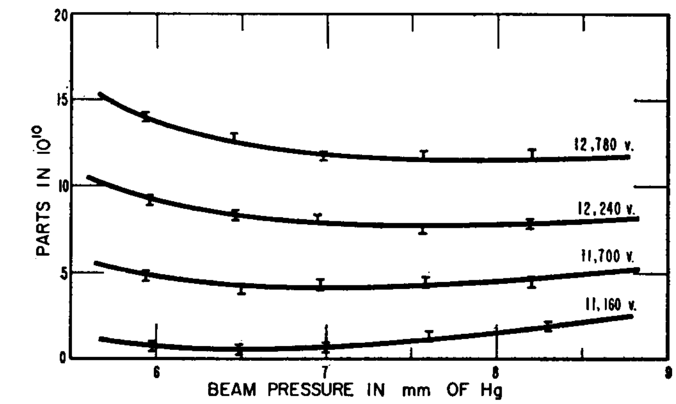

Fig. 4—Relative shift in maser frequency vs beam pressure.

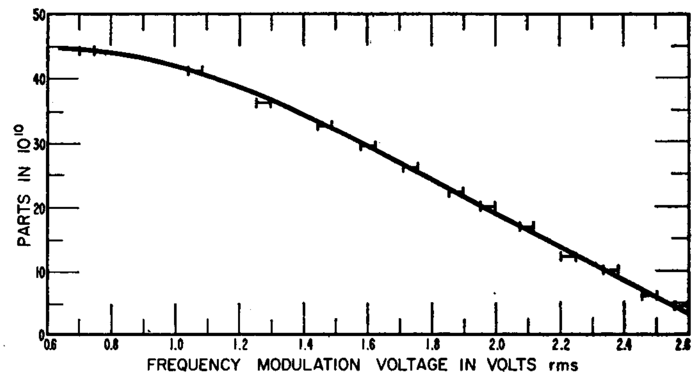

Fig. 5—Relative maser frequency vs modulation voltage applied to magnetic field coils.

Prior to the installation of the cavity-tuning servo-loop, background pressure changes caused sizeable frequency shifts; for example, when the liquid-nitrogen cold traps on the maser's vacuum system were filled, of course the background pressure would change, and frequency shifts as high as 1 part in 10^9 were observed. Since the installation of the above-mentioned servo-loop, cold trap filling and fairly large changes in the background pressure cause no measurable frequency shifts. Pressures typically used in the maser's vacuum system are 2×10^{-6} mm of mercury; no frequency shift has been observed up to pressures of 6×10^{-6} mm. Higher frequency shifts than this have been indicated though very marginal up to 1×10^{-5} mm.

The NBS maser has been taken apart and reassembled several times; each time a frequency shift has occurred along with changes in the parametric curves of Figs. 4 and 5, although the basic character of the curves has not changed. If pains are taken to duplicate alignment and configuration of nozzles, focusers, and cavity, the shift can be kept within 3 parts in 10^{10}—showing the critical nature alignment plays in the maser as a frequency standard. However, for any one alignment and configuration the stability for short term and long term is shown in Figs. 6 and 7. Such stability brings the maser to the status of a very good secondary standard.

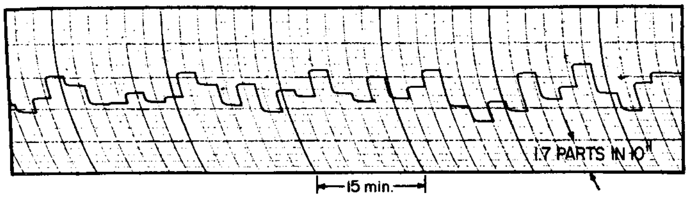

Fig. 6—Frequency of maser as a function of time. Each step represents an average frequency over an interval of approximately 78 sec. The record is $1\frac{1}{2}$ hr long.

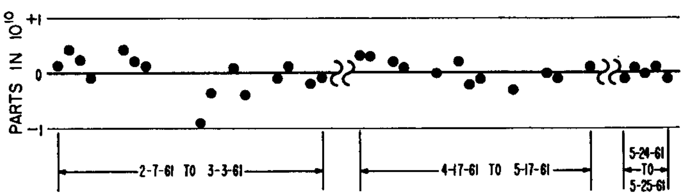

Fig. 7—NBS double-beam ammonia maser vs cesium beam (National Frequency Standard).

Maser Stability

In order to test the long- and short-term stability of the maser, comparisons have been made with a very stable (drift rate of about 6×10^{-12}/hr) quartz crystal oscillator and with the National Frequency Standard, cesium beam.

For short-term comparisons the oscillator that is phase locked to the maser is mixed with the oscillator mentioned above, and the period of the beat note is measured. Eight-second averages gave a standard deviation of 3 parts in 10^{11}; a large part of this is due to the oscillating cavity-tuning servoloop because the short-term stability of the free-running maser is only a few parts in 10^{12}.

The reader will remember that the period of oscillation of the cavity-tuning servoloop is about $\frac{1}{2}$ sec; so if the time of averaging is long compared with the period of oscillation, the statistics should average out this oscillatory characteristic. Therefore, 78-sec period averages were taken, and a typical trace of the relative frequency fluctuations is shown in Fig. 6. This trace is 96 min long and gives a standard deviation of the mean of 1.3 parts in 10^{12}.

The long-term stability of the maser has been obtained by direct comparisons with the cesium beam. A plot of the day-by-day comparisons is shown in Fig. 7. Note that there are three sections; each one represents a new alignment and hence a different frequency (not indicated); the mean of each set is plotted on the same axis. Since the cesium beam usually has a standard deviation of the mean of less than 1 part in 10^{11} for any one measurement and the standard deviation for the plot in Fig. 7 is 3 parts in 10^{11}, one concludes that there is an unknown parameter in the maser system which is not in statistical control. It is felt at present that this is probably temperature-dependent elements in the maser system.

Conclusion

The experimental results show that an ammonia beam maser using the $J=3$, $K=3$ transition in ordinary $N^{14}H_3$ can be a very reliable secondary standard of frequency. As long as the alignment of the maser is not disturbed, its frequency is resettable to a precision which makes it quite competitive with the cesium beam and gas cell. The authors see no reason why such a maser system could not be run for years without deterioration of its resettability. With an improved servosystem, the short-term stability could probably approach that of the free-running maser itself and thus have this advantage over either the cesium beam or gas cell which depends on a quartz crystal oscillator for their short-term stability.

Perhaps one of the most encouraging aspects of the system is that everyone who has used $N^{15}H_3$ has reported a marked improvement over $N^{14}H_3$ in all operating parameters as theory predicts. It is hoped that the change to $N^{15}H_3$ may relegate this maser servosystem to a competitive primary standard of frequency.

Appendix

When a magnetic field is applied to the ammonia molecule, the spectral line for $J=3$, $K=3$ is split as shown in Fig. 1. Such a splitting, of course, changes the effective line width $\Delta\nu_l$ of the transition. Since this effective line width must be a function of the magnitude of the splitting only, it is reasonable, at least as far as functional dependence is concerned, to assume that the effective line width is related to the second moment of the perturbed line by the relation

$$\left(\frac{\Delta\nu_l}{2}\right)^2 = \frac{\int P(\nu)(\nu-\nu_0)^2 d\nu}{\int P(\nu) d\nu} \tag{13}$$

where $P(\nu)$ is the probability of transition for the frequency ν, and ν_0 is the center of gravity of the line.

By a simple application of the parallel axis theorem one obtains

$$\left(\frac{\Delta\nu_l}{2}\right)^2 = \left(\frac{\Delta\nu_0}{2}\right)^2 + \left(\frac{\Delta\nu_H}{2}\right)^2 \tag{14}$$

or equivalently

$$\Delta\nu_l = \sqrt{(\Delta\nu_0)^2 + (\Delta\nu_H)^2}. \tag{15}$$

Acknowledgment

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