Coherent Spontaneous Microwave Emission by Pulsed Resonance Excitation*

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Summary—**This paper describes an investigation of the coherent microwave emission from pulse-excited ammonia molecules. Coherent and periodic pulses of near resonance frequency and 1-μsec duration excited the gas from its initial thermal equilibrium condition. Self-induced coherent emission (molecular ringing) continued after the excitation field was removed. This radiation was observed during a period of 10 μsec. In an actual experiment performed, a new Doppler bandwidth reduction method was used in the gas cell. The observed spectral width of the ammonia 7,7 line was about 5 kc. The emission was used to stabilize the excitation signal source to a short-term frequency stability of 2×10^{-10}.**

DURING the period 1952–1955, the author was concerned with an investigation of a new way to reduce the Doppler contribution to total microwave spectral line bandwidth.[1] The basic concept on which the Doppler reduction aspect hinged was due to a proposal by Dicke. In one of the Doppler reduction experiments which were performed, the signal was obtained as a small, nonsustained self-induced coherent microwave emission (frequently termed spontaneous coherent emission, or molecular ringing) from the ammonia gas. This emission was induced by coherently pulsed resonance excitation of the gas molecules. In view of the present interest in spontaneous emission and its relation to molecular and atomic oscillators and amplifiers, it appears desirable to describe these early experiments and results which have not hitherto been published. The difference between the described resonance-radiation excited nonthermal equilibrium condition and the beam deflection state separation utilized by Townes and Gordon[2] in the now well-known Maser is quite apparent. A different but related system has also been reported by Hahn,[3] and Hahn and Maxwell,[4] who used pulse-induced spin flipping in their pulse echo work. While this paper was in final stages of preparation still another publication,[5] with a vastly different point of view, made its appearance.

One of the experimental arrangements is shown in Fig. 1. The Doppler bandwidth reduction details[1] are omitted in this figure. Detailed operation of Fig. 1 is described later. For the moment, the important detail is that one of the repetitively pulsed sidebands provided microwave resonance excitation for the two quantum levels involved. Additionally, the excitation frequency was swept slowly and cyclically to record either the envelope or the dispersion of the line.

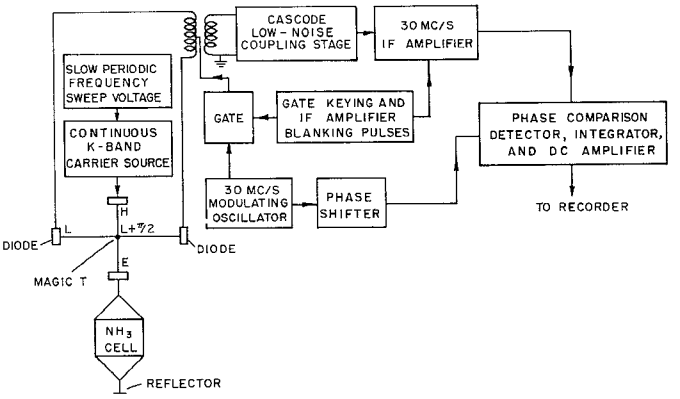

Fig. 1—Block diagram of simplified system for obtaining envelope and dispersion of spectral lines by spontaneous coherent emission.

The basic operation of Fig. 1 is shown in Fig. 2. The primary excitation field E_D was applied at $t=0$ and removed at $t=t_0$. The receiver was actuated from $t=t_0$ to $t=t_1$ so that the signal, emitted from the ringing molecules, could be obtained during this interval. The excitation was applied again and the receiver disabled at $t=t_1$ to initiate a new cycle. Solutions to this problem are contained implicitly in a paper by Dicke[6] and more specifically in a forthcoming theoretical paper by Bloom.[7] However, for the microwave region of the spectrum, and under experimental conditions which permit omission of all collision effects, it is possible to go directly, and with good approximation, to expressions for self-induced coherent emission after removal of primary drive.

A particle with unperturbed normal Hamiltonian H_0 is subjected suddenly to an excitation field which adds perturbation energy

$$V(x, t) = -eE_D x \sin \omega t. \tag{1}$$

The electric excitation field E_D is at frequency ω and its electric vector is directed along x.

Considering only two nondegenerate states at energy levels W_1, W_2, the time-dependent probability amplitudes may be put in the form

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[1] "Investigation and Study of Practical Utilization of Molecular Absorption for Frequency Control," Contract No. DA 36-039-sc-15525, Dept. of the Army Project No. 3-99-11-021, Signal Corps Project No. 33-142B. Reps. No. 1–No. 10 for period May 1, 1952 to April 22, 1955.
[2] J. P. Gordon, H. J. Zeiger, and C. H. Townes, *Phys. Rev.*, vol. 99, pp. 1264–1274; August 15, 1955.
[3] E. L. Hahn, *Phys. Rev.*, vol. 80, pp. 580–594; November 15, 1950.
[4] E. L. Hahn and Maxwell, *Phys. Rev.*, vol. 88, pp. 1070–1084; December 1, 1952.
[5] R. H. Dicke and R. H. Romer, "Pulse techniques in microwave spectroscopy," *Rev. Sci. Inst.*, vol. 26, pp. 915–928; October, 1955.
[6] R. H. Dicke, *Phys. Rev.*, vol. 93, pp. 99–110; January 1, 1954.
[7] S. Bloom, *J. Appl. Phys.*, vol. 27, pp. 785–788; July, 1956.

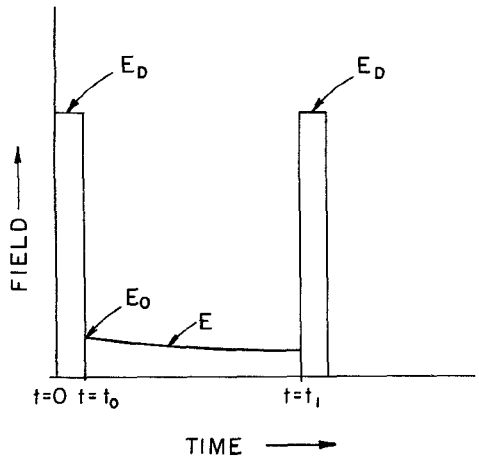

Fig. 2—Time application of coherently pulsed excitation to molecular system, and received spontaneous coherent emission. E_D is primary drive field, and E is time dependent spontaneous coherent emission field with initial value $E_0 \cdot dE/dt$ does not change sign in the particular illustration plotted.

$$\left.\begin{aligned} i\dot{a}_1 &= -\sin \omega t \quad (p_{11}a_1 + p_{12}a_2\epsilon^{-i\omega_0 t}) \\ i\dot{a}_2 &= -\sin \omega t \quad (p_{12}^{*}a_1\epsilon^{i\omega_0 t} + p_{22}a_2) \end{aligned}\right\} \tag{2}$$

where

$$p_{ij} = E_D\mu_{ij}/\hbar, \quad \text{and} \quad \omega_0 = (W_2 - W_1)\hbar.$$

The probabilities of state occupancy under initial thermal equilibrium conditions (before application of excitation) for $\hbar\omega_0/kT \ll 1$, are

$$\left.\begin{aligned} N_1(0)/N &= 1/2 + \hbar\omega_0/4kT \\ N_2(0)/N &= 1/2 - \hbar\omega_0/4kT \end{aligned}\right\} \tag{3}$$

where $N_1(0)$, $N_2(0)$ are the thermal equilibrium numbers of particles in states 1 and 2 in a system of N particles.

First, consider the case where, to the time scale of interest, particle density is chosen sufficiently small and particle container dimensions sufficiently large so that collision processes are relatively unimportant. Upon application of resonance excitation at frequency $\omega = \omega_0$ at time $t = 0$, neglecting incoherent spontaneous drop-downs in this microwave region of the spectrum, from (3) and a solution of (2), the probability of occupancy of the upper state after $t = 0$ is

$$N_2(t)/N = 1/2 - \hbar\omega_0/4kT \cdot \cos\left[(\mu E_D/\hbar)t\right] \tag{4}$$

where μ is the effective dipole moment for the transition involved.

The power input to the system of N particles just before excitation shutoff at $t = t_0$ is

$$\begin{aligned} P_{\text{in}} &= \hbar\omega_0 N \cdot d/dt[N_2(t)/N] \\ &= m\omega_0^2\mu E_D N/4kT \cdot \sin\left[(\mu E_D/\hbar)t_0\right]. \end{aligned} \tag{5}$$

Similarly, the initial power emission from the system just after excitation removal at $t = t_0$ is

$$P_0 = \hbar\omega_0^2\mu E_0 N/4kT \cdot \sin\left[(\mu E_D/\hbar)t_0\right] \tag{6}$$

where E_0 is the field initially emitted.

The value of E_0 may be obtained by noting that, after excitation shutoff, E_0 is the field in the transmission path containing the particles undergoing transitions. Hence, neglecting any field dependence as a function of position in the transmission path,

$$cE_0^2A/4\pi = \hbar\omega_0^2\mu E_0 N/4kT \cdot \sin\left[(\mu E_D/\hbar)t_0\right]$$

and

$$E_0 = \pi\hbar\omega_0^2\mu N/ckTA \cdot \sin\left[(\mu E_D/\hbar)t_0\right] \tag{7}$$

where A is the cross sectional area of the transmission path containing the molecules.

Finally, from (6) and (7), the initial coherent power radiated by the particles at excitation shut-off is

$$P_0 = \pi(\hbar\omega_0^2\mu/2kT)^2 \cdot N^2/cA \cdot \sin^2\left[(\mu E_D/\hbar)t_0\right]. \tag{8}$$

Excitation periods given by

$$\mu E_D/\hbar \cdot t_0 = (2n + 1)\pi/2 \tag{9}$$

provide state saturation conditions (equality of state populations) and produce maximum initial coherent emission; those given by

$$\mu E_D/\hbar \cdot t_0 = (2n + 1)\pi \tag{10}$$

define "inverted" states in which the sense of population inequality is appropriate for a negative interpretation of T in the Boltzmann factor exponential.

Under conditions where collision processes may be neglected, the same simple model may be used to evaluate with good accuracy the change of coherent emission P with time from its initial value P_0. During primary excitation, transitions are induced by drive of constant amplitude; after removal of primary drive the situation is similar except that the self-induced driving field intensity changes with time. In practical cases the change is very slow on the time scale of interest, for the following interrelated reasons:

1) Available coherently pulsed microwave driving ing power is small.
2) To keep the effect of competing collision processes small, especially for electric dipole transitions, pressures must be low; even so, corresponding reasonable drive intervals are of the order of a microsecond, with following observation intervals about ten times longer.
3) Since coherent emission intensity after primary excitation removal goes as N^2, increase in A is desirable but this in turn reduces the field due to the small driving power. Without accompanying increase in the excitation power, the improvement factor is only slightly greater than unity.
4) At microwave frequencies $\hbar\omega_0/kT < 1$. Under these circumstances, the ratio of self-induced driving field to primary field is ordinarily very small. As a result, intensity changes in the self-induced driving field during the limited observation period (usually 10^{-5} seconds, or less) are very small. For this case, from (6) and by extending (6) to time after primary drive removal, we write immediately

$$P/P_0 = (E/E_0)^2$$

$$= E/E_0 \sin [(\mu E_D/\hbar \cdot t_0) - \mu E/\hbar \cdot t]/\sin [(\mu E_D/\hbar)t_0]$$

or

$$E/E_0 = \sin (\alpha - E/E_0 \cdot \mu E_0/\hbar \cdot t)/\sin \alpha \qquad (11)$$

where $\alpha = \mu E_D/\hbar \cdot t_0$, E is the self-emissive field after $t = t_0$, and P is the corresponding power.

Because E_0/E_D is so small, the departure and eventual decay with time of E from E_0, in any reasonable experimental circumstances, inevitably requires a much longer time than the primary excitation interval. Hence, eventually other competing processes (collisions, for example), which are not a part of this simple model, must be taken into account. The same slow decay also indicates that when the next coherent pulse is applied at $t = t_1$, there may still be appreciable energy storage in the particles. Results, taking into account thermal collisions, are included in the solution of Bloom.[7] In general, the effect of thermal collisions is to superpose an exponential time decay on the coherent emission.

Since reduction of Doppler contribution to spectral line bandwidth was the objective of the experiment as performed, the spectral envelopes and dispersion characteristics were displayed as functions of slowly varying excitation frequency near resonance. Hence, the time decay characteristics of the initial spontaneous coherent emission (P_0, E_0) were never observed directly.

However, the foregoing results are still directly applicable to the slowly-varying excitation frequency case. Specifically, when the excitation and resonance frequencies are identical, the initial coherent emissive power is P_0. The situation for small departures from the resonance frequency is interpreted as follows. The product terms $\epsilon^{-i\omega_0 t} \sin \omega t$, and similarly $\epsilon^{i\omega_0 t} \sin \omega t$, appear in the probability amplitude espressions of (2). Expansion of the product terms lead to sum and difference frequencies $(\omega+\omega_0)$ and $(\omega-\omega_0)$. Upon integration, the corresponding terms are $\sin (\omega+\omega_0)t \cdot 1/(\omega+\omega_0)$ and $\sin (\omega-\omega_0)t \cdot 1/(\omega-\omega_0)$. The sum frequency term may be neglected since it has little influence on the magnitudes of the time dependent probability amplitudes. The difference term $\sin (\omega-\omega_0)t \cdot 1/(\omega-\omega_0)$ in the probability amplitudes produces maxima at resonance, $\omega = \omega_0$.

Typical experimental recordings show that the results predicted by the foregoing, indeed, do have physical reality. The dispersion characteristic of the NH_3 7,7 line as a function of slowly varying excitation frequency is shown in Fig. 3. Itwas obtained as a spontaneous coherent emission in the periods $t = t_0$ to $t = t_1$ by the use of the coherently pulsed resonance excitation system of Fig. 1. The corresponding envelope appears in Fig. 4. Time intervals $t = 0$ to $t = t_0$, and $t = t_0$, to $t = t_1$ in Fig. 2 were 10^{-6} and 10^{-5} seconds, respectively, in the experiment as performed. The Doppler reduction system was designed for a spectral line bandwidth of 5 kc and was achieved by reduction of Doppler bandwidth from its normal value

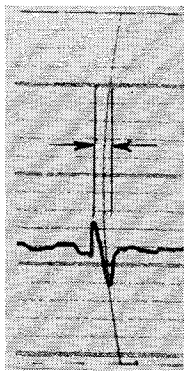

Fig. 3—5-kc bandwidth dispersion of NH_3 7,7 spectral line.

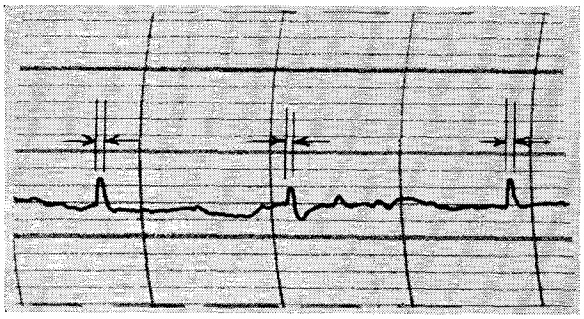

Fig. 4—5-kc bandwidth envelope of NH_3 7,7 spectral line.

of about 70 kc for ammonia at 25,000 mc and room temperature. For observation of a 5-kc-bandwidth line, bandwidth contributions due to particle-particle thermal collisions, and particle-cell wall thermal collisions, must be of the order of 5 kc or less. To achieve this condition, the ammonia pressure was about 1.85×10^{-4} mm Hg; the required 10-cm minimum cell dimension was obtained by using a cube 10 cm on a side. The corresponding total number of ammonia molecules, N', was 7.5×10^{15}. However, because of the Doppler bandwidth reduction in the experiment as performed,[1] the "molecular effectiveness" was only about 6 per cent. That is, the intensity of the reduced bandwidth signal was as if only 6 per cent of the actual total number of molecules were contributing to it.

Because of the requirement for narrow spectral distribution, and coherence in its pulsing, the K-band excitation source (Fig. 1) became rather complex. It consisted of a periodically frequency-swept 13.2+mc crystal controlled oscillator, followed by many stages of frequency multiplication with an over-all multiplication factor of 1944. The output frequency was smaller by 30 mc than 25,715+mc, the resonance frequency of the $N^{14}H_3$ 7,7 line. One simplified form of several arrangements used is shown as a block diagram in Fig. 1. The gate-keying and IF-blanking pulser opens the gate in the 30-mc oscillator output circuit, and simultaneously disables the IF amplifier during intervals $t = 0$ to $t = t_0$. Hence, continuously supplied K-band carrier and gated 30-mc output are applied to the magic-T and associated diodes, which functions as a balanced modulator during this time. The upper sideband produced by the balanced

modulator served as cell excitation. In this arrangement, application of the gated 30-mc output to the center of the transformer winding avoids (by phase balancing) overloading and delayed blocking of the cascode low-noise coupling stage in the receiving channel.

The same gate-keying and IF-blanking pulser closes the 30-mc gate, and actuates the IF amplifier during receiving intervals $t=t_0$ to $t=t_1$. Hence, the sideband intensity (excitation field E_D) becomes zero and spontaneous coherent emission from the cell and continutinuously applied K-band carrier output impinge on the diodes attached to the magic-T. During these intervals this unit operates as a superheterodyne mixer, supplying output to the input transformer preceding the cascode low-noise coupling stage. The push-pull to single-ended connection of the transformer in conjunction with the $\pi/2$ differential path length in the similar arms of the magic-T, insures complete uncoupling of the cascode-input circuit and gate-input circuit; hence none of the small intensity signal output is wasted in the gate-output circuit. Reference phase from the 30-mc modulating oscillator and 30-mc signal phase are compared in the phase sensitive detector, yielding output signals of the types shown in Figs. 3 and 4. To attain a favorable signal-to-noise ratio with the small signal intensity available, the signal bandwidth imposed by the integration circuit following the phase sensitive detector was made about 0.1 cycle. This required that the frequency sweep rate of the cell excitation unit be very slow; it was usually made about 1/40 cps.

Because of the nature of the primary-excitation unit and of the components available when the experiment was performed, available peak drive in the 10^{-6} second excitation periods was small. It was in the range from 10^{-4} to 10^{-5} watts, and usually was nearer the latter. Neglecting field position dependency in the cell, by assuming the mean field to prevail throughout the cube, the corresponding E_D was about 2×10^{-2} volts/cm and $\mu E_D/\hbar\cdot t_0$ was only 8.6×10^{-2}. From (4), the departure from thermal equilibrium conditions and the energy storage in the molecular system by the small perturbing resonance excitation, were slight. Unavoidably, and due only to the inadequacy of the primary excitation intensity, the initial spontaneous coherent emission, P_0, as given in (8) was small. For $E_D=2\times10^{-2}$ volts/cm, P_0 was 4.4×10^{-18} watts. Taking into account the "molecular effectiveness" in the reduced Doppler bandwidth signal,[1] even if the initial emission were completely monochromatic, and neglecting collisions, the time required for the self-induced emissive power to fall to half its initial value would be about 2.8×10^{-2} seconds. This is about 2800 ties the 10^{-5} second listening period, and therefore the change in P from P_0 was insignificant in the listening period for E_D as indicated. For a receiver noise figure of 10 and the previously mentioned 0.1-cycle signal bandwidth due to the integration circuit following the phase sensitive detector, the room temperature signal-to-noise ratio was calculated to be about 1×10^3, at best. For a variety of reasons, including the space dependency of the field in the cell, and utilization of only some of the discrete Stark components, the actual signal-to-noise ratio was always less. Assuming a worsening of the ratio, due to these causes, of somewhat less than an order of magitude, the actual signal-to-noise values as observed in Figs. 3 and 4 appear quite reasonable.

By using the received signal as a reference frequency, and assuming that the primary excitation source is servocontrolled to 1/1000 of the 5-kc spectral bandwidth (a fairly severe but reasonable assumption), by long time-averaging servoaction, the frequency stability of the excitation source is 2×10^{-10}.

ACKNOWLEDGMENT

The author wishes to express appreciation to Dr. D. O. North of RCA Laboratories for many helpful and stimulating discussions relating to the theoretical aspects of this problem, and to G. W. Leck, also of RCA who built much of the required specialized equipment.

Corrections

Karle S. Packard, author of "Planar Transmission Lines—Part III," a correspondence which appeared on page 163 of the April, 1957 issue of these TRANSACTIONS, wishes to make the following correction to his paper.

The quantity $\sqrt{k}$ should be replaced by $\sqrt{K}$, where K is the relative dielectric constant.

Leonard Sweet, co-author with M. Sucher of the correspondence, "The Available Power of a Matched Generator from the Measured Load Power in the Presence of Small Dissipation and Mismatch of the Connecting Network," which appeared on pages 167–168 of the April, 1957 issue of these TRANSACTIONS, has brought the following correction to the attention of the editors.

Eq. (6) on page 168 should be

$$(1-|S_{22}|^2)^2-|S_{22}|^2-|S_{11}|^2+|S_{11}S_{22}|^2 \geq 2\ \mathrm{Re}\ S_{12}{}^{*2}S_{11}S_{22}.$$