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[3] This sample, as well as some of the others, exhibits hysteresis and anomalies in the T_1's measured, probably as a result of residual traces of O_2 adsorbed on the glass sample tubes. It is hoped that more reliable data can be obtained in experiments now under way with "getter" techniques to eliminate O_2. .See H. S. Sandhu, J. Lees, and M. Bloom, Can. J. Chem. 38, 493 (1960).

[4] N. F. Ramsey, Nuclear Moments (John Wiley & Sons, Inc., New York, 1953), p. 73.

[5] M. R. Baker and N. F. Ramsey, Bull. Am. Phys. Soc. 5, 344 (1960). See also M. R. Baker, H. M. Nelson, J. A. Leavitt, and N. F. Ramsey, Phys. Rev. 121, 807 (1961), p. 815, on which the implications for nuclear relaxation of the large c_F are discussed.

[6] A study was made of the proton and fluorine T_1 and T_2 in liquid HF at room temperature by I. Solomon and N. Bloembergen, J. Chem. Phys. 25, 261 (1956), and the results were interpreted without recourse to spin-rotation interaction. This is reasonable for pure HF, because in it the strong hydrogen bonding and resultant polymerization could quench the interaction. [See also the footnote in the paper on T_1 in liquid H_2: W. P. A. Hass, G. Seidel, and N. J. Poulis, Physica 26, 834 (1960)]. However, in aqueous solutions of HF they found the fluorine T_1 to be much shorter than the proton T_1 and attributed the differences to differential molecular association and chemical exchange effects. Another possibilty is that the water decreases the quenching of the spin-rotation interaction in the HF. The importance of the latter probably could be assayed by observing the temperature dependence of T_1 and T_2.

[7] The details can be developed in a manner similar to that described by I. Solomon, Phys. Rev. 99, 559 (1955); see also Solomon and Bloembergen, reference 6.

GENERAL ANALYSIS OF OPTICAL, INFRARED, AND MICROWAVE MASER OSCILLATOR EMISSION[*]

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(Received February 13, 1961)

In this paper we will generalize the equations governing coherent emission from quantum mechanical amplifiers (microwave, infrared, and optical masers) using either electric or magnetic dipole transitions. Earlier analyses by Gordon, Zeiger, and Townes,[1] Feynman, Vernon, and Hellworth,[2] and Wang and Singer,[3] are incorporated in our generalization. The principal new result of our analysis is that we find that an amplitude modulation of the output is to be expected from all maser oscillators excepting those in which excited atoms are supplied at a notably greater rate than the depopulation rate due to coherent induced emission. This analysis also suggests that energy population inversions of electric dipole moments can be performed using coherent light sources in the same manner as with magnetic dipole moments. It appears, therefore, that modulation of an optical or infrared maser (e.g., for communication purposes) may be easily accomplished by varying the population of excited states through control of the pumping energy. This modulation may be both by absorption and by amplification. Amplitude modulation of maser oscillators has been observed in this laboratory[3] and elsewhere[4-6] and we will explain these observations.

The total density of active atoms is composed of n_2, the density of upper energy state molecules, times the relative quantum (statistical) weights g_1/g_2, and n_1, the density of lower energy state atoms. We define the difference as the effective number of molecules given by

$$N_e \equiv n_2(g_1/g_2) - n_1. \tag{1}$$

N_e is dependent on the pumping system. For nondegenerate levels, $g_1/g_2 = 1$. The superposition state of a molecule may be described by

$$\psi = a_1(t)u_1 + a_2(t)u_2, \tag{2}$$

where $|a_1(t)|^2$ is the time-dependent probability coefficient of the lower state and $|a_2(t)|^2$ is the time-dependent coefficient for the upper state. The two states are orthogonal and may be normalized, but the existence of other states must be considered in such a normalization procedure. The frequency of transition is given by

$$\omega_0 = (W_2 - W_1)\hbar, \tag{3}$$

where W_2 is the energy of the upper energy level and W_1 is the energy of the lower level of the maser. The starting population distribution of the maser is assumed to be an inverted Boltzmann distribution (often referred to as a negative temperature). The inversion is assumed to satisfy the requirement that the molecular radiation rate will exceed the rate of energy dissipation (losses) so that a sustained oscillation can occur. After a given population distribution is established, the coherent induced radiation rate of the excited molecules, neglecting relaxation, collision processes, and spontaneous emission, is given by

$$\frac{d}{dt}[\hbar\omega_0 N_e |a_2(t)|^2]. \tag{4}$$

The radiation wavelength is, in general, much greater than the distance between molecules. In fact, it is physically realistic to assume that, for any maser system, the radiation field acts upon the entire molecular system. The molecular system is the entire collection of maser molecules.

The value of $|a_1(t)|^2$ and $|a_2(t)|^2$ can be determined by a perturbation calculation.[7] The interaction energy between an oscillating electric field E_1 and an effective electric dipole moment p is given by Eq. (5). (Alternatively, a magnetic dipole μ and oscillating magnetic field H_1 may be substituted.)

$$H' = E_1 \cos\omega t \int u_1^* P u_2 dv = pE_1 \cos\omega t. \tag{5}$$

P is the dipole moment operator of the pertinent electric or magnetic dipole transition, ω is the angular frequency of the radiation, and p is the matrix element between the two states along the direction of E_1 (suitably averaged, e.g., see Shimoda, Wang, and Townes[8]). The values of a_1 and a_2 are then obtained (assuming no deviation from resonance) $[(\omega - \omega_0) = 0]$ as

$$|a_1(t)|^2 = \sin^2\left(\int_0^t \frac{pE_1(t)}{\hbar} dt\right),$$
$$|a_2(t)|^2 = \cos^2\left(\int_0^t \frac{pE_1(t)}{\hbar} dt\right), \tag{6}$$

where the integral of $E_1(t)$ over time is needed to account for the variation of $|E_1|$ with the emission. For masers using magnetic dipole transitions, $\Delta m = \pm 1$, and $H' = -\frac{1}{2}(\mu_x \pm \mu_y)(H_x \mp iH_y) \times \cos\omega t$. Then in Eq. (6) the term $pE_1(t)$ should be replaced by $\mu H_1(t)$. Note that $\mu/\hbar$ is generally defined as γ (the magnetogyric factor) in the magnetic case. Hence, $p/\hbar$ may also be defined as γ for electric dipole transitions. By substituting (6) into (4) we obtain the rate of coherent induced emission and absorption.

Next consider the equation governing the growth and decay of the radiation field. The change in stored energy over the bandwidth ($\Delta\nu$) and the cavity loss rate, in a cavity of quality factor Q, is described by

$$\frac{d}{dt}\left[\frac{\epsilon E_1^2}{4\pi}\right] + \frac{\omega_0 \epsilon E_1^2}{2Q \times 4\pi}. \tag{7}$$

We next set this equal to the molecular energy emission rate. It is convenient to utilize a filling factor[9] F to account for the variation in density and dielectric constant (or permeability) between the radiation field volume and the volume of the active molecules. The equation governing the exchange of energy between molecules and the radiation is

$$\frac{d}{dt}(E_1^2) + \frac{\omega_0 E_1^2}{2Q} = 4\pi N_e F h\nu_0 \frac{d}{dt}\left[\cos^2\left(\int_0^t \frac{pE_1(t)}{\hbar} dt\right)\right]. \tag{8}$$

By using (6) for the value of a_2, we have a nonlinear differential equation which can be solved approximately or numerically. If N_e is assumed to be constant, Eq. (8) describes a maser which is excited and then permitted to radiate such as an ammonia maser, a two-level paramagnetic maser, an optical ruby or samarium maser when excited by pulsed light, etc. If we wish to describe a maser which is continuously pumped,[10] additional terms are added, and the right-hand side of (8) becomes

$$4\pi F\hbar\omega_0\left[N_e + \int R_e dt\right]\frac{d}{dt}(|a_2|^2), \tag{9}$$

where R_e is the rate of pumping to decrease the density of lower state molecules relative to the density of upper state molecules.

The solution of (8) gives the line shape of the energy of the emitted radiation. The amplitude and period of the amplitude modulations of a maser are related to the coherence and intensity of the emitted radiation. We have carried out numerical solutions using an electronic computer

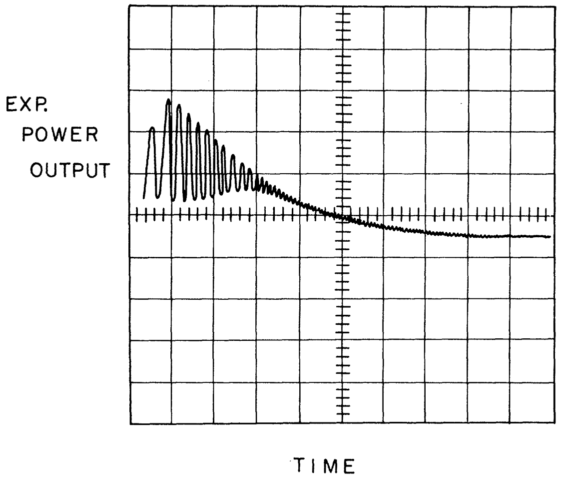

FIG. 1. Experimentally observed radiation envelope from a neutron-radiation-damaged calcite sample at X band [J. C. Kemp, Ph.D. dissertation, Electrical Engineering Department, University of California; issued as Electronics Research Laboratory Report No. 275, 1960 (unpublished)]. Activation is by a microwave adiabatic fast passage.

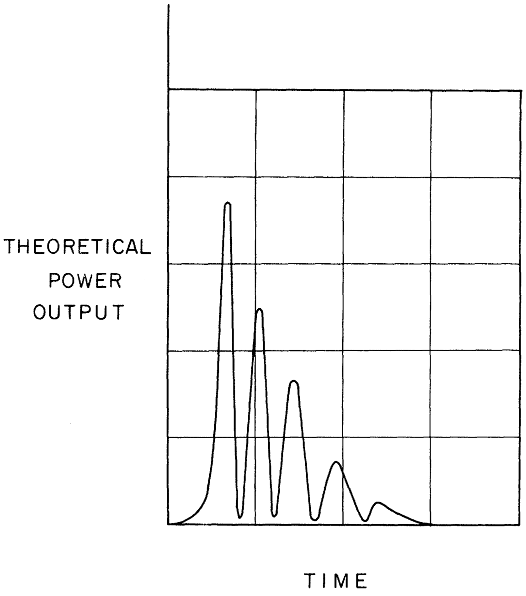

FIG. 2. Digital computer solution to Eq. (8). The scale is not the same as in Fig. 1. The amplitude and periods of amplitude modulation agrees with the experimental results within the experimental error.

for the case of a paramagnetic maser. The results agree with experiments as illustrated by Figs. 1 and 2.

An approximate result of the solution of (8) is that the output pulse will exponentially decay and will have amplitude modulation of a frequency

$$\omega_1 \sim (p/\hbar)(FN_e h\nu_0)^{1/2} \sim (p/\hbar)E_1. \qquad (10)$$

Since E_1 decays exponentially with time, the frequency of amplitude modulation will be higher at the peak amplitude than at lower output amplitudes. This has been experimentally observed. It may be noted from (10) that electric dipole two-level population inversion optical masers may be designed by supplying a pulse of radiation for a time given by $T=h/(2pE_1)$, where E_1 is the field strength of the coherent pulse of radiation. (This inversion is analogous to a paramagnetic spin echo first observed by Hahn.[11])

Amplitude modulation of the ammonia beam maser[1] is not prominent for the reason suggested in the first paragraph. A molecule passes through the maser cavity in an average time of about 10^{-4} second. Due to the low radiation energy density in the cavity, the period of amplitude modulation is greater than 10^{-4} second. Hence, the supply of upper state molecules exceeds the rate of loss by coherent radiation. In calculating the power output of the ammonia maser, Gordon, Zeiger, and Townes[1] were able to use an approximation which eliminated the nonlinear terms in our Eq. (9). The linearization is correct as noted in their paper for low-power masers such as the ammonia maser.

A more detailed analysis of the generalized equations, taking account of the molecular linewidth, is in the process of solution and will be published.

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*This work was supported in part by the Office of Scientific Research, by the Miller Institute for Basic Research in Science, and by the Faculty Research Committee.

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POLARIZATION AND THE TRIPLET ELECTRON-HYDROGEN SCATTERING LENGTH

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Recently, upper bounds for the triplet and singlet scattering lengths of electrons scattered from atomic hydrogen have been calculated by Rosenberg, Spruch, and O'Malley.[1] In the triplet case the upper bound was found to be $a_t \leq 1.91$.[2] This bound was obtained from an expression involving an arbitrary wave function ψ having a correct asymptotic form of an s-scattered wave at infinity. The particular function used by Rosenberg et al. did not contain a slowly vanishing part ($\sim -r^{-2}$) coming from the induced polarization in the hydrogen atom. A recent calculation[3] in which such a term was included gave a scattering length practically identical with the RSO bound, from which it may have been inferred that the polarization has an almost negligible effect on the scattering length and that the value $a_t = 1.91$ is very close to the correct scattering length. We wish to show that the polarization has a very important effect on the scattering length (of the order of 10%). Secondly, we want to report an accurate calculation of the triplet scattering length:

$$a_t = 1.74, \tag{1}$$

where the error is estimated to be less than three percent. The difference between this value and that of RSO is almost wholly due to the long-range polarization.

The effect of polarization as well as the calculation may be understood from a method which has been outlined in this journal.[4] The basic formula of that approach is

$$\sin(\delta-\delta_0) = \frac{-1}{k}\sum_{l=1}^{\infty}\frac{2}{(2l+1)^{1/2}}\int_0^{\infty} dr_1 \int_0^{r_1} dr_2 \Phi_0^{(0)} \frac{r_2^{\,l}}{r_1^{\,l+1}}\Phi_l. \tag{I.5}$$

δ is the exact phase shift of the whole problem and $\Phi_l(r_1 r_2)$ are "coefficients" in the expansion of the exact s-wave wave function in terms of Legendre polynomials $P(\cos\theta_{12})$, Eq. (I.1). δ_0 is the phase shift associated with a zeroth order problem, Eq. (I.4), which in particular does not contain any induced multipole distortion (polarization) effects.

Assume that we have computed δ_0 and all $l > 1$ terms on the right-hand side of (I.5). Assume also that only that part of the $l = 1$ term has been calculated by integrating up to a point R beyond which the difference between Φ_1 and its asymptotic form,

$$\lim_{r_1\to\infty} \Phi_1 = \frac{-2}{\sqrt{3}}\,\frac{\sin(kr_1+\delta)}{r_1^{\,2}}\,e^{-r_2}\left(\tfrac{1}{2}r_2^{\,3}+r_2^{\,2}\right), \tag{I.6}$$

can be considered negligible. Let $\delta(R)$ be the phase shift so calculated, and let $a(R)$ be the associated scattering length:

$$\lim_{k\to 0} \delta(R) = \pi - a(R)k.$$

$a \equiv a(\infty)$ is the exact scattering length. We find