

Induced and Spontaneous Emission in a Coherent Field. V. Theory of Molecular Beam Amplification

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The theory of the previous articles of this series is extended to allow analysis of the spectral distribution of induced and spontaneous emission in a cavity and to study the steady-state condition when a molecular beam traverses the cavity.

It is shown that the induced emission energy, when averaged over a reasonable time, depends only on the energy spectrum of the driving field. Expressions are obtained for the spectral distribution of induced, spontaneous, and thermally induced emission. These results are then applied to a molecular beam analysis and conditions for a steady state in the cavity are obtained.

The amplification by the molecular beam of the driving field (signal) and of the thermal and spontaneous emission energies (noise) is studied. Signal and noise energies are compared and the dependence of the results on both the bandwidth of the signal and the bandwidth of detection is examined.

INTRODUCTION

IN the previous articles of the present series,¹ a study was made of the emission of two-level molecules in a sinusoidally driven resonant cavity. The molecules were present in the cavity initially, and their induced and spontaneous emission were studied for a time limited by the fact that a perturbation approach toward the molecules was used. In the present article, the theory is extended in two directions. First, the driving field need no longer be sinusoidal; it is described by an arbitrary power spectrum, and the spectral distribution of both induced and spontaneous emission is studied. Secondly, instead of considering a single group of molecules subject to identical conditions, we consider a molecular beam passing through the cavity, and discuss the steady-state conditions existing in the cavity.

The first generalization, which permits the consideration of driving fields of various bandwidths, leads to a greater understanding and unification of results obtained in the earlier articles, where only two types of fields acting on the molecules were encountered: the sinusoidal driving field which was labeled "coherent," and the thermal field which was labeled "incoherent." These two types of fields produced different effects on the molecules. We are now in a position to consider fields which are intermediate between the two, and see how the results for one type transform into those for the other. The second generalization applies the theory developed to a realistic experimental situation, that of a molecular beam amplifier (maser)² in steady-state operation. The main question of interest is the interaction between the beam and the cavity field, and little attention is given to the circuit aspects of the amplifier, since the latter depend on particular experimental arrangements and are not within the scope of the present discussion. However, some general results

concerning signal and noise in molecular beam amplifiers is derived, since they are of fundamental interest.

Some of the preliminary aspects of the present article are the same as those of earlier articles of the present series. For the sake of intelligibility, we summarize these aspects briefly, and refer the reader to the appropriate earlier article for a more thorough discussion. It should not be assumed, however, that all the developments of the previous articles are used as the foundation upon which to build the present extension of the theory. Only those developments are used which are essential to the problems under consideration. Thus, although a molecular frequency spread was considered in III, we assume in the present analysis, as was done in II, that all the molecules are identical. Similarly, we do not utilize the large-signal approach (toward the field) developed in IV, but use perturbation-theory methods, which are satisfactory for present purposes.

Part I contains the theory needed for the discussion of the behavior of a single molecule, and Part II consists of a study of this behavior. Part III treats the interaction between a molecular beam and a steady-state cavity field, and Part IV considers questions related to signal and noise.

PART I

In the cavity under consideration (the same as that considered in II-IV) we consider only a single mode of (angular) frequency ω and loss constant β . (The quality factor Q_e is ω/β .)

The field is described in the usual manner by

$$\mathbf{E} = -4\pi c \mathbf{u}(\mathbf{r}) P(t), \quad \mathbf{H} = \nabla \times \mathbf{u}(\mathbf{r}) Q(t), \quad (1)$$

where $\mathbf{u}(\mathbf{r})$ is a normalized function—determined by the cavity geometry—which describes the spatial dependence of the field, and $Q(t)$ and $P(t)$ are the quantum mechanical field operators satisfying the commutation relationship $[Q(t), P(t)] = i\hbar$. For convenience, we shall refer to either Q or P (as well as to

¹ I. R. Senitzky, *Phys. Rev.* **111**, 3 (1958); **115**, 227 (1959); **119**, 1807 (1960); **123**, 1525 (1961), hereafter referred to as I, II, III, and IV, respectively.

² J. P. Gordon, H. J. Zeiger, and C. H. Townes, *Phys. Rev.* **95**, 282 (1954); *Phys. Rev.* **99**, 1264 (1955).

E and II, of course) as the field. The field energy is given by

$$H_f = 2\pi c^2 P^2 + (\omega^2/8\pi c^2) Q^2. \quad (2)$$

We consider the case in which the cavity is driven by a field of external origin, the driving field $P_D(t)$, about which we say at this time only that it is a steady-state—or stationary—field; that is, its square, when averaged over a sufficient number of cycles, is constant.

We consider, further, the situation in which a single molecule is introduced into the cavity at time $t=0$, and leaves the cavity before it has undergone a large change. The field, also, is assumed to be changed only slightly by the presence of the single molecule. Perturbation theory may therefore be used. The molecule is the same as that considered in II. It has two energy levels, and is in resonance with the cavity, so that

$$\hbar\omega = E_2 - E_1, \quad (3)$$

where E_2 and E_1 are the upper and lower molecular energies, respectively. Its initial state is given by

$$\psi = a_1 \varphi_1 + a_2 \varphi_2, \quad (4)$$

in obvious notation. The molecule couples to the field through an electric dipole moment $\gamma(t)$ with an interaction energy (see Appendix B)

$$4\pi cu \gamma P, \quad (5)$$

where u is the magnitude of $\mathbf{u}(\mathbf{r}_m)$ at position of the molecule \mathbf{r}_m and γ (hereafter referred to as dipole moment) is the component of the dipole moment along $\mathbf{u}(\mathbf{r}_m)$. For the sake of simplicity, we assume that $\mathbf{u}(\mathbf{r}_m)$ is constant during the entire time the molecule spends in the cavity. The behavior of the molecule is described in the Heisenberg picture by two dynamical variables, the Hamiltonian H_m , and dipole moment γ . The representation used is that which diagonalizes the energy of the free molecule. The dipole-moment properties of the free molecule are given by

$$\gamma^{[0]} = \tilde{\gamma} \begin{pmatrix} 0 & e^{-i\omega t} \\ e^{i\omega t} & 0 \end{pmatrix}, \quad (6)$$

where $\tilde{\gamma}$ is a real constant. (A square-bracketed superscript indicates the perturbation-theory order.)

We come now to the equations of motion. For our purposes, it is most convenient to have all but one of them in integral form. In the preceding articles, these integral equations were derived from the conventional formulation in terms of the Heisenberg equations of motion together with an analysis of the effect of cavity dissipation.³ We refer the reader interested in this derivation to the preceding articles, and begin here

³ For a general treatment of cavity dissipation in quantum mechanics, see I. R. Senitzky, Phys. Rev. **119**, 670 (1960).

with a statement of the equations of motion:

$$H_m(t) = H_m^{[0]} + \frac{2\pi cu_m}{i\hbar} \int_0^t dt_1 \{ [H_m(t_1), \gamma(t_1)], P(t_1) \}, \quad (7)$$

where the notation for the symmetrized product, $\{A, B\} = AB + BA$, is used;

$$\begin{aligned} \gamma(t) = & \gamma^{[0]}(t) + \frac{4\pi cu}{\hbar^2} \int_0^t dt_1 \int_0^{t_1} dt_2 U(t-t_1) \\ & \times [\gamma(t_1), [\gamma(t_2), H_m(t_2)] P(t_2)] U^{-1}(t-t_1), \end{aligned} \quad (8)$$

where

$$U(\tau) = \exp[(i/\hbar) H_m^{[0]} \tau]; \quad (8a)$$

$$P(t) = P_D(t) + P_F(t) + P_\gamma(t), \quad (9)$$

where

$$P_\gamma(t) = -\frac{\omega}{c} \int_0^t dt_1 \gamma(t_1) e^{-i\beta(t-t_1)} \sin\omega(t-t_1), \quad (10)$$

and P_F is the “free” cavity field exhibiting both quantum-mechanical and thermal fluctuations,³

$$P_F = -\frac{1}{\omega} \int_{-\infty}^t dt_1 F(t_1) e^{-i\beta(t-t_1)} \sin\omega(t-t_1), \quad (11)$$

F being a fluctuation operator the properties of which are given by the expectation values,

$$\langle F(t) \rangle = 0, \quad (12)$$

$$\langle F(t_1) F(t_2) \rangle = \frac{\omega^2 \hbar \beta}{4\pi^2 c^2} \left[i \frac{\mathcal{P}}{t_2 - t_1} + \pi \delta(t_1 - t_2) f(T) \right], \quad (13)$$

where

$$f(T) = 1 + 2(e^{\hbar\omega/kT} - 1)^{-1} \quad (13a)$$

and T is the temperature of the cavity. To complete the set of equations of motion, we add the differential equation

$$Q = -(4\pi c^2/\omega^2) \dot{P}. \quad (14)$$

Equations (7), (8), (9), and (14) determine completely the behavior of the interacting field and molecule in terms of the dynamical variables H_m , γ , P , and Q .

PART II

We now ask the following question: What is the expectation value of the induced and spontaneous emission power radiated by the molecule while in the cavity? From Eq. (7), we have

$$\dot{H}_m = (2\pi cu/i\hbar) \{ [H_m(t), \gamma_m(t)], P(t) \}. \quad (15)$$

A straightforward calculation based on the equations of motion (some of this calculation is carried out in detail in II and III) shows that the part of $\dot{H}_m^{[2]}$ which

depends on $P_D(t)$ is given by

$$-(\dot{H}_m^{[2]})_{\text{ind}} = \frac{16\pi^2 c^2}{\hbar} \omega n^2 \bar{\gamma}^2 I \int_0^t dt_1 \times \{P_D(t), P_D(t_1)\} \cos \omega(t-t_1), \quad (16)$$

where I is a matrix that operates on the initial state vector of the molecule and is given by

$$I \equiv \begin{pmatrix} -1 & 0 \\ 0 & 1 \end{pmatrix}. \quad (16a)$$

For the expectation value of I we have, from Eq. (5),

$$\langle I \rangle = |a_2|^2 - |a_1|^2. \quad (17)$$

Denoting the expectation value of power emitted by the molecule by p , we have

$$p_{\text{ind}} = 4\pi c^2 M \int_0^t d\tau P_D(t) P_D(t-\tau) \cos \omega \tau, \quad (18)$$

where

$$M \equiv 8\pi \omega n^2 \bar{\gamma}^2 \langle I \rangle / \hbar. \quad (18a)$$

We are not really interested in the instantaneous value of the induced power emitted by the molecule, but rather in a smoothed out value, averaged over a reasonable number of cycles, which will yield information about the secular changes in the molecule. It might be said that we are interested in a value of the induced emission which is averaged over a time long compared to a cycle but short compared to other significant times, such as the cavity relaxation time and the time the molecule spends in the cavity. We can therefore average both sides of Eq. (18) over such a time. Now, the average of $P_D(t)P_D(t-\tau)$ over this short time is the same as its average over a long time—or over the entire time it acts in the cavity—since $P_D(t)$ represents a steady-state field. We, therefore, replace the short-time average of $P_D(t)P_D(t-\tau)$ by its long-time average and use no special notation to indicate the short-time average. Equation (18) then becomes

$$p_{\text{ind}} = 4\pi c^2 M \int_0^t d\tau \langle P_D(t) P_D(t-\tau) \rangle_{\text{av}} \cos \omega \tau. \quad (19)$$

The function $\langle P_D(t) P_D(t-\tau) \rangle_{\text{av}}$, which is a function of τ only, is the correlation function of the driving field, and bears a well-known relationship to the energy spectrum of the field. Let us assume that the driving field acts in the interval $-\frac{1}{2}T \leq t \leq \frac{1}{2}T$, where T is large compared to other times of interest. Then,

$$\langle P_D(t) P_D(t-\tau) \rangle_{\text{av}} = \frac{1}{T} \int_{-\frac{1}{2}T}^{\frac{1}{2}T} dt P_D(t) P_D(t-\tau). \quad (20)$$

Setting

$$P_D(t) = \int_0^\infty d\omega' P_D(\omega') \cos \omega' t, \quad (21)$$

we obtain by a routine calculation

$$\langle P_D(t) P_D(t-\tau) \rangle_{\text{av}} \approx \frac{\pi}{T} \int_0^\infty d\omega' P_D^2(\omega') \cos \omega' \tau, \quad (22)$$

for T sufficiently large. We therefore have

$$p_{\text{ind}} = \frac{4\pi^2 c^2 M}{T} \int_0^t d\tau \int_0^\infty d\omega' P_D^2(\omega') \cos \omega' \tau \cos \omega \tau, \\ \approx \frac{2\pi^2 c^2 M}{T} \int_0^\infty d\omega' P_D^2(\omega') \frac{\sin(\omega' - \omega)t}{\omega' - \omega}, \quad (23)$$

where the approximation holds for $t \gg \omega^{-1}$.

It is convenient to define a driving energy per unit frequency range $\mathcal{E}_D(\omega')$ in the cavity, so that the total driving energy is given by

$$\mathcal{E}_D = \int_0^\infty d\omega' \mathcal{E}_D(\omega'). \quad (24)$$

Since the total driving energy may also be expressed as $4\pi c^2 \langle P_D^2(t) \rangle_{\text{av}}$, we have, from Eq. (22),

$$\mathcal{E}_D(\omega') = 4\pi^2 c^2 P_D^2(\omega') / T, \quad (25)$$

and from Eq. (23)

$$p_{\text{ind}} = \frac{1}{2} M \int_0^\infty d\omega' \mathcal{E}_D(\omega') \frac{\sin(\omega' - \omega)t}{\omega' - \omega}. \quad (26)$$

We can thus eliminate T by the use of $\mathcal{E}_D(\omega')$, which is given directly by

$$\mathcal{E}_D(\omega') = 8c^2 \int_0^\infty d\tau \langle P_D(t) P_D(t-\tau) \rangle_{\text{av}} \cos \omega' \tau. \quad (27)$$

From Eq. (26) we obtain the induced emission per unit frequency range⁴:

$$p(\omega')_{\text{ind}} = \frac{1}{2} M \mathcal{E}_D(\omega') \frac{\sin(\omega' - \omega)t}{(\omega' - \omega)}. \quad (28)$$

Equation (26) shows that the induced emission of interest to us is determined completely by the energy spectrum of the driving field. This fact yields a deeper insight into the results of the previous articles of the present series, where it was found that the induced emission produced by the sinusoidal signal field was different from that produced by the thermal field, and the difference was classified on the basis of the coherence or incoherence of the driving field. We see now that we have a more sensitive classification in the energy (or power) spectrum of the driving field, which may be

⁴ In the case of energy and power, use of the argument ω' or ω indicates density in frequency space, and absence of the argument (or use of the argument t) indicates the total quantity. Note that $p(\omega)$ and p have different dimensions. In connection with Eq. (28), see also reference 6, Chap. V.

applied to both coherent and incoherent fields, and are in a position to consider incoherent fields having different energy spectra from that of the thermal field.

For purposes of computation, it is often simpler to use the correlation function and Eq. (19), rather than the energy spectrum and Eq. (26). Also, when dealing with incoherent fields, an ensemble average may be used instead of a time average.

It is instructive to study a specific driving-field distribution. Consider a Lorentzian energy spectrum of width α given by

$$\mathcal{E}_D(\omega') = \frac{1}{2\pi} \frac{\alpha \mathcal{E}_D}{(\omega' - \omega)^2 + \frac{1}{4}\alpha^2}, \quad (29)$$

to which there corresponds a correlation function

$$\langle P(t)P(t-\tau) \rangle_{av} = \frac{1}{4\pi c^2} \mathcal{E}_D e^{-(1/2)\alpha\tau} \cos\omega\tau. \quad (30)$$

By substituting Eq. (29) into Eq. (26), or Eq. (30) into Eq. (19), we obtain

$$p_{ind} = M \mathcal{E}_D \alpha^{-1} (1 - e^{-(1/2)\alpha t}). \quad (31)$$

It is interesting to compare this expression with the induced emission produced by the thermal field in the cavity. From Eq. (II 81), we have

$$p_{th} = M \hbar \omega (e^{\hbar\omega/kT} - 1)^{-1} \beta^{-1} (1 - e^{-(1/2)\beta t}). \quad (32)$$

We see that the thermal field produces the same induced emission as a driving field of total energy

$$\mathcal{E}_{0th} = \hbar\omega (e^{\hbar\omega/kT} - 1)^{-1}, \quad (33)$$

with a Lorentzian distribution of width β . We can show directly from Eq. (11) that Eq. (29), with $\alpha = \beta$, is, indeed, the distribution of the thermal field energy. One calculates first that (the averaging is now over an ensemble)

$$\frac{1}{2} \langle \{P_F(t), P_F(t-\tau)\} \rangle = (4\pi c^2)^{-1} (\mathcal{E}_{0th} + \frac{1}{2}\hbar\omega) e^{-(1/2)\beta\tau} \cos\omega\tau. \quad (34)$$

The $\frac{1}{2}\hbar\omega$ term represents quantum-mechanical vacuum fluctuations, and the remaining term yields immediately

$$\mathcal{E}_{0th}(\omega') = \frac{1}{2\pi} \frac{\beta \mathcal{E}_{0th}}{(\omega' - \omega)^2 + \frac{1}{4}\beta^2}, \quad (35)$$

which, for $\alpha = \beta$, is in agreement with Eq. (29).

It is of interest to consider also a Gaussian spectral distribution for the driving field. Let us set

$$\mathcal{E}_D(\omega') = (2\mathcal{E}_D/\pi^{\frac{1}{2}}\alpha') \exp[-(\omega' - \omega)^2/\frac{1}{4}\alpha'^2]. \quad (36)$$

The correlation function for this distribution is

$$\langle P(t)P(t+\tau) \rangle_{av} = \frac{\mathcal{E}_D}{4\pi^{\frac{1}{2}}c^2} \exp\left(-\frac{\alpha'^2\tau^2}{16}\right) \cos\omega\tau. \quad (37)$$

For the induced emission we obtain, by using either Eq. (19) or Eq. (23),

$$p_{ind} = \pi^{\frac{1}{2}}\alpha'^{-1} M \mathcal{E}_D \operatorname{erf}(\frac{1}{4}\alpha' t), \quad (38)$$

where the error function notation is

$$\operatorname{erf} x \equiv 2\pi^{-\frac{1}{2}} \int_0^x dy e^{-y^2}. \quad (38a)$$

We see that in the case of both spectral distributions, the Lorentzian and the Gaussian, the induced emission power approaches a constant value after a transient period which is of the order of magnitude of the inverse spectral width. In both cases the constant value of the induced emission is inversely proportional to the spectral width. As the spectral width becomes infinitesimal, that is, as the driving field becomes a pure sine wave, the induced emission of either distribution becomes

$$p_{ind} = \frac{1}{2} M \mathcal{E}_D l, \quad (39)$$

which is identical with Eq. (II 84) for the induced emission produced by a sinusoidal driving-field in resonance with the molecules. We also note that each type of spectral distribution of the driving field produces its own characteristic time dependence of the induced emission during the transient period.

We come now to spontaneous emission. It, too, is obtained from Eq. (15). In absence of a driving-field, the second-order part of Eq. (15) gives both the thermally induced and the spontaneous emission. We have already examined the former; the latter is given by (see II or III for the details of the derivation)

$$p_{sp} = M' \hbar \omega \beta^{-1} (1 - e^{-(1/2)\beta t}), \quad (40)$$

where

$$M' = (8\pi\omega u^2 \tilde{\gamma}^2 |a_2|^2)/\hbar. \quad (40a)$$

By comparison with Eqs. (31), (29), and (28), one sees that the induced emission power per unit frequency range may be expressed as

$$p_{sp}(\omega') = \frac{1}{2} M' \mathcal{E}_{0sp}(\omega') \frac{\sin(\omega' - \omega)t}{\omega' - \omega}, \quad (41)$$

where

$$\mathcal{E}_{0sp}(\omega') = (1/2\pi)\beta\hbar\omega [(\omega' - \omega)^2 + \frac{1}{4}\beta^2]^{-1}. \quad (41a)$$

It is interesting to note that when the molecule is completely in the upper state (that is, when $|a_2|^2 = 1$), $M' = M$, and then the spontaneous emission looks like induced emission produced by a field of total energy $\hbar\omega$ (twice the zero point energy) and Lorentzian distribution of width β . However, this is purely a formal resemblance, since, when we reach the condition $|a_2|^2 = 1/2$, the induced emission is zero, while the spontaneous emission is half its maximum. The detailed derivation of Eq. (40) from Eq. (15) shows that, from a strictly formal quantum-mechanical viewpoint,

spontaneous emission is due to the zero-point fluctuations of *both* the field and the molecular moment.⁵

We conclude this discussion with expressions for the energy per unit frequency range $\epsilon(\omega')$ radiated by the molecule during the time τ that it spends in the cavity. An integration of the expressions for the expectation value of the power yields

$$\epsilon_{\text{sig}}(\omega') = M \mathcal{E}_{0\text{sig}}(\omega') f(\omega', \tau), \quad (42)$$

$$\epsilon_{\text{th}}(\omega') = M \mathcal{E}_{0\text{th}}(\omega') f(\omega', \tau), \quad (43)$$

$$\epsilon_{\text{sp}}(\omega') = M' \mathcal{E}_{0\text{sp}}(\omega') f(\omega', \tau), \quad (44)$$

where

$$f(\omega', \tau) \equiv \sin^2[\tfrac{1}{2}(\omega' - \omega)\tau] / (\omega' - \omega)^2, \quad (42a)$$

and where $\mathcal{E}_{0\text{th}}(\omega')$ and $\mathcal{E}_{0\text{sp}}(\omega')$ are given by Eqs. (35) and (41a), respectively. The frequency distribution of the signal driving-field energy $\mathcal{E}_{0\text{sig}}(\omega')$ is of course arbitrary, and is expressed as $\mathcal{E}_0 \delta(\omega' - \omega'')$ when the driving field is a pure sine wave of frequency ω'' . On the other hand, $\mathcal{E}_{0\text{sp}}(\omega')$ is determined solely by the cavity loss and $\mathcal{E}_{0\text{th}}(\omega')$ is determined by both the cavity loss and the temperature. The latter has physical significance, being the thermal energy distribution in the cavity in the absence of molecules, while the former has no physical significance. Only the spontaneous emission itself has physical significance, and not any "driving field" which seems, formally, to produce it.

PART III

We consider now a molecular beam. Equations (42) to (44) apply to each molecule in the beam. We cannot, however, speak any longer of a specific transit time, since in a molecular beam, as usually prepared, there is a spread of velocities and therefore a spread of transit times. In order to be able to use Eqs. (42) to (44), we must average the expressions over the beam velocities, which involves the averaging of $f(\omega', \tau)$. Now if the beam comes from a source in which there is a Maxwell-Boltzman velocity distribution, the velocity distribution in the beam is given by⁶

$$\rho(v) = 2\eta^{-4} v^3 \exp(-v^2/\eta^2), \quad (45)$$

where

$$\eta = (2kT/m)^{1/2}. \quad (45a)$$

We therefore have

$$\bar{f}(\omega', \tau) = \int_0^\infty dv \frac{\sin^2[\tfrac{1}{2}(\omega' - \omega)(l/v)]}{(\omega' - \omega)^2} \rho(v), \quad (46)$$

where l is the length of the molecular path through the cavity. It is convenient to rewrite this integral as

$$\bar{f}(\omega', \tau) = \tfrac{1}{4} \tau_0^2 \psi[(\omega' - \omega)\tau_0], \quad (47)$$

⁵ Compare with comment by E. T. Jaynes, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 290, and A. E. Siegman, *Proc. Inst. Radio Engrs.* **49**, 633 (1961).

⁶ Norman F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956), p. 20.

where

$$\tau_0^2 \equiv (l/\eta)^2 = \langle \tau^2 \rangle_{\text{av}} \quad (47a)$$

and

$$\psi(z) \equiv 8 \int_0^\infty dy y^3 \exp(-y^2) \frac{\sin^2[\tfrac{1}{2}(z/y)]}{z^2}. \quad (47b)$$

The function $\psi(z)$ cannot be evaluated in closed form. It is not difficult to see, however, that it is a bell-shaped distribution curve with a maximum of unity at $z=0$, and with an asymptotic value of $2/z^2$. We also have

$$\int_0^\infty d\omega' \psi[(\omega' - \omega)\tau_0] = \pi^{1/2} / \tau_0. \quad (48)$$

A tabulation has been made by Ramsey and Cruse⁷ of the function

$$I(z) = \int_0^\infty dy y^3 \exp(-y^2) \cos(z/y), \quad (49)$$

in terms of which $\psi(z)$ may be expressed:

$$\psi(z) = 4[\tfrac{1}{2} - I(z)] / z^2. \quad (50)$$

Using the notation

$$\varphi(\omega') \equiv \psi[(\omega' - \omega)\tau_0], \quad (51)$$

we have now, instead of Eqs. (42) to (44), the equations

$$\bar{\epsilon}_{\text{sig}}(\omega') = \tfrac{1}{4} \tau_0^2 M \mathcal{E}_{0\text{sig}}(\omega') \varphi(\omega'), \quad (52)$$

$$\bar{\epsilon}_{\text{th}}(\omega') = \tfrac{1}{4} \tau_0^2 M \mathcal{E}_{0\text{th}}(\omega') \varphi(\omega'), \quad (53)$$

$$\bar{\epsilon}_{\text{sp}}(\omega') = \tfrac{1}{4} \tau_0^2 M' \mathcal{E}_{0\text{sp}}(\omega') \varphi(\omega'), \quad (54)$$

where it is recalled that

$$\mathcal{E}_{0\text{th}}(\omega') = [e^{\hbar\omega/kT} - 1]^{-1} \mathcal{E}_{0\text{sp}}(\omega'), \quad (53a)$$

and

$$\mathcal{E}_{0\text{sp}}(\omega') = \frac{1}{2\pi} \beta \hbar \omega [(\omega' - \omega)^2 + \tfrac{1}{4}\beta^2]^{-1}. \quad (54a)$$

Equations (52) to (54) form the basis of our molecular beam calculation.

We consider a steady-state situation with a molecular beam passing through the cavity, N being the number of molecules entering the cavity per second. Our attention will be confined to the situation in which the molecules undergo only a small change during their transit time (except for the very slow ones, of which there is a negligible number); the above perturbation theory is therefore applicable to each molecule. We further restrict our consideration to those cases where the average transit time is much larger than the cavity relaxation time,⁸ which implies

$$\tau_0 \gg \beta^{-1}. \quad (55)$$

⁷ Norman F. Ramsey, *Molecular Beams* (Oxford University Press, New York, 1956), Appendix D.

⁸ In a typical ammonia beam apparatus, $\tau_0 \sim 10^{-4}$ and $\beta \sim 10^7$.

We are interested in a steady-state situation in which a small amount of signal power is fed into the cavity and amplified by the molecular beam. Let the power per unit frequency range fed into the cavity be specified by $\mathcal{P}(\omega')$ and the energy per unit frequency range in the cavity be specified by $\mathcal{E}_{\text{sig}}(\omega')$. The average induced energy emitted by each molecule as it passes through the cavity is given by Eq. (52), with $\mathcal{E}_{0\text{sig}}(\omega')$ replaced by $\mathcal{E}_{\text{sig}}(\omega')$. In any given frequency range, the input power plus induced emission power must equal the dissipated signal power.⁹ We, therefore, have (assuming β independent of frequency in the frequency range of interest)

$$\mathcal{P}(\omega') + \frac{1}{4}\tau_0^2 NM \mathcal{E}_{\text{sig}}(\omega') \varphi(\omega') = \beta \mathcal{E}_{\text{sig}}(\omega'), \quad (56)$$

which yields

$$\mathcal{E}_{\text{sig}}(\omega') = [\mathcal{P}(\omega')/\beta] G(\omega'), \quad (57)$$

where

$$G(\omega') \equiv [1 - \frac{1}{4} NM \tau_0^2 \varphi(\omega')/\beta]^{-1}. \quad (57a)$$

$G(\omega')$ may be regarded as the gain, since it is unity in the absence of molecules, and increases with beam intensity. Its maximum value is

$$G_m = G(\omega) = [1 - \frac{1}{4} NM \tau_0^2/\beta]^{-1}. \quad (58)$$

If this quantity becomes infinite, oscillation occurs. For $(\frac{1}{4} NM \tau_0^2/\beta) \geq 1$, our theory is inapplicable. As a matter of fact, the theory becomes inapplicable when the amplified field is large enough to produce large changes in the state of the average molecule.¹⁰

The same reasoning that was used to derive Eq. (57) can be applied to the steady-state thermal and spontaneous emission energies in the cavity.¹¹ In the calculation of thermal energy, the "input power" is the thermal power $\beta \mathcal{E}_{0\text{th}}$ [given by Eq. (53a)] radiated by the walls, so that

$$\mathcal{E}_{\text{th}}(\omega') = \mathcal{E}_{0\text{th}}(\omega') G(\omega'); \quad (59)$$

and in the calculation of spontaneous emission energy, the "input power" is the spontaneous emission power $N \bar{\epsilon}_{\text{sp}}(\omega')$ [given by Eq. (54)], so that

$$\mathcal{E}_{\text{sp}}(\omega') = [N \bar{\epsilon}_{\text{sp}}(\omega')/\beta] G(\omega'). \quad (60)$$

⁹ We are not considering explicitly an output. In order to avoid involvement in circuit considerations, we assume that, as far as the field in the cavity is concerned, the output is indistinguishable from—and is incorporated into—the loss.

¹⁰ For $(\frac{1}{4} NM \tau_0^2/\beta) > 1$, a steady state cannot exist within the framework of our theory, since the (theoretical) induced emission power exceeds the dissipated power. If experimental conditions are such that this inequality is satisfied, then the steady state which does occur in the experimental situation is such that the amplified field is large enough to produce significant changes in the state of the average molecule, so that the molecular amplification is no longer linear, and our induced emission formula is not applicable. It is easy to see that in the case of the high gain and narrow-band signal, the condition that our theory be valid is that $\mathcal{E}_{\text{sig}} \beta < N \hbar \omega$.

¹¹ We are applying the energy balance equation to each type of energy separately: signal energy, thermal energy, and spontaneous emission energy. This separation is permissible because each type involves independent processes.

It is instructive at this point to study more closely the gain function $G(\omega')$, or better yet, the "reduced" gain function $g(\omega')$ defined by

$$g(\omega') = G(\omega') - 1. \quad (61)$$

For brevity the word "gain" will henceforth refer to the reduced gain. If we are interested only in the situation in which

$$G_m \gg 1, \quad (62)$$

then, from Eqs. (57a), (58), and (59), we obtain

$$g(\omega') \approx G_m \frac{\varphi(\omega')}{1 + G_m [1 - \varphi(\omega')]} \quad (63)$$

It is easy to see that $g(\omega')$ is, like $\varphi(\omega')$, a bell-shaped distribution curve with maximum at $\omega' = \omega$, but is much narrower than $\varphi(\omega')$. Thus, for $|\omega' - \omega|$ such that $g(\omega')$ drops to one-half of its maximum value, $\varphi(\omega')$ is $(1 + G_m)/(2 + G_m)$ of its maximum value, a very small change from maximum, according to Eq. (62). Conversely, for $|\omega' - \omega|$ such that $\varphi(\omega')$ drops to one half its maximum, $g(\omega')$ is $(2 + G_m)^{-1}$ of its maximum value, a much greater change. More will be said about the gain function later, but meanwhile we can utilize the fact that $\varphi(\omega')$ changes only slightly within the width of $g(\omega')$ to rewrite Eqs. (59) and (60), which give the amplified thermal and spontaneous emission energy.

$$\mathcal{E}_{\text{th}}(\omega') = \mathcal{E}_{0\text{th}}(\omega) g(\omega') + \mathcal{E}_{0\text{th}}(\omega'), \quad (64)$$

$$\mathcal{E}_{\text{sp}}(\omega') = [N \bar{\epsilon}_{\text{sp}}(\omega)/\beta] g(\omega') + [N \bar{\epsilon}_{\text{sp}}(\omega')/\beta]. \quad (65)$$

Equation (45) for the signal energy may likewise be rewritten. If the signal width is much narrower than the amplifier width,¹² we obtain,

$$\mathcal{E}_{\text{sig}}^{(n)}(\omega') = [\mathcal{P}(\omega')/\beta] G_m; \quad (66)$$

if it is much wider than the signal width, we may write

$$\mathcal{E}_{\text{sig}}^{(w)}(\omega') = [\mathcal{P}(\omega)/\beta] g(\omega') + \mathcal{P}(\omega')/\beta. \quad (67)$$

There are three physically significant spectral widths¹³ associated with the problem under consideration: First—and widest—there is the cavity width. It is the width of the unamplified thermal energy in the cavity described by Eq. (35). This width is given by $\frac{1}{2}\pi\beta$. Another spectral width is that of the unamplified spontaneous emission energy, $N \bar{\epsilon}_{\text{sp}}(\omega')/\beta$, which is the same, of course, as the width of the spontaneous emission energy of a single molecule $\bar{\epsilon}_{\text{sp}}(\omega')$, given by Eq. (54). Inspection of this equation shows that the variation of $\bar{\epsilon}_{\text{sp}}(\omega')$ is determined by two factors, $\varphi(\omega')$ and the Lorentzian which determines the cavity

¹² By amplifier width, we mean the width of the (reduced) gain function $g(\omega')$.

¹³ Since we are considering different types of spectral distributions, it is useful to have a precise definition of spectral width W of the energy distribution $S(\omega')$. A convenient definition is

$$W = \frac{1}{S_{\text{max}}} \int_0^\infty d\omega' S(\omega').$$

width. The width of $\varphi(\omega')$ is given by Eq. (48) as $\pi^{1/2}\tau_0^{-1}$, and this is much narrower than the cavity width, according to Eq. (55). We have, therefore,

$$\bar{\epsilon}_{\text{sp}}(\omega') \approx \frac{1}{4}\tau_0^2 M' \mathcal{E}_{0\text{sp}}(\omega) \varphi(\omega') \\ = \tau_0^2 M' \hbar \omega \varphi(\omega') / 2\pi\beta. \quad (68)$$

If we specialize our situation to that in which the molecules enter the cavity in the upper state ($|a_2|^2 = 1$) than, as mentioned previously, $M' = M$. When the amplifier is in a high gain condition, we can make use of Eqs. (62) and (58) to obtain for the unamplified power spontaneously emitted by the molecular beam

$$N \bar{\epsilon}_{\text{sp}}(\omega') = 2\hbar\omega \varphi(\omega') / \pi, \quad (69)$$

a particularly simple expression. Either Eq. (68) or (69) shows that the spectral width of the unamplified spontaneous emission is much less than the spectral width of the unamplified thermal energy in the cavity.

The third significant width is that of the amplification, or the gain. It was shown previously that this width is much less than that of $\varphi(\omega')$. In accordance with our definition,¹³ it is given by

$$W = \int_0^\infty d\omega' \frac{\varphi(\omega')}{1 + G_m[1 - \varphi(\omega')]} \\ = \frac{2}{\tau_0} F(G_m), \quad (70)$$

where

$$F(G_m) \equiv \int_0^\infty dz \frac{\psi(z)}{1 + G_m[1 - \psi(z)]}, \quad (70a)$$

z being a dimensionless variable. The function $F(G_m)$ can be evaluated only numerically. This is done in Appendix A, where it is shown by numerical comparison that, for $10 \leq G_m \leq 100$, we can conveniently set

$$F(G_m) \approx \tilde{F}(G_m), \quad (71)$$

where

$$\tilde{F}(G_m) \equiv \left[\int_0^\infty dz \psi(z) \right] G_m^{-1/2} = \frac{1}{2}\pi^{3/2} G_m^{-1/2}, \quad (71a)$$

which yields

$$W \approx \pi^{3/2} \tau_0^{-1} G_m^{-1/2}. \quad (72)$$

[It is seen that this is $G_m^{-1/2}$ times the width of $\varphi(\omega')$.] In our further discussion we will consider W to be a characteristic property of the amplifier; in other words, the amplifier will be characterized by G_m and W , its gain at center frequency and its width, respectively. The amplifier may also be characterized, when convenient, by the pair of constants G_m and τ_0 , if W is expressed in terms of G_m and τ_0 by means of Eq. (72).

PART IV

There are questions of fundamental interest in connection with amplifiers involving a comparison of signal and noise. The noise energy in our case is, of course, the (amplified) thermal and spontaneous emis-

sion energy. These questions usually refer not to energy per unit frequency range—in which our relationships are so far expressed—but to total energy. However, “total energy” does not always mean energy integrated over the entire frequency spectrum. Frequently, it refers to energy integrated over a small range of frequency determined by the bandwidth of the apparatus being used to measure the energy in the cavity (the receiver). We must therefore be careful to point out, whenever we speak of total energy, within what range the energy is being totaled.

Let us consider, first, only that field energy which is within the molecular amplifying range, that is, within the range of $g(\omega')$. The total thermal and spontaneous energies within this range are given by

$$\mathcal{E}_{\text{th}}^{(a)} = (2/\pi\beta) \hbar \omega [e^{\hbar\omega/kT} - 1]^{-1} G_m W \quad (73)$$

and

$$\mathcal{E}_{\text{sp}}^{(a)} = (2/\pi\beta) \hbar \omega G_m W, \quad (74)$$

respectively, where we have substituted from Eqs. (69) and (53a) into Eqs. (64) and (65), integrated over ω' within the limits of $g(\omega')$, and neglected the contribution of the second terms of the right sides of Eqs. (64) and (65) compared to the first terms in the range of ω' under consideration. If the signal is much narrower than the amplifier bandwidth, we have from Eq. (66)

$$\mathcal{E}_{\text{sig}}^{(n)} = (\mathcal{P}/\beta) G_m; \quad (75)$$

if it is much wider, then Eq. (67) yields

$$\mathcal{E}_{\text{sig}}^{(w)} = [\mathcal{P}(\omega)/\beta] G_m W. \quad (76)$$

The factor $[e^{\hbar\omega/kT} - 1]^{-1}$ determines the relative importance of the thermal and spontaneous emission noise. As the temperature approaches zero, the thermal noise disappears, of course, and only the spontaneous emission noise remains. The ratio R of signal energy to noise energy for narrow signal is then

$$R^{(a)(n)} = \frac{\mathcal{P}/W}{\frac{1}{2}\pi \frac{\hbar\omega}{\beta}}, \quad (77)$$

and for wide signal

$$R^{(a)(w)} = \frac{\mathcal{P}(\omega)}{\frac{1}{2}\pi \frac{\hbar\omega}{\beta}}. \quad (78)$$

This is the fundamental limit in the amplification of a molecular beam amplifier when the limiting bandwidth is W .

We consider now the case in which the bandwidth of interest is the entire cavity bandwidth; that is, we consider the true total energy of the single mode under consideration. The total thermal and spontaneous energies are then obtained by integrating the right sides of Eqs. (64) and (65) over all frequencies. The result is

$$\mathcal{E}_{\text{th}}^{(a)} = \frac{\hbar\omega}{e^{\hbar\omega/kT} - 1} \left[\frac{2G_m W}{\pi\beta} + 1 \right], \quad (79)$$

$$\mathcal{E}_{\text{sp}}^{(a)} = \frac{2\hbar\omega}{\pi\beta} \left[G_m W + \frac{\pi^{3/2}}{\tau_0} \right]. \quad (80)$$

We see from Eq. (72) that

$$G_m W = \pi^{3/2} \tau_0^{-1} G_m^{1/2}, \quad (81)$$

which, in accordance with Eq. (55), and for reasonable values of G_m (~ 100), is much smaller than β . The first term in the bracket of Eq. (79) is therefore negligible, which means that the unamplified thermal energy (in the entire cavity bandwidth) is much larger than the thermally induced energy coming from the molecular beam (in the narrow amplification range). As for the spontaneous emission energy, we have, using Eq. (72),

$$\mathcal{E}_{sp}^{(e)} = (2\pi^{1/2}/\beta\tau_0)\hbar\omega(G_m^{1/2}+1). \quad (82)$$

In contrast to the situation in the case of the thermal noise, the amplified spontaneous-emission noise is considerably larger than the unamplified noise. The reason for this contrast is the fact that the difference in frequency spread between unamplified noise and molecular amplification is much larger for the thermal energy than for the spontaneous emission energy. It is also evident that the relative importance of the thermal and spontaneous emission energies depends on the bandwidth under consideration. Thus, if the energy within the amplification bandwidth only is considered, the temperature at which the thermal noise becomes equal to the spontaneous emission noise is given by $(\hbar\omega/k)(\ln 2)^{-1}$; if the energy within the entire cavity bandwidth is considered, this temperature is approximately $(\hbar\omega/k)\{\ln[\beta\tau_0/2(\pi G_m)^{1/2}]\}$, a much lower value. The ultimate signal to noise ratio as the temperature approaches zero (note that lower temperatures are involved now than those in the previous case), is approximately the same as in the previous case—if we neglect unity compared to $G_m^{1/2}$ —and is given by Eqs. (77) and (78).¹⁴

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APPENDIX A

Ramsey and Cruse⁷ tabulate the function

$$I(z) = \int_0^\infty dy y^3 \exp(-y^2) \cos(z/y) \quad (A1)$$

for values of z ranging from 0 to 20. The function

$$F(x) = \int_0^\infty dz \frac{\psi(z)}{1+x[1-\psi(z)]}, \quad (A2)$$

¹⁴ The comparison of signal and noise in maser amplifiers has been considered by the following: K. Shimoda, H. Takahasi, and C. H. Townes, J. Phys. Soc. Japan **12**, 868 (1957); M. Muller, Phys. Rev. **106**, 8 (1957); M. W. P. Strandberg, *ibid.* **106**, 617 (1957); R. Pound, Ann. Phys. (New York) **1**, 24 (1957); J. Weber, Revs. Modern Phys. **31**, 681 (1959). These authors do not discuss in detail the bandwidth aspects of either the signal or the noise.

TABLE I. Comparison of values of $F(x)$ and $\tilde{F}(x)$.

x	$F(x)$	$\tilde{F}(x)_{\text{comp}}$	$\tilde{F}(x)_{\text{anal}}$
0	2.684
10	0.853	0.849	0.880
20	0.606	0.600	0.623
30	0.491	0.490	0.508
40	0.423	0.424	0.440
50	0.376	0.380	0.394
60	0.341	0.347	0.359
70	0.314	0.321	0.333
80	0.292	0.300	0.311
90	0.275	0.283	0.293
100	0.260	0.268	0.278

where

$$\psi(z) \equiv 4[\frac{1}{2} - I(z)]/z^2, \quad (A3)$$

has been calculated numerically by Simpson's rule in conjunction with the tabulation of Ramsey and Cruse, and is given in the second column in Table I. An idea of the error introduced in the numerical integration can be obtained by comparing the computed value of $F(0)$ with the true value of $F(0)$ given by

$$\int_0^\infty dz \psi(z) = \frac{1}{2}\pi^{3/2} = 2.784, \quad (A4)$$

an error of less than 4%. In the third column of Table I is presented the value of

$$\tilde{F}(x)_{\text{computed}} \equiv (F(0)_{\text{computed}}/x^{1/2}) = (2.684/x^{1/2}), \quad (A5)$$

and we see that this value is very close to that of $F(x)$, the difference being less than 4% for $10 \leq x \leq 100$. It is more convenient for analytic purposes, however, to use the function

$$\tilde{F}(0)_{\text{analytic}} \equiv F(0)_{\text{analytic}}/x^{1/2} = \frac{1}{2}\pi^{3/2}/x^{1/2}, \quad (A6)$$

which is given in the fourth column of Table I.

APPENDIX B

The interaction energy of Eq. (5),

$$-\gamma \cdot \mathbf{E}, \quad (A7)$$

has been used as the part of the Hamiltonian expressing the interaction between molecule and field in the derivation of the present equations of motion. This term is the standard interaction Hamiltonian when the molecule is in a *prescribed* (external) electric field (see, for instance, reference 6, p. 290). If we describe the molecule in terms of its Hamiltonian H_m (which involves the internal coordinates only) and its dipole moment, then Eq. (A7) yields, as the differential equations of motion for the molecule [Eqs. (7) and (8) are integral versions of these]

$$\begin{aligned} \dot{\gamma} &= -(i/\hbar)[\gamma, H_m], \\ \dot{H}_m &= (i/\hbar)[H_m, \gamma] \cdot \mathbf{E}. \end{aligned} \quad (A8)$$

The expression for the absorption of power by the

dipole from the field is formally in accord with Newton's laws of motion and the definition of work. For the case in which the electric field is not prescribed but is considered as a dynamical variable of the system under consideration, we obtain from Eqs. (2) and (5), if we ignore the loss and driving mechanisms,

$$\begin{aligned}\dot{P} &= -(\omega^2/4\pi c^2)Q, \\ \dot{Q} &= 4\pi c^2 P + 4\pi c \mathbf{u} \cdot \dot{\boldsymbol{\gamma}}.\end{aligned}\quad (\text{A9})$$

These equations are also found in I.

Let us now consider the molecular dipole moment as prescribed, and study the behavior of the field. As far as Eqs. (2) and (5) are concerned, it does not matter whether the dipole moment is prescribed or is part of the system under consideration; Eqs. (A9) remain unchanged. However, in the present instance, we need not resort to a Hamiltonian, but may also start with Maxwell's equation and a prescribed localized *current* $\dot{\boldsymbol{\gamma}}$. We obtain then

$$\begin{aligned}\dot{P} &= -(\omega^2/4\pi c^2)Q + (\mathbf{u}/c) \cdot \dot{\boldsymbol{\gamma}}, \\ \dot{Q} &= 4\pi c^2 P,\end{aligned}\quad (\text{A10})$$

which appear different from Eqs. (A9). Before we consider whether the difference is significant, we ask if Eqs. (A10) can be obtained from a Hamiltonian. Had we used an interaction Hamiltonian

$$-\dot{\boldsymbol{\gamma}} \cdot (\mathbf{u}/c)Q$$

instead of (A7), then the equations for the field obtained from this Hamiltonian would have been just Eqs. (A10). Such a procedure is possible, however, only if the dipole current is considered as an external, prescribed, current. Otherwise, $\boldsymbol{\gamma}$ is a dynamical variable, and its time derivative may not occur in the Hamiltonian. We see, then, that if we consider the combined system of molecule and field, and if the molecule is described (phenomenologically) by an energy and dipole moment, then only expression (A7) may be used as the interaction Hamiltonian, and the equations of motion for the field appear somewhat different from Maxwell's equations.

Let us now investigate the difference between the two sets of equations. Equations (A9) lead to

$$\begin{aligned}\ddot{P} &= -\omega^2 P - (\omega^2/c) \mathbf{u} \cdot \dot{\boldsymbol{\gamma}}, \\ \ddot{Q} &= -\omega^2 Q + 4\pi c \mathbf{u} \cdot \dot{\boldsymbol{\gamma}},\end{aligned}\quad (\text{A11})$$

while Eqs. (A10) imply

$$\begin{aligned}\ddot{P} &= -\omega^2 P + (1/c) \mathbf{u} \cdot \partial^2 \boldsymbol{\gamma} / \partial t^2, \\ \ddot{Q} &= -\omega^2 Q + 4\pi c \mathbf{u} \cdot \dot{\boldsymbol{\gamma}}.\end{aligned}\quad (\text{A12})$$

The equations for Q (the magnetic field) are identical. The equations for P differ in the interaction term. If the coupling between the field and molecule is sufficiently weak, then, on the one hand, the interaction term is small, and, on the other, $\partial^2 \boldsymbol{\gamma} / \partial t^2$ is approximately equal to $-\omega^2 \boldsymbol{\gamma}$ (in the case of resonance or near resonance between molecule and field). Under these

circumstances the equations for P become equivalent. Equations (A9) and (A10) are thus equivalent for weak coupling. (It might be pointed out that only in the case of weak coupling is it meaningful to relate parts of the total energy to the field and molecule, respectively. The analysis in the present series of articles is based explicitly on the condition of weak coupling.)

It can be said further that these two sets of equations are equivalent when we are dealing with secular changes occurring in resonance phenomena, changes which are cumulative over a large number of periods, the change during each period being small. This follows from the fact that both equations for the magnetic field are identical, and changes in the electric field show up in the magnetic field within a period. The use of Eq. (5) as the interaction Hamiltonian for the problems of the present series of articles, as well as other problems involving secular effects in resonance phenomena, is therefore justified. If, however, there are large changes during a single cycle, or if we are interested in nonresonant effects (in which the time of interest is a single cycle), then it is not possible to use a Hamiltonian formalism in which the molecule is described phenomenologically by its energy and dipole moment.

It is instructive to see how a phenomenological Hamiltonian may be obtained from a microscopic Hamiltonian. (The following argument has been suggested by Professor Julian Schwinger.) We consider a simple model in which the dipole moment is due to motion of a single particle of mass m and charge e in a potential $\varphi(\mathbf{q})$, the entire molecule being localized about \mathbf{r}_0 in the cavity. The internal (canonically conjugate) coordinates and momenta of the particle are \mathbf{q} and \mathbf{p} , respectively; the mode distribution is assumed to have negligible variation over the molecule. The Hamiltonian for the combined system of single mode and molecule (the loss and driving mechanisms are of no concern here) is

$$H = \frac{1}{2m} \left[\mathbf{p} - \frac{e}{c} \mathbf{u}(\mathbf{r}_0) Q \right]^2 + \varphi(\mathbf{q}) + 2\pi c^2 P^2 + \frac{\omega^2}{8\pi c^2} Q^2. \quad (\text{A13})$$

Neither of our two molecular phenomenological variables, the energy and the dipole moment, appear explicitly in this Hamiltonian. The action for the combined system is given by

$$W = \int_{t_1}^{t_2} dt (\mathbf{p} \cdot \dot{\mathbf{q}} + P \dot{Q} - H). \quad (\text{A14})$$

Consider now the transformation

$$\begin{aligned}\mathbf{q}' &= \mathbf{q}, & \mathbf{p}' &= \mathbf{p} - (e/c) \mathbf{u}(\mathbf{r}_0) Q, \\ Q' &= Q, & P' &= P - (e/c) \mathbf{u}(\mathbf{r}_0) \cdot \dot{\mathbf{q}}.\end{aligned}\quad (\text{A15})$$

Substitution from Eq. (A15) into Eq. (A14) shows that this is a canonical transformation, the new Hamiltonian

being

$$H' = H_f' + H_m' + H_{\text{int}}',$$

where

$$H_f' = 2\pi c^2 P'^2 + (\omega^2/8\pi c^2) Q'^2,$$

$$H_m' = (1/2m) p'^2 + \varphi(\mathbf{q}') + 2\pi e^2 u^2 q'^2,$$

$$H_{\text{int}}' = 4\pi c \mathbf{u} \cdot (e \mathbf{q}') P' = 4\pi c \mathbf{u} \cdot \boldsymbol{\gamma} P'.$$

We see that the transformed Hamiltonian refers to the molecules only in terms of the (transformed) phenomenological variables.

So far there has been no approximation. The approximation consists of identifying H_f' and H_m' as the field and molecular energy, respectively. Such an approximation is justified for sufficiently weak coupling.

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Electron Impact Ionization of Atomic Nitrogen*

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The total cross sections for the ionization of atomic nitrogen by electron impact have been measured between 25 and 750 eV using a modulated molecular beam technique. The neutral beam source was a pulsed dc discharge which dissociated about 20% of the molecules. The number of ions formed in a region defined by the intersection of an electron beam and the modulated molecular beam was compared with the number of ions formed when the neutral beam was partially dissociated. All ions were collected, including those energetic ones formed in the dissociative ionization of molecular nitrogen. The degree of dissociation was measured with a mass spectrometer. From the data, the ratios of the atomic to the molecular ionization cross sections were obtained. The absolute atomic values were calculated by multiplying these ratios by the molecular ionization cross sections measured by Tate and Smith. The atomic cross sections are lower than those calculated by Seaton.

INTRODUCTION

MODULATED molecular beam techniques have been extensively used to study chemically unstable atoms. In particular, many collision properties of atomic hydrogen and of atomic oxygen have been measured, while those of atomic nitrogen have been neglected primarily because of the greater difficulty of dissociating the molecule. We have produced a nitrogen beam in which about 20% of the molecules were dissociated, and have applied it to measure the electron impact ionization cross section of atomic nitrogen between 25 and 750 eV.

EXPERIMENTAL

The method was similar to that previously used to obtain the ionization cross sections for H and O.¹ Since the radio-frequency discharge used in those experiments would not significantly dissociate nitrogen, a new source was required. In addition, a different ionizer was required for some of the data. A schematic arrangement of the apparatus is shown in Fig. 1.

The atomic nitrogen source, a pulsed dc discharge, is shown schematically in Fig. 2. The tube is of Pyrex and the electrodes are aluminum. The beam source was a slit fixed to the tip of the exit tube which was 2 cm in

length. The beam thus originated from the afterglow in the exit tube and not from the discharge column. The slit was sufficiently narrow to satisfy effusive flow conditions. Typically, the discharge operated at 1.6 kV and 50 mA, with a nitrogen pressure of about 0.2 mm Hg. The current pulse, similar in shape to a one-cycle damped sine wave, had a duration of about 26 μsec . The positive and negative peak currents were about 750 and 350 A, respectively. Although the interval between

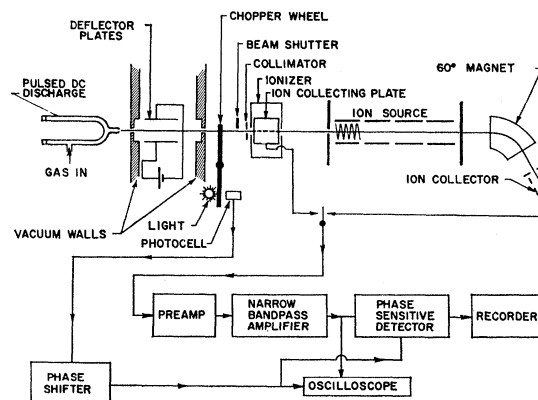


Fig. 1. Experimental arrangement for the ionization of atomic nitrogen by electron impact. A magnetically collimated electron beam intercepts the modulated neutral beam within the ionizer. The ions formed are collected and the resultant ac signal is measured by the electronic system. The mass spectrometer is used to determine the degree of dissociation of the nitrogen beam.

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¹ E. W. Rothe, L. L. Marino, R. H. Neynaber, and S. M. Trujillo, *Phys. Rev.* **125**, 582 (1962).