Steady-State Stimulated Emission in Resonant and Nonresonant Cavities

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The value of the Einstein B coefficient for stimulated emission for a collection of atoms interacting with a radiation field is shown to depend upon the velocity distribution of the excited atoms and also upon the variation of field intensity with frequency. An emission line narrowing phenomenon for radiating matter enclosed in a nonresonant cavity with reflecting walls is predicted. It is shown that a loss mechanism must be introduced if significant line narrowing is to result from a single pass of radiation through an amplifying medium. An additional contributing effect to the mode pushing and mode pulling phenomena in a gas laser is demonstrated. A method of setting a gas laser to a predetermined frequency is described.

1. INTRODUCTION

THE investigations of Planck[1] and Einstein[2] into the problem of blackbody radiation were of great importance to the development of modern physics. This well-known problem involves the frequency distribution of the radiation emanating from a small hole in a heated cavity in thermal equilibrium. The idea of stimulated emission and the A and B coefficients were introduced by Einstein to describe this equilibrium state. The ratio of these two coefficients is known in this theory, but until the development of wave mechanics no method was available for finding their separate values.

In the equilibrium considered by Einstein, the interaction of the radiation field with the cavity walls obscures the sharp emission lines of the enclosed gaseous matter in the cavity. Over the small range of frequencies encompassed by a given emission line of the gaseous enclosed matter, the radiation field in the cavity is essentially constant. For this reason, as we shall show in detail, the B coefficient appears as a constant.

We will consider the problem of a gas in thermal motion interacting with a radiation field whose intensity varies significantly over the range of frequencies associated with a given emission line of the gas. It will be shown that the average value of the B coefficient for the collection of matter depends upon the form of the radiation field intensity function and upon the velocity distribution of the matter. These considerations will be of importance in our discussion of gas lasers and also for the nonresonant cavity to be discussed. Such dependence can also be significant for a gas radiating in free space. As stated, the effect is obscured if the field is constant over the range of frequencies associated with the emission line of the gas.

An emission line narrowing phenomenon is predicted for radiating gaseous matter enclosed by a nonresonant cavity with reflecting walls if the gas has a sufficient population inversion.

All of our work will presuppose that correlation effects between the gas atoms are absent.[3]

A weighting function is introduced to discuss problems involving the interaction of an excited gas with a radiation field. This function is used in a discussion of gas lasers and in the analysis of a postulated effect which contributes to mode pulling and mode pushing phenomena of the sort discussed by Bennett.[4]

An experiment is reported in which the higher order terms in the mode pulling phenomenon are directly measured. Discrepancy between the results of this experiment and the predictions of the Bennett theory is explained by resort to the additional mode pushing effect we have postulated and analyzed.

Several methods of setting a gas laser to a known frequency which are convenient and simple are outlined.

Zeeman and Stark effects are not considered in this paper. The generalization of our work to cases where Zeeman or Stark effects must be considered, however, is direct and simple.

2. THEORY

Let us consider a collection of gaseous matter interacting with a radiation field and focus our attention upon a particular transition in the gas. We shall designate by N_2 the number of gas atoms per unit volume in the upper energy level E_2 and by N_1 those in the lower energy level E_1. We shall ignore all interactions except $2 \rightarrow 1$ or $1 \rightarrow 2$.

In a nonresonant cavity or in free space, the Einstein coefficient for spontaneous decay A_{21} is equal to $(\tau)^{-1}$, where τ is the lifetime of level 2 for the decay $2 \rightarrow 1$ when stimulation effects are absent.[5]

The Einstein coefficient B_{21} represents the probability that a member of N_2 will decay to level 1 and emit a photon of frequency $\nu_0 = (E_2 - E_1)/h$ (h is the Planck constant) when in the presence of a radiation field which appears to have unit energy density at the frequency ν_0 in the rest frame of the atom considered. The Einstein B_{12} coefficient represents the probability that a member of N_1 will absorb such a photon under these conditions. If the field is described in the rest frame of an enclosing cavity, this definition presupposes that the absorbing

[1] M. Planck, Ann. Physik 4, 553 (1901).
[2] A. Einstein, Physik Z. **18**, 121 (1917).
[3] I. R. Senitzky, Phys. Rev. **121**, 171 (1961).
[4] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962).
[5] A. Mitchell and M. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, Cambridge, England, 1934), pp. 94, 99, 100, 161, 268.

atom is stationary in the rest frame of the cavity unless the energy density of the field is constant over the range of Doppler-shifted frequencies which can appear as ν_0 to a moving atom.

We shall initially adopt the viewpoint that an atom can be stimulated to absorb or emit a photon only when radiation satisfying the Bohr condition $E_2-E_1=h\nu_0$ in the rest frame S of the atom is incident on the atom and consider only Doppler-broadening effects. We shall then extend the argument to include the net effect of all processes which symmetrically broaden the emission line shape. Cases where asymmetric broadening effects operate shall not be considered, but the generalization of our work to such cases is direct.

Doppler-shift effects are of considerable importance in problems involving the interaction of radiation with matter.[4] While, in the rest frame S of the atom, any radiation energy emitted or absorbed appears to be of frequency ν_0, this radiation will appear in the rest frame S_0 of the mass center of the radiating gas to have a Doppler-shifted frequency ν. The relativistic connection between ν and ν_0 is[6]

$$\frac{\nu}{\nu_0}=\frac{1+\beta\cos\theta}{\{1-\beta^2\}^{1/2}}. \tag{1}$$

In (1), $\beta=s/c$, where s is the speed of the atom in the S_0 frame, c is the speed of light in the gas, and θ is the angle between the velocity of the atom and the direction of emission of emitted radiation or, in the case of absorption, of incidence of the stimulating radiation. Let us assume $c\gg s$ so that (1) can be approximated by the familiar formula:

$$\nu/\nu_0=1+\beta\cos\theta. \tag{2}$$

With every $(\theta,d\theta)$ we can associate a conical element of solid angle $d\Omega$ whose axis lies along the direction of the velocity of the atom—which we take as the polar axis—in the S_0 frame. This is shown in Fig. 1 below. In Fig. 1, θ is the polar angle and ϕ is the azimuthal angle. Only through the $d\Omega$ associated with an appropriate $(\theta,d\theta)$ can radiation of frequency ν stimulate an absorption or emission process for the atom. The value of $d\Omega$ in steradians is found below:

$$d\Omega=\int_0^{2\pi}(\sin\theta d\theta)d\phi$$
$$=2\pi\sin\theta d\theta. \tag{3}$$

Combining (2) and (3), we obtain

$$d\Omega=2\pi[1-(c/s)^2(\nu-\nu_0)^2/\nu_0^2]^{1/2}d\theta. \tag{4}$$

Let us now assume that the velocity distributions of N_1 and N_2 are isotropic Maxwell-Boltzmann (M-B) distributions. Then we can write for the distributions of speeds of members of N_1 and N_2 the following[7]:

$$n_{1,2}(s)=4\gamma^3N_{1,2}s^2(\pi)^{-1/2}\exp(-\gamma^2s^2). \tag{5}$$

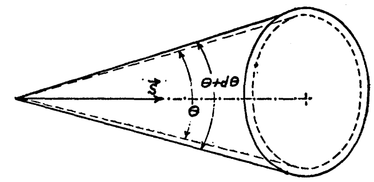

FIG. 1. Solid-angle element associated with stimulation by radiation of frequency ν.

In (5), $\gamma=m(2kT)^{-1}$, where m is the atomic mass, k is the Boltzmann constant, and T is the gas temperature.

Consider the line shape of the radiation from a gas in free space which is associated with a transition for which Doppler broadening is overwhelmingly dominant over other broadening effects. This is a problem which has a familiar treatment in which one merely substitutes (2) into the velocity distribution in order to obtain the change in wavelength due to the Doppler effect of the component of velocity of the emitting atom along the line of sight.[8] We shall, however, treat it by a different method which is more cumbersome in this particular case but has greater power in more difficult calculations.

We consider the problem in free space and assume the radiation field is sufficiently weak that stimulation processes can be ignored. We will obtain the line shape and the intensity of the spontaneously emitted radiation by weighting the Einstein A_{21} coefficient by a frequency-dependent weighting function which we shall designate by $n_2(\nu)$. The function will be seen to bear an intimate relation to the absorption coefficient used, for example, by Milne.[9] We shall generalize, however, to three dimensions and consider the problem in somewhat greater detail. This function, if integrated over ν, must equal N_2. This condition is set by the requirement that our calculation technique, if correct, must give the same result Einstein obtained[2] if $n(\nu)$ is used as a weighting function to replace N in the blackbody radiation problem he considered. The function $n_2(\nu)$ depends upon the thermal distribution of speeds of the radiating matter and involves the solid-angle effect we have considered. The idea of such a function is familiar in a one-dimensional form in absorption problems and seems to be original with Milne.[9] Milne did not explicity evaluate $n(\nu)$ and he assumed that $n_2(\nu)/n_1(\nu)=N_2/N_1$ which we shall show is not valid in general.

Let us evaluate $n_2(\nu)$ and $n_1(\nu)$ assuming that N_2 and N_1 have a M-B distribution of speeds.

$$n_{1,2}(\nu)=\oint\int_0^\infty n_{1,2}(s)\frac{d\Omega}{d\theta}ds. \tag{6}$$

[6] C. Møller, *The Theory of Relativity* (Oxford University Press, London, England, 1952), p. 62.

[7] E. H. Kennard, *Kinetic Theory of Gases* (McGraw-Hill Book Company, Inc., New York, 1938), p. 47.

[8] H. E. White, *Introduction to Atomic Spectra* (McGraw-Hill Book Company, Inc., New York, 1934), p. 419.

[9] E. A. Milne, Monthly Notices Roy. Astron. Soc. **85**, 117 (1924).

Using

$$N_{1,2}=\int_0^\infty n_{1,2}(\nu)d\nu\,, \tag{7}$$

to evaluate the constant $\cancel{c}$ in (6), we obtain

$$n_{1,2}(\nu)=\gamma c N_{1,2}(\pi)^{-1/2}(\nu_0)^{-1}\exp[-\{\gamma c(\nu-\nu_0)/\nu_0\}^2]. \tag{8}$$

Let $\rho(\nu)$ designate the radiation energy density in the radiating gas. When stimulation effects are negligible, $\rho(\nu)$ is

$$\rho(\nu)=\gamma c N_2(\pi)^{-1/2}(\nu_0)^{-1}A_{21}h\nu \times\exp[-\{\gamma c(\nu-\nu_0)/\nu_0\}^2]. \tag{9}$$

Equation (9) has the proper well-known Gaussian shape.[8] This result is of particular interest to us in our study of stimulated emission, since spontaneous emission can be considered to arise from stimulation of emission by a zero-point oscillating field.[10]

If we have an isotropic M-B velocity distribution and if Doppler broadening is not overwhelmingly dominant, then (6) must be replaced by

$$\begin{aligned} n(\nu)=&\frac{\cancel{c}}{\Delta\nu_N+\Delta\nu_L}\\ &\times\int_{-\infty}^{\infty}\frac{\exp[-(2\zeta/\Delta\nu_D)^2\ln 2]}{1+\{[2/(\Delta\nu_N+\Delta\nu_L)](\nu-\nu_0-\zeta)\}^2}d\zeta\\ =&\frac{\cancel{c}}{\Delta\nu_N+\Delta\nu_L}\int_0^\infty ds\,n(s)\\ &\times\int_{\nu^{-1}(\nu-\nu_0-s/c)}^{\nu^{-1}(\nu-\nu_0+s/c)}\frac{\{1+[c/s\{\nu_0-\nu(1-y)^2]^2\}^{1/2}}{1+4y^2/(\Delta\nu_N+\Delta\nu_L)^2}dy, \end{aligned} \tag{10}$$

where $\Delta\nu_N$ is the natural line breadth, $\Delta\nu_D$ is the Doppler breadth, and $\Delta\nu_L$ is the Lorentz breadth.[5] Stark-effect broadening and Holtsmark broadening shall be ignored but the extension of our work to such effects is direct and obvious. In a gas or vapor that is not electrically excited and whose pressure is kept below 0.01 mm, Stark-effect broadening and Holtsmark broadening may be ignored. If a foreign gas is present, however, whose pressure is over about 5 mm, the contribution to the line breadth due to Lorentz broadening can be significant.[5]

Doppler effect and the natural-breadth effect are enentirely independent broadening processes.[5] Consequently, we could calculate the combined result of these effects in (10) by considering every infinitesimal frequency region of (8) to be broadened by the natural linewidth effect. We shall ignore the interdependence of Doppler and Lorentz broadening. This latter simplification enabled us to combine Lorentz broadening with Doppler broadening in (10) in the same way that natural broadening was combined with Doppler broadening.

3. NONRESONANT CAVITY

Let us now investigate the steady-state radiation field inside a nonresonant cavity. We assume the enclosed gas has constant populations of its relevant energy levels and an isotropic M-B velocity distribution. The cavity walls are to be made diffuse reflectors so that the enclosed radiation field will be uniform and isotropic.

The equilibrium condition for the radiation energy density $\rho(\nu)$ in the cavity is obtained by equating energy gains to losses.[2]

$$h\nu B_{21}\rho(\nu)n_2(\nu)+A_{21}h\nu n(_2\nu) =B_{12}\rho(\nu)h\nu n_1(\nu)+L\rho(\nu). \tag{11}$$

The term $L\rho(\nu)$ in (11) brings the reflection losses into the energy balance. In this model, L depends only upon the wall reflectivity r which we assume is constant.

At points on the cavity walls, radiation is incident from one-half the solid angle that it is at interior points. Thus, $\rho(\nu)$ at wall points is one-half $\rho(\nu)$ at interior points. Therefore,

$$L=\tfrac{1}{2}c(1-r)(\text{wall area/cavity volume}). \tag{12}$$

If the cavity is a hollow sphere of radius R, (12) becomes

$$L=(3c/2R)(1-r). \tag{13}$$

The Einstein coefficients must obey the following relationships when thermodynamic equilibrium exists[2]:

$$A_{21}/B_{12}=8\pi h(\nu/c)^3(g_1/g_2)\,, \tag{14}$$

$$B_{21}/B_{12}=g_1/g_2. \tag{15}$$

In the above, g_2 and g_1 are the statistical weights of the upper and lower states, respectively. While the A and B coefficients are related to atomic constants, the B coefficient will be shown to be a constant *only* for atoms which are stationary in the rest frame S_0 if the radiation field varies over the range of frequencies associated with an emission line.

If we substitute for B_{12} and B_{21}, (11) becomes

$$\frac{A_{21}c^3\rho(\nu)n_2(\nu)}{8\pi\nu^2}+A_{21}h\nu n_2(\nu) =\frac{A_{21}c^3\rho(\nu)n_1(\nu)g_2}{8\pi\nu^2 g_1}+L\rho(\nu). \tag{16}$$

We shall henceforth assume that Doppler-broadening effects are overwhelmingly dominant. Then, $n_1(\nu)$ and $n_2(\gamma)$ are expressed by (8). Under this assumption, we

[10] L. Schiff, *Quantum Mechanics* (McGraw-Hill Book Company, Inc., New York, 1955), 2nd ed., p. 400.

can solve (16) to obtain

$$\rho(\nu)=\left[\frac{(\pi)^{1/2}3(1-r)}{2Rh\gamma N_2}\exp\{(\gamma c/\nu_0)^2(\nu-\nu_0)^2\} -\frac{c^3}{8\pi h(\nu)^3}\left(1-\frac{N_1g_2}{N_2g_1}\right)\right]^{-1}. \quad (17)$$

Suppose the background gas has a M-B distribution of speeds. Let the excitation process be such that atoms enter into N_2 and N_1 with a M-B distribution of speeds. In the steady-state this implies that atoms leaving N_1 and N_2 have a M-B distribution of speeds. However, this does *not* imply that the atoms belonging to N_1 and N_2 have a M-B distribution of speeds. This is true even when thermodynamic equilibrium exists.

An argument will be made now to show that in such circumstances the atoms in N_1 and N_2 may have a distribution of speeds which differs from a M-B distribution. The argument will be based upon deducing that the probability for a stimulation process for an atom is a function of the atom's speed.

Consider an atom moving through the isotropic radiation field in the cavity. Such an atom can be stimulated by radiation in the frequency range

$$\nu_0(1-s/c)\leqslant\nu\leqslant\nu_0(1+s/c). \quad (18)$$

Equation (18) reveals the frequency range of the radiation which can stimulate an absorption or an emission process by an atom is directly proportional to the atom's speed. Thus, for the frequency range of the half-width of an emission line, an atom of high speed will have a smaller solid angle through which stimulation can occur than will an atom of low speed. The typical peaking of the radiation field associated with an emission line thus means that a stimulation process is less likely for excited atoms of high speed than for those of low speed. On the other hand, except in a resonant cavity, the probability of a spontaneous decay is independent of the presence of a radiation field. Therefore, for atoms of different speeds considered in the S_0 frame, there are apparent differences in the value of the Einstein B coefficient because of the difference in the relative probabilities of stimulated processes. The weighting functions $n_1(\nu)$ and $n_2(\nu)$, however, give us a means of taking these apparent shifts into the calculation when we know the velocity distribution of N_1 and N_2. If the velocity distributions are unknown, these weighting functions, although not explicitly known, give us a means of finding it by enabling us to compute the rate of change of the probability of stimulating a transition with respect to changes in atomic speed. The values of the Einstein coefficients that apply to an atom stationary in the rest frame of the cavity can be used in our equations if we employ $n_1(\nu)$ and $n_2(\nu)$ as weighting functions since these functions allow us to refer the calculations into the rest frame of the cavity.

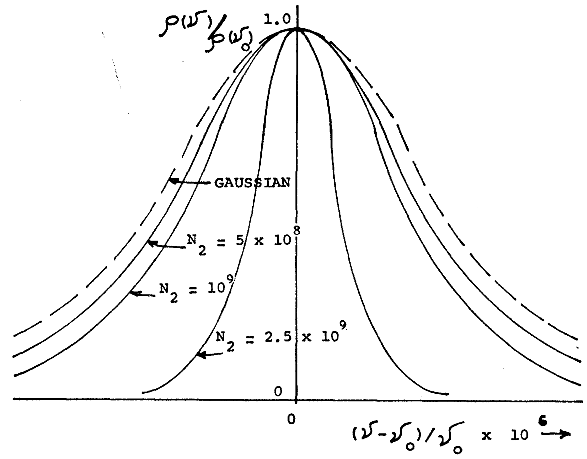

FIG. 2. Neon 6328 Å line shape as a function of N_2 for a particular nonresonant cavity.

Thermalizing interactions tend to bring $n_2(s)$ and $n_1(s)$ to the speed distribution of the background atoms. To the extent that such thermalizing interactions occur, those systems leaving N_1 and N_2 will deviate from the speed distribution of the background gas. However, in a sufficiently intense radiation field, the time between collisions is much larger than the average lifetime in state 2 because of stimulation processes.[11]

Let us now proceed to a detailed consideration of (17). As N_2 increases, the peak value of $\rho(\nu)$ increases and the line narrows. As the net stimulated emission approaches to within an order of magnitude of the losses, this narrowing increases rapidly. This narrowing process is limited by the attainable population inversion.

In Fig. 2 below, (17) is plotted for the 6328 Å line of neon gas located in a cavity having an R of 10 cm and an r of 0.98. In this case, τ is about 10^{-7} sec.[12] Thus, when $N_2g_1\gg N_1g_2$, (17) becomes

$$\rho(\nu)=6.6\times10^{-13}[(2.6\times10^9/N_2)\times\exp\{3.7\times10^{11}(\nu-\nu_0)^2/\nu_0^2\}-1]^{-1}. \quad (19)$$

Notice the very sharp line narrowing that develops as N_2 approaches to within an order of magnitude of 2.6×10^9 atoms/cc.

Let us define $\Delta\nu$ to be the emission line half-width. Then, in terms of $\Delta\nu$, (17) can be written

$$N_2-N_1g_2/g_1=\pi^{1/2}L/(B_{21}h\gamma c)\times[2-\exp\{(\gamma c/\nu_0)^2(\Delta\gamma/2)^2\}]. \quad (20)$$

Equation (20) reveals clearly that an increase in $N_2-N_1g_2/g_1$ leads to a decrease in $\Delta\nu$. For the neon example, (20) becomes

$$N_2-Ng_2/g_1=2.6\times10^9[2-\exp\{4\times10^{-19}(\Delta\nu)^2\}]. \quad (21)$$

[11] A. L. Schawlow and C. H. Townes, Phys. Rev. **113**, 1940 (1958).

[12] W. J. Bennett, Jr., in *Quantum Electronics*, edited by J. Singer (Columbia University Press, New York, 1961), p. 28.

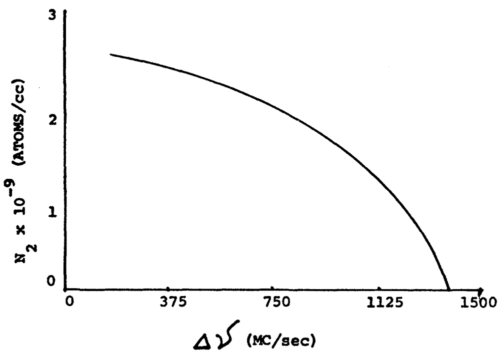

FIG. 3. Neon 6328 Å linewidth as a function of N_2 for a particular nonresonant cavity.

Equation (21) is plotted in Fig. 3 below, assuming $g_1N_2 \gg g_2N_1$.

If we now combine (19) and (17), we obtain

$$\rho(\nu)=8\pi h(\nu/c)^3 \times\left\{-1+\frac{\exp\{\gamma c/\nu_0)^2(\nu-\nu_0\}^2}{2-\exp\{(\gamma c/\nu_0)^2(\Delta\nu/2)^2\}}\right\}^{-1}. \quad (22)$$

In the limit of significant line narrowing, (22) becomes approximately

$$\rho(\nu)=\frac{8\pi h(\nu/c)^3\{\nu_0/(\gamma c)\}^2}{(\nu-\nu_0)^2+(\Delta\nu/2)^2}. \quad (23)$$

The similarity of the shape of (23) to the Lorentzian natural line shape has some interesting implications.

For a homogeneously broadened emission line, a narrowing of the emission line if a population inversion exists also occurs due to the peaking of the Lorentzian line shape of such emission lines and to the fact that the cavity we have assumed is lossy. This lossy characteristic comes from the fact that the walls are imperfect reflectors and that, therefore, radiation of all frequencies is subject to an attenuation. On the other hand, all atoms (as we have shown) are preferentially available for stimulation processes to radiation of frequency ν_0.

The power that must be supplied to the system for the transition we have considered can now be estimated by use of the following approximation:

$$\text{power input/cc}=L\rho(\nu_0)\Delta\nu. \quad (24)$$

For a spherical nonresonant cavity, (24) becomes:

$$\text{power input/cc}=\frac{12(1-r)(\Delta\nu)\lambda_0^{-2}\pi h\nu_0R^{-1}}{-1+[2-\exp\{\gamma\lambda_0)^2(\Delta\nu/2)^2\}]^{-1}}. \quad (25)$$

In (25), λ_0 is the wavelength in the cavity of radiation of frequency ν_0. Figure 4 shows (25) for the example. Notice the typical sharp increase of the required power input/cc as $\Delta\nu$ becomes small.

Let $P(s)$ be the probability per second of stimulating an emission photon from an excited atom of speed s moving in an isotropic radiation field of energy density $\rho(\nu)$. We shall show that $P(s)$ decreases with an increase of s if $\rho(\nu)$ is peaked at $\nu=\nu_0$. We can write that

$$P(s)=K\int_{\nu_0(1-s/c)}^{\gamma_0(1+s/c)}\rho(\nu)[1-\{(c/s)(\nu-\nu_0)/\nu_0\}^2]^{1/2}d\nu, \quad (26)$$

where K is a positive constant.

Letting $y=(\nu-\nu_0)cs^{-1}\nu_0^{-1}$ in (26), we obtain

$$P(s)=Ks\nu_0c^{-1}\int_{-1}^{1}\rho(\nu_0+s\nu_0y/c)[1-y^2]^{1/2}dy.$$

If $\rho(\nu)$ is a constant equal to ξ, this gives

$$P(s)=\tfrac{1}{2}\xi s\nu_0c^{-1}K\pi,$$

which must be equal to$B_{21}\xi$, where B_{21} is the Einstein coefficient for stimulated emission for a stationary atoms $(s=0)$. Therefore,

$$P(s)=2(\pi)^{-1}B_{21}\int_{-1}^{1}\rho(\nu_0+s\nu_0y/c)[1-y^2]^{1/2}dy. \quad (27)$$

For a stimulated absorption, we need only replace $(g_2/g_1)\times B_{21}$ by B_{12} in the above. By assumption, $\rho(\nu)$ is peaked at $\nu=\nu_0$—which corresponds to $y=0$—and decreases as $|\nu-\nu_0|$ increases. Therefore, we see that (27) tells us that

$$\partial P(s)/\partial s<0$$

when $\rho(\nu)$ is peaked at $\nu=\nu_0$, which is what we set out to demonstrate.

We see now that the Einstein B coefficient for a collection of matter in thermal motion can be treated as constant *only* if the radiation field is constant over the range of frequencies associated with a given transition. In addition to the Doppler-shift effects due to the thermal motion of the radiating matter, it is clear that effects

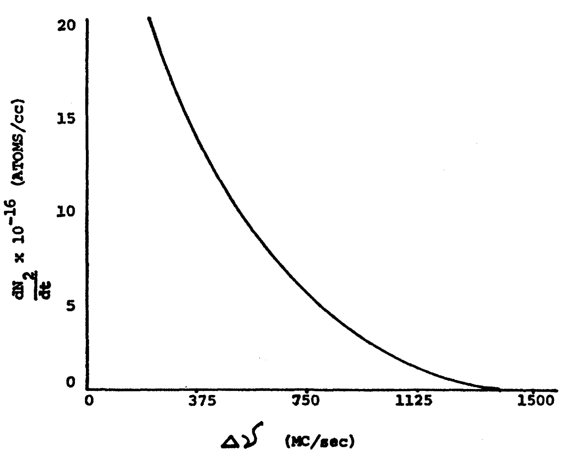

FIG. 4. Rate of creation of members of N_2 as a function of linewidth for a particular nonresonant cavity.

due to the natural line shape and intensity are important. If the cavity walls are merely lossy reflectors or if the gas is in free space the typical sharp emission lines of the gas make such considerations of importance.

As a result of our work, we see that Milne's use of $n_2(\nu)/n_1(\nu)=N_2/N_1$ is correct only when the members of N_1 and N_2 have the same velocity distributions.

4. LINE NARROWING BY SINGLE TRAVERSE THROUGH A REGION WITH AN INVERTED POPULATION

A narrowed emission line can also be obtained by passing a radiated emission line through a region in which a gas with a population inversion is enclosed. This approach, however, is not capable of giving as narrow a line as can potentially be obtained from the nonresonant cavity (provided sufficient population inversions can be obtained) because of saturation-broadening effects. This is so because in this approach all excited atoms in the gas cannot be stimulated to emit by radiation of frequency ν_0. In this technique, radiation of a given frequency propagating through the excited gas can stimulate only certain atoms because of Doppler-shift effects. By the usual line-of-sight argument, (11) describes the gain function for such a traveling radiation field.

The experimental result to be expected is now clear. At first, line narrowing will occur. This narrowing will continue until all atoms available for stimulation by radiation of frequency ν_0 *are* stimulated. After this optimum point, the line will broaden and square off. This occurs as the intensity of the radiation at other frequencies increases to the point where the excited atoms preferentially available at these other frequencies are stimulated to emit radiation centered at these frequencies. This limitation can be defeated by introducing a frequency-independent energy loss mechanism since $n_2(\nu)$ is typically peaked at ν_0.[12a] Such a technique, however, limits the maximum output intensity of the radiation to the incipient saturation value. In the nonresonant cavity, no such limitation on the line narrowing exists since *all* excited atoms in the upper energy level of the transition are preferentially available for stimulation by radiation of frequency ν_0. It may, however, be impossible to produce population inversion in a nonresonant cavity of sufficiently large dimensions to expect line narrowing to develop as large as those that can be obtained in a narrow tube.

5. GAS LASER

The relative gain curve of a gas laser operating on an inhomogeneously broadened line has an essentially Gaussian shape with a dip at the center frequency ν_0. The function $n_2(\nu)$ describes approximately the emission which can be stimulated inside the laser before oscillations start. However, a dip in gain at the center of the relative gain curve when the gas has a single isotope results because only one group of excited atoms in this frequency region can be stimulated to emit radiation, whereas at other frequencies two groups of excited atoms are potential contributors of radiation. This effect results from reflection of the oscillating radiation field at the mirrors of the laser. Obviously, once the laser threshold condition[11] is satisfied, the amplitude of the radiation field at resonant frequencies will build up until limited by energy losses from the cavity and by the ability to excite atoms into N_2. In general, two regions of depletion will be burnt in $n_2(\nu)$ for each oscillating frequency. These regions of depletion in $n(\nu)$ are equivalent to Bennett's "holes" in the relative gain curve. It is the gain coefficient $k(\nu)$ in Beer's law for propagating radiation which is directly proportional to $n_2(\nu) - n_1(\nu)$.[5]

Consider a laser operating on an inhomogeneously broadened emission line. Since the relative gain curve is close to Gaussian, this means the atoms which can be stimulated to emit have a M-B distribution of velocities along the laser axis. Because of the holes in $n_2(\nu)$, $n_2(s)$ is *not* a M-B eistribution of speeds. The Gaussian shape of the relative gain curve, however, is predicted by our analysis. This is so because only those atoms recently excited into N_2 will interact with the oscillating radiation in the laser, all other members of N_2 (neglecting thermalizing) being essentially nonavailable to stimulation processes. These recent arrivals will have a velocity distribution close to that of the background matrix—which should be close to M-B.

6. MODE PULLING AND MODE PUSHING

Mode pulling and mode pushing in gas lasers have been investigated by previous workers.[4,13,14] An additional effect contributing to the mode pulling phenomenon will now be discussed. This effect seems to contribute also to the mode pushing phenomenon.

Consider the interaction of an excited atom with an oscillating radiation field inside the cavity of a gas laser. Because of Doppler-shift effects, such an interaction involves the thermal motion of the excited atom with respect to the cavity, as we have shown. Before any oscillations commence, any stimulated radiation will have a Lorentzian distribution centered at the Doppler-shifted value corresponding to s in the rest frame of the emitting atom.

In a gas laser, emission into an oscillating field is al-

[12a] *Note added in proof*. The author, in a manuscript which has been submitted for publication, has shown it is possible when proper losses are introduced—both in the single traverse case and also in certain types of resonant and nonresonant cavities—to achieve output half-widths only 2 orders of magnitude greater than gas laser half-widths and which are sharply defined at ν_0, the center frequency of the emission line.

[13] C. H. Townes, in *Quantum Electronics*, edited by C. H. Townes (Columbia University Press, New York, 1960), p. 3.

[14] J. P. Gordon, H. J. Zeiger, and C. H. Townes, Phys. Rev. **99**, 1264 (1955).

most completely due to stimulation.[15] An atom stimulated to emit into such a field will donate its energy primarily at the frequency of the oscillating field ν_m and in phase with it.[15] This is true despite the fact that if the atom were to spontaneously emit energy into the direction involved, this energy might be centered at a different frequency ν.[16]

Let us designate the center frequency of a resonance of a Fabry-Perot interferometer by ν_c. The response of such a cavity is a symmetric function of $\nu-\gamma_c$.[17] In general, however, $n_2(\nu_c)$ will have a nonzero slope. Thus, most of the emission into the oscillating field is stimulated from excited atoms that would *spontaneously* emit into the direction of the oscillating field radiation of frequencies greater or less than ν_c.

Mode pulling effects are typically of the order of 10^3–10^5 cycles.[4] Thus, the following picture of the build-up of the oscillating field in an active laser is reasonable. Because $\Delta\nu_c$ and $\Delta\nu_N$ are of the order of megacycles, the half-width of the oscillating field during the early stages of its build-up is at least of the order of tens of kilocycles. The final frequency of the oscillation will thus not be ν_c necessarily, but will be set by the most favorable gain balance between the passive cavity response and the relative gain with respect to frequency of the laser. The cavity response of the *active* cavity may thus differ from that of the *passive* cavity. This effect gives a contribution to mode pulling which is independent of the contribution due to the anomalous dispersion effect.

We shall make the assumption that the excited atoms have a Maxwell-Boltzmann velocity distribution along the spatial mode of the oscillation. Then, as we have already seen, the distribution of potential contributors of spontaneous emission of frequency ν is approximately equal to

$$n_2(\nu)=\gamma c(\pi)^{-1/2}\nu_0^{-1}N_2\ \exp[-\{\gamma c/\nu_0(\nu-\nu_0\}^2]. \quad (28)$$

Equation (28) assumes that the line is inhomogeneously broadened and also that an isotopic mixture is present in the laser which obscures the drop in the center of the relative gain curve assignable to each isotope.[18] If only a single isotope directly participates in the laser action, then the Gaussian factor in the numerator of (10) must be corrected to take account of this effect. If several isotopes are present, the relative gain curve of the laser will be the weighted sum of the $n(\nu)$ functions for the various isotopes.

The relative probability for stimulating emission centered at frequency ν_c into a given direction from an atom which would *spontaneously* emit radiation centered at ν into that direction is given by the Lorentzian[4]

$$[1+\{2(\nu-\nu_c)/(\Delta\nu_N+\Delta\nu_L)\}^2]^{-1}. \quad (29)$$

The mirror reflectance coefficient varies by the order of magnitude of a factor of 2 over 1000 Å.[4] This variation introduces a completely negligible effect over the width of an emission line.

The expression for the cavity response of a Fabry-Perot interferometer is an Airy function.[17] If the finesse of such a device is high, this function becomes very close to a Lorentzian. The cavity of a gas laser must have such a high finesse in order to achieve oscillation. In terms of $\Delta\nu_c$, the half-width of the passive cavity response, the passive cavity response of a gas laser is thus approximately

$$[1+(\nu-\nu_c)^2/(\Delta\nu_c/2)^2]^{-1}. \quad (30)$$

Mode pulling effects are small in comparison to the half-width $\Delta\nu_c$.[4] Thus, the cavity response will be relatively flat over the region of mode pulling since it is near the maximum of the passive cavity response. This is not true, however, of the relative gain curve of the laser in the neighborhood of ν_c unless the gain curve has a local maximum or minimum.

Let us assume that he excited atoms in N_2 have a M-B distribution of velocities along the laser axis. Then, the contribution to the quantity $(\nu_m-\nu_c)$—which is known as the mode pulling—which is due to the effect we are considering is equal to the following:

$$\int_{-\nu_c}^{\infty}(\nu-\nu_c)\exp[-\{\gamma c/\nu_0(\nu-\nu_0)\}^2]\times[1+(\nu-\nu_c)^2/(\Delta\nu_c/2)^2]^{-1}d\nu \div\int_{-\nu_c}^{\infty}\exp[-\{\gamma c/\nu_0(\nu-\nu_0)\}^2]\times[1+(\nu-\nu_c)^2/(\Delta\nu_c/2)^2]^{-1}d\nu. \quad (31)$$

Expression (31) for the contribution of this effect to $\nu_m-\nu_c$ can be written since the cavity resonance is an effect independent of the emission line broadening

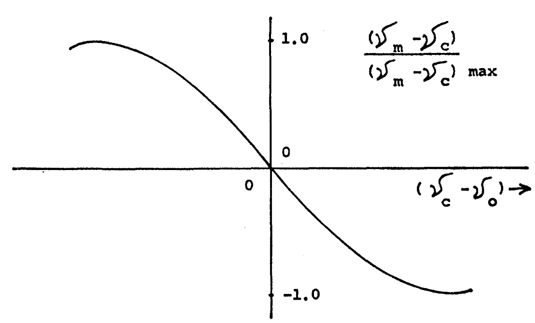

FIG. 5. Contribution to mode pulling phenomenon in a gas laser of effect considered for an isotopic mixture.

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[15] O. S. Heavens, Appl. Opt., Suppl. **1**, 1 (1962).
[16] W. R. Bennett, Jr., Appl. Opt., Suppl. **1**, 24 (1962).
[17] M. Born and E. Wolf, *Principles of Optics* (Pergamon Press, Inc., New York, 1959), p. 327.
[18] A. Szoke and A. Javan, Phys. Rev. Letters **10**, 521 (1963).

effects we have considered. An approximation can be made to solve (31), and we thereby obtain

$$(\Delta\nu_c/2)\tan\{(\gamma c/\nu_0)^2(\Delta\nu_c)(\nu_0-\nu_c)\}. \qquad (32)$$

Expression (32) is plotted in Fig. 5. Notice that (32) is zero for $\nu_c=\nu_0$. Thus, (32) satisfies a necessary condition imposed by symmetry considerations.

When a laser with a single active isotope is operated at high power at a frequency near ν_0, the type of contribution to mode pulling to be expected from the effect considered is that shown in Fig. 6.

In terms of the Doppler half-width, $\Delta\nu_D$, (32) is

$$(\Delta\nu_c/2)\tan[\{2\ln 2/(\Delta\nu_D/2)^2\}(\Delta\nu_c/2)(\nu_c-\nu_0)]. \qquad (33)$$

Suppose several frequency modes are oscillating in the same spatial region. Then holes will be burnt in the local $n(\nu)$. By changing the local slope of $n(\nu_c)$, the mode pulling that they individually experience is affected since the most favorable gain location is changed. Thus, it is clear that the mechanism we have discussed is responsible for "hole repulsion" effects of the type considered by Bennett.[4] These holes will be burnt at the frequencies $\nu_0\pm\nu_m$ due to the reflection effects discussed by Bennett.[4] This effect, of course, applies only when holes in $n_2(\nu)$ overlap.

If $\Delta\nu_c=1$ Mc/sec, then for the 1.1523 μ line of a helium-neon laser, (33) gives a mode pulling contribution of 2.7 kc/sec when $\nu_c-\nu_0=\Delta\nu_D/2$, and two modes symmetrically positioned about ν_0 would be pulled together by 5.4 kc/sec. Somewhat larger hole repulsion effects are possible and can be tied into laser frequency stabilization schemes as we shall show later.

We have excluded any consideration of the degeneracies which can lead to Zeeman and Stark effects in our discussion of lasers and mode pulling. It is worth making a remark about Zeeman effects, however, because of the inherent interest of such effects.

Consider a gas laser lacking Brewster angle windows. If the upper level of the transition is degenerate, in the presence of an appropriate magnetic field the levels corresponding to different magnetic quantum members will separate. The transitions from these separated levels to the lower levels will have different energy values and polarizations.[5] In fact, if the population inversion is sufficient, two oscillating fields slightly separated in frequency will appear in the laser for each cavity resonance.[19] Again, the effect we have considered is a contributor to this splitting.

The functions $n_2(\nu)$ for each of these levels separated by the applied magnetic field will be somewhat displaced from each other in frequency space. The local slopes of these functions will therefore be different at ν_c. Thus, our model predicts that the contribution to the mode pulling of the effect considered will be different for each of these separated levels. This frequency

[19] H. Statz, R. Paananer, and G. F. Koster, J. Appl. Phys. 33, 2319 (1962).

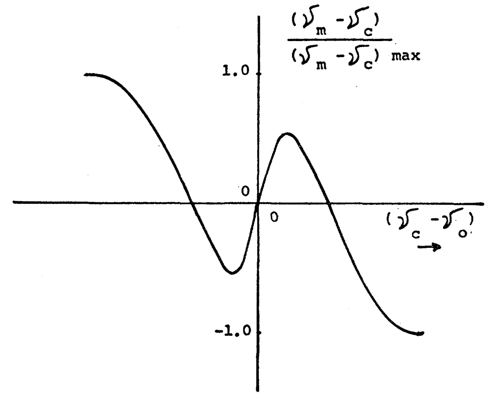

FIG. 6. Contribution to mode pulling phenomenon in a gas laser of effect considered for a single isotope.

difference will be a function of the intensity and direction of magnetic field.

Bennett obtained the following expression for the mode pulling due to anomalous dispersion effect for an inhomogeneously broadened line near ν_0 (in our notation[16]):

$$\nu_m-\nu_c=\frac{2(\ln 2)^{1/2}}{\pi^{1/2}}\left(\frac{\Delta\nu_c}{\Delta\nu_D}\right)(\nu_0-\nu_c)+\frac{8(\ln 2)^{3/2}}{3(\pi)^{1/2}}\frac{\Delta\nu_c}{(\Delta\nu_D)^3}(\nu_0-\nu_c)^3. \qquad (34)$$

If we compare this with (33) in the neighborhood of ν_0, the effect we have discussed is a much smaller contributor to mode pulling than the anomalous dispersion effect discussed by Bennett. However, it does give rise to a detectable mode *pushing* effect. An experiment which directly detected this mode pushing effect will be described further on in this paper.

7. MODE PULLING EXPERIMENT

In the experiment to be described, a convenient means of frequency tuning a gas laser was required. The means adopted was a thin dielectric plate mounted in the cavity on an indexed rotatable table. The plate employed was a microscope slide cover glass of 0.147-mm thickness and index of refraction 1.5 and was selected for minimum prismatic angle by observing fringes under monochromatic light. The table was adapted from a Spencer (American Optical Company) spectrometer, model No. 10025. The position of this table could be read to ± 0.5 min.

Consider the change in frequency of an oscillating mode of a gas laser as the plate undergoes a small rotation (up to a maximum of 20 min) from the Brewster angle. The optical path in the cavity changes from L_1 to $L_1+\Delta L$ and the frequency of the oscillating mode changes from ν_1 to $\nu_1+\Delta\nu$. If the number of wavelengths traveled by the oscillating field in a single round trip around the cavity is unaltered, we can write

$$\frac{2L_1\nu_1}{c}=\frac{2(L_1+\Delta L)(\nu_1+\Delta\nu)}{c}, \tag{35}$$

which yields:

$$\Delta L/L_1=-\Delta\nu/\nu_1. \tag{36}$$

We have assumed ΔL arises only from the rotation of the tuning plate. Therefore, ΔL is a function of the variable angle ϕ between the normal to the plane and the direction of incidence of the oscillating field in the cavity and is also a function of the constant plate thickness D and the constant (for a given emission line) index of refraction n of the plate. The value of $\Delta\phi$ for any rotation of the tuning plate is equal to the angle through which the indexed table is rotated and can thus be read directly.

We can write

$$\Delta L=(\Delta L/\Delta\phi)\Delta\phi. \tag{37}$$

Combining (36) and (37) gives

$$\Delta\nu/\nu=-(1/L)(\Delta L/\Delta\phi)\Delta\phi. \tag{38}$$

The expression for $dL/d\phi$ is well known from the theory of compensating plates as used in Michelson interferometers to be

$$dL/d\phi=-D(d/d\phi)\{(n^2-\sin^2\phi)^{1/2}-\cos\phi\}. \tag{39}$$

Combining (38) and (39) and integrating gives

$$\ln\{(\nu_1+\Delta\nu)/\nu_1\}=-(D/L)\times\{(n^2-\sin^2\phi)^{1/2}-\cos\phi\}\big|_{\phi_1}^{\phi_1+\Delta\phi}. \tag{40}$$

It would also have been possible to adapt a Kösters-type compensator[20] to frequency tune the laser. In that case, two microscope slide cover glasses of similar prismatic angles would be inserted at the Brewster angle into the laser cavity but set in opposite directions. If one prism is moved along the bisector of the prism angle a distance Δh, then an analysis similar to the above yields the expression (for a prism angle α):

$$\ln\{(\nu_1+\Delta\nu)/\nu_1\}=2(L\cos\phi)^{-1}\sin(\alpha/2)(n-1)(\Delta h). \tag{41}$$

This latter technique offers the promise of an order of 1 order of magnitude greater sensitivity than the tuning plate method actually used in the experiment.

If a gas laser is excited such that only a single frequency mode can oscillate, then laser action can occur only over the frequency range

$$\nu_a\leqslant\nu\leqslant\nu_b. \tag{42}$$

To simplify the discussion, let us initially assume that the frequency range given by (42) is such that only a single frequency mode can show laser action in the TEM_{00} spatial mode. We shall also assume the tuning plate is inserted inside the gas laser cavity at the Brewster angle to the field. By observing the positions of the indexed table as the laser goes on and off, the ϕ_a and ϕ_b corresponding to ν_a and ν_b, respectively, can be determined. Using this information and employing (40), the frequency of the laser mode can be set to ν_0 or to any other predetermined frequency in the range given by (42). This procedure directly determines the loss line of the laser also.

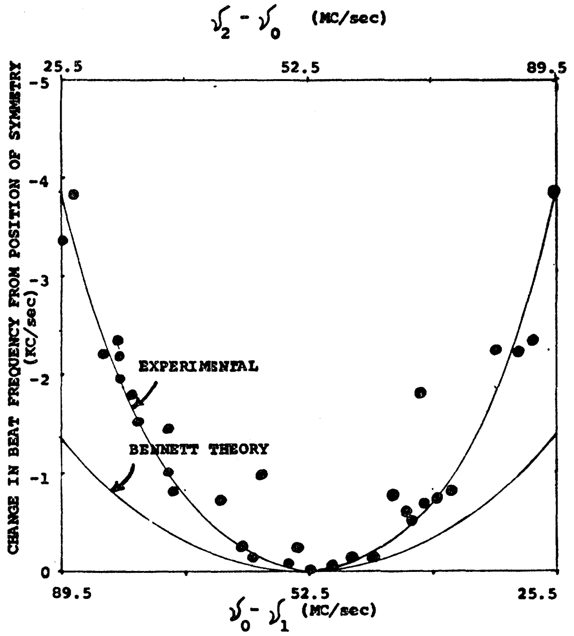

FIG. 7. Comparison of Bennett's theory with mode pulling experiment.

Let us now consider a situation where the excitation level of the laser is set so that two but not three frequency modes can simultaneously laser. For simplicity, assume that only a single isotope of the material showing laser action is present so that the gain curve is essentially symmetric about ν_0. We shall restrict ourselves to situations where the two simultaneously oscillating frequency modes are close to being symmetrically positioned about ν_0. If the laser is operated close to threshold for these modes, hole burning effects should be quite small as indicated by the results of Szöke and Javan.[21]

In the experiment, two simultaneously oscillating frequency modes operated under the above conditions were focused on the detector surface of a 7102 photomultiplier tube and the beat frequency was electronically detected by a heterodyning technique.[22] The geometry was confocal with a 140-cm mirror separation enclosing a 1-m helium-neon filled laser tube with Brewster angle windows. A tuning plate was in

[20] C. Candler, *Modern Interferometry* (The University Press, Glasgow, Scotland, 1950), p. 474.

[21] A. Szöke and A. Javan, Phys. Rev. Letters **10**, 521 (1963).

[22] A. T. Forrester, R. A. Gudmundsen, and P. O. Johnson, Phys. Rev. **99**, 1691 (1955).

the cavity. The change in the beat frequency was observed as the tuning plate was rotated through small angles. By changing the local oscillator frequency, the fact that this beat frequency was a maximum in the symmetric position of the oscillating modes about ν_0 was determined. The experimental data obtained are shown in Fig. 7 and a schematic of the equipment in Fig. 8. In Fig. 7, we also show the predictions of Bennett's theory[4] for the $1.1523\,\mu$ line of neon investigated in this experiment (assuming hole repulsion is absent) for the cavity Q of the laser used. The experimental data indicate that either mode pushing is significant even just above threshold or that there is some error in Bennett's result. The atter possibility cannot be ruled out since Bennett's results are based upon best fits to curves, but the data were taken close enough to the line center such that Bennett's results should be close to reality. If then we accept the conclusion that mode pushing is responsible for the discrepancy between our data and the predictions of Bennett's theory, the fact that we operated close to threshold indicates very shallow holes were responsible for this discrepancy. This means an effect due to the local slope of the depleted gain curve is responsible. The size of the discrepancy and the fact that the local slope of the depleted gain curve is responsible means our previous calculation of an additional mode pushing effect due to the local slope of the gain curve is justified since the size of the discrepancy is just what would be expected from our theory. This is so since in the asymmetrical situation, one hole will be closer to the laser loss line than the other and thus effectively operating in the hole created by the other mode while exerting no reciprocal effect.

It is worth pointing out that the technique involved in this experiment can be employed to detect successively higher order mode pulling effects. Suppose the excitation level is set so that four but not five frequency modes can oscillate simultaneously in the TEM_{00} spatial mode:

$$\nu_1<\nu_2<\nu_3<\nu_4,$$

and the beat frequency between ν_1 and ν_4 is observed when these modes are symmetrically displaced about ν_0. In that case, higher order terms in Bennett's mode pulling results can be compared with experiment and corrected. Incidentally, if three modes are showing laser action in a single isotope laser, and if the beat frequency between ν_1 and ν_3 is set to a minimum, then ν_2 should be very close to ν_0 if the gain curve is symmetric as can be seen from Fig. 6. This effect gives an error signal of $\sim$1 kc/sec per Mc/sec displacement of ν_2 from ν_0. If the excitation level is lowered until ν_1 and ν_3 drop out, ν_2 will be the frequency of the sole remaining oscillating mode. It should also be pointed out that at the symmetrical location of ν_1 and ν_3 about ν_0, a minimum mode pushing effect on the beat frequency between these modes will be observed as the excitation level is varied.

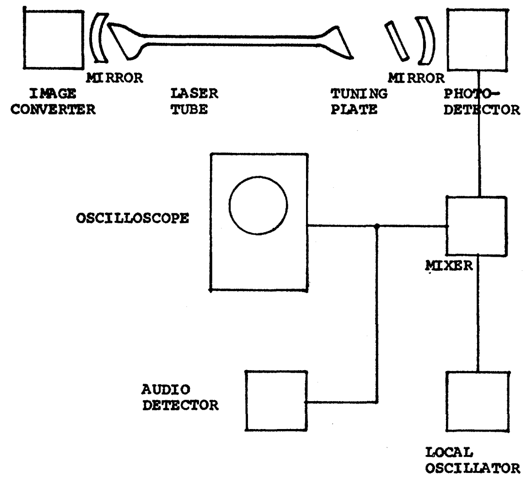

FIG. 8. Schematic of apparatus used in mode pulling experiment.

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