Effects of Collisions on the Saturation Behavior of the 6328 Å Transition of Ne Studied with a He-Ne Laser*

R. H. CORDOVER† AND P. A. BONCZYK‡

Department of Physics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

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The power output of a single-mode 6328 Å He-Ne laser is studied as a function of cavity tuning and for different values of gas pressure. A theory due to Szöke and Javan is used to interpret the results. Two parameters γ and γ', which characterize a homogeneous line shape in the atomic polarization to third order, are determined as a function of the total pressure P. The results for two different gas mixtures are as follows (in units of MHz/Torr): $d\gamma/dP=22\pm7$ and $d\gamma'/dP=58\pm13$ for an 8:1 He^3-Ne^{20} mixture; $d\gamma/dP=12\pm4$ and $d\gamma'/dP=39\pm9$ for a 5.5:1 He^3-Ne^{22} mixture. Linear extrapolation of γ and γ' to zero pressure yields the radiative linewidth $\gamma_N=13\pm12$ MHz. An asymmetry Δ in the frequency dependence of the power output is determined as a function of P, with the result that $d\Delta/dP=2.3\pm0.6$ MHz/Torr. The shift of the minimum of the power dip is toward increasing frequency for both mixtures.

1. INTRODUCTION

It is well known that the power output of a gas laser as a function of cavity tuning may exhibit a sharp dip.[1,2] Conditions for the dip to appear are given explicitly by Lamb.[3] The dip reflects the nonlinear properties of the gaseous medium when coupled to a standing-wave optical-frequency (SWOF) field. The presence of the dip allows one to extract information concerning a line shape associated with an individual atom which, in general, is interacting with the laser field, radiating spontaneously, and colliding with neighboring atoms.

In Lamb's original theory,[3] the power output versus the frequency of the SWOF field is estimated, with allowance for radiative decay but neglect of the effects of atomic collisions. The induced polarization is evaluated up to terms of third order. The latter have a sharp Lorentzian frequency dependence centered at the exact resonance frequency of the atomic transition and have a width $\gamma_N=\frac{1}{2}(T_a^{-1}+T_b^{-1})$, where T_a and T_b are the lifetimes for radiative decay of the upper and lower laser levels, respectively. However, appreciable departure from this width is observed when the collision rate approaches or exceeds γ_N.

In a preliminary theory of the influence of collisions on the saturation behavior of a Doppler-broadened atomic resonance,[4] it is shown that the laser power output versus tuning may be better accounted for if the frequency behavior of the third-order polarization is described by two line-shape parameters γ and γ', where γ determines the magnitude of a Lorentzian, and γ' determines its width. Further, a parameter Δ is introduced to account for a slight asymmetrical behavior of the power output versus tuning.

A recent detailed theory of Gyorffy, Borenstein, and Lamb[5] predicts a frequency behavior for the power output consistent with the foregoing. However, this theory yields details concerning the pressure dependences of the quantities γ and γ', which are not available from the largely preliminary work of Szöke and Javan.

Preliminary results of the present work were presented earlier.[6]

2. EXPERIMENT

A Pyrex tube having quartz windows oriented at Brewster's angle was used. The tube was evacuated to 5×10^{-8} Torr before filling. The purity of the Ne isotopes was 99.8%; the He isotope was of comparable purity. The partial gas pressures were measured with a variable capacitance-type manometer with initial calibration against a McLeod gauge. The uncertainties in the final partial pressures were estimated to be 5%. A Pirani-type gauge, fixed to the tube, and calibrated as above, monitored the total pressure P once the tube was sealed off and removed from the filling area. Any change of P over the course of the measurements was estimated not to have exceeded 5%. The pressure P was varied by changing the gas volume with a pistonlike assembly. This allowed a change of P up to a factor of 2.

A 10-cm length of discharge was excited by a static voltage. For a single-mode operation, the discharge current ranged from 5–20 mA.

The mirror spacing of 20 cm gave a 750-MHz axial mode spacing. Off-axis modes were not present; their introduction required a deliberate mirror misalignment.

The Doppler width was 900 MHz based on an estimated gas kinetic temperature of 400°K.

The frequency of oscillation was varied by changing the cavity length. This was done with a piezoelectric device which moved one of the mirrors. Application of

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† Present address: American Science Corp., 1501 Broadway, New York, N. Y. 10036.

‡ Present address: Physikalisches Institut der Universität, 53 Bonn, Nussallee 12, Germany.

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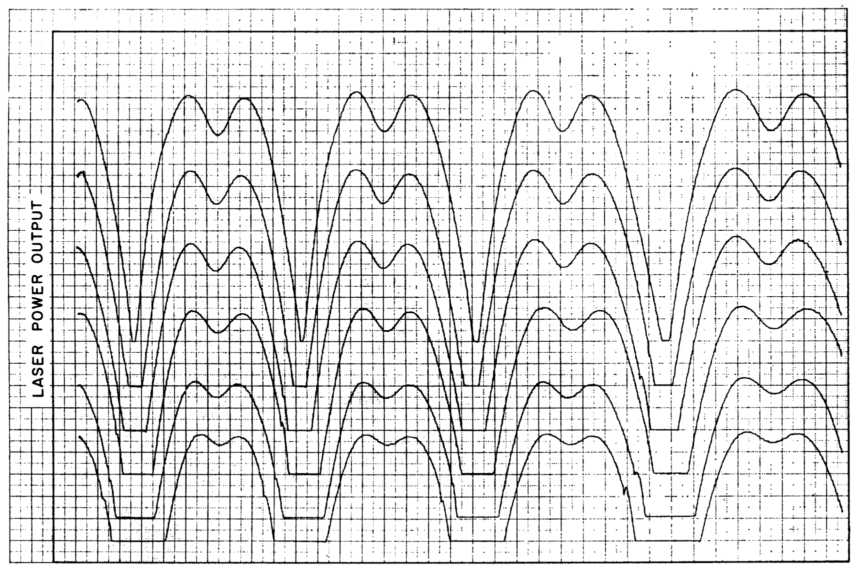

FIG. 1. Laser power output is given as a function of cavity tuning for six levels of discharge excitation. The data are for $P=1.60$ Torr and for the 8:1 mixture.

500 V to this transducer allowed tuning through 3 adjacent modes. The cavity length versus transducer voltage had a 3% nonlinearity. However, this was wholly reproducible and was compensated for in the data handling.

The discharge was shielded from the earth's magnetic field. The residual or uncancelled component of this field along the laser axis was 0.1 G. The effect of this residual field on the data was negligible.

A periodic check was made for simultaneous laser oscillation at 3.39 μ. A mechanical chopper, within the cavity, periodically interrupted laser oscillation. Spontaneous emission at 3594 Å, originating from the lower level of the 3.39-μ transition in Ne, was viewed at 90° to the laser axis. The light intensity at 3594 Å was observed to be unaffected by the chopping, indicating, as desired, no oscillation at 3.39 μ.

The drift of the frequency of the laser was about 150 kHz/sec, and the time required to measure the dip was about 15 sec.

The laser power was measured with a photomultiplier. An X-Y recorder was used to plot the power versus tuning.

A tube of 2 mm i.d. was filled with an 8:1 He^3-Ne^{20} mixture, and data were taken for P between 1 and 2 Torr. Further, a tube of 1 mm i.d. was filled with a 5.5:1 He^3-Ne^{22} mixture, and data were taken for P between 2 and 4 Torr. Two different mixtures were studied in order to attempt to separate Ne*-Ne from Ne*-He collisions. Very pure isotopes of Ne were used in order to prevent the known dip distortions due to isotope shift.[2] Different Ne isotopes were used for the two mixtures in order to be absolutely certain that the observed asymmetry Δ was not a residual effect of isotope shift. The sign of the asymmetry was observed to be the same for both mixtures. Hence, the asymmetry is a collision-induced effect.

A sample of our data is given in Fig. 1.

3. THEORY

Earlier, Javan[7] pointed out how to allow for the effects of collisions in the Lamb's theory.[3] One takes the following modified expression for the imaginary part of the susceptibility of the medium:

$$\chi''=\alpha\left[1-\beta(E)^2\left(1+\frac{\gamma\gamma'}{(\gamma')^2+(\omega-\omega_0-\Delta)^2}\right)\right]\times\exp[(\omega-\omega_0)/\omega_D]^2, \quad (1)$$

where α and β are constant factors. In the limit of zero pressure, where $\gamma=\gamma'=\gamma_N$ and $\Delta=0$, this expression agrees with Lamb's. This expression also gives the gain within constant factors. If gain is set equal to loss and one solves for E^2 in Eq. (1), the power output is then easily shown to be

$$I(\omega)=K\{G_0-(L)\exp[(\omega-\omega_0)/\omega_D]^2\}\times\left[1+\frac{\gamma\gamma'}{(\gamma')^2+(\omega-\omega_0-\Delta)^2}\right]^{-1}. \quad (2)$$

TABLE I. Dependence of γ and γ' on total gas pressure P.

Mixture	$d\gamma/dP$ (MHz/Torr)	$d\gamma'/dP$ (MHz/Torr)
8:1 He^3-Ne^{20}	22±7	58±13
5.5:1 He^3-Ne^{22}	12±4	39± 9

[7] A. Javan, in *Quantum Optics and Electronics*, edited by C. DeWitt, A. Blandin, and C. Cohen-Tannoudji (Gordon and Breach Science Publishers, Inc., New York, 1965), p. 390.

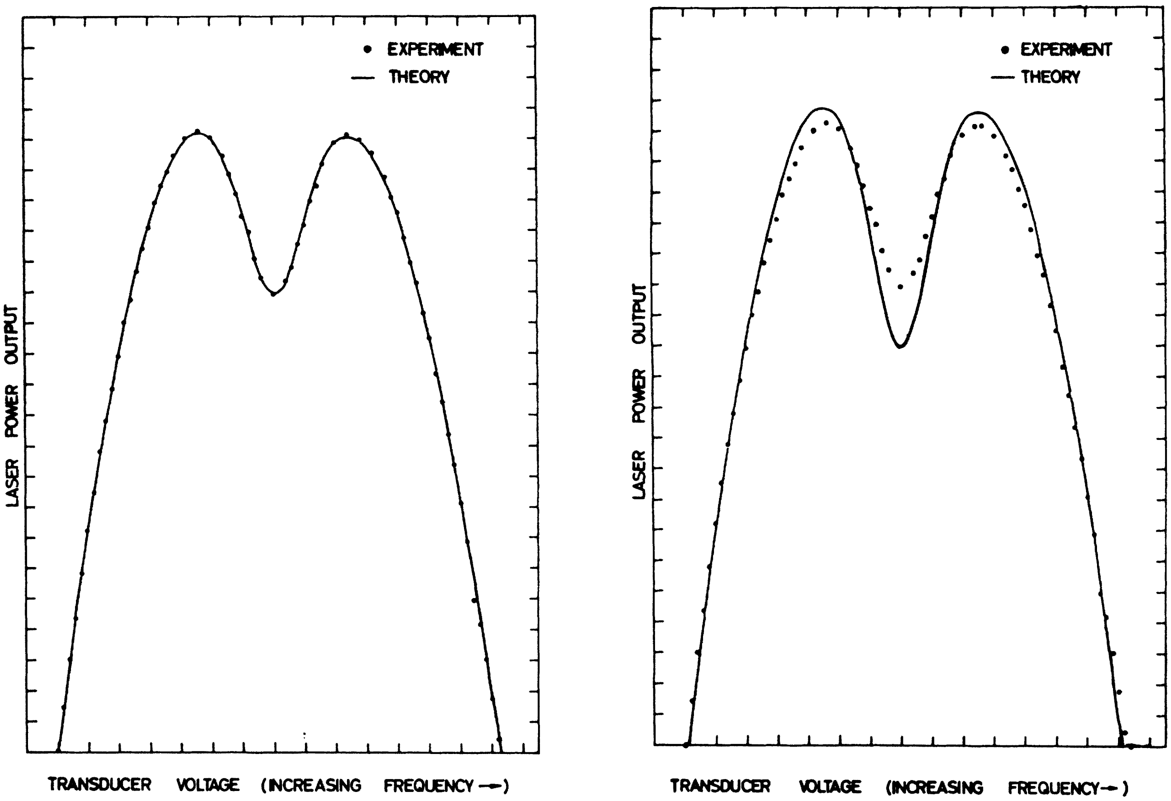

FIG. 2. (a) Best fit of power output versus cavity tuning is given using Eq. (4). The data are for $P=1.18$ Torr and for the 8:1 mixture. The values $A=713$, $B=0.830$, $C=0.0798$, $D=0.656$, and $E=0.00173$ and Eq. (4) determine the theoretical curve. (b) Best fit of power output versus cavity tuning is given using Eq. (4), but with $D=1$. The experimental curve is the same as in (a). The values $A=785$, $B=0.835$, $C=0.0686$, and $E=0.00146$ and Eq. (4) determine the theoretical curve. Note the relatively poor agreement between experiment and theory in the vicinity of the dip.

This result is essentially that of Szöke and Javan.[4] If in the calculation of χ'', the integration over atomic velocities is extended to include terms of order $[(\omega-\omega_0)/\omega_D]^2$, the following final form is obtained for the power output:

$$I(\omega)=K\{G_0-(L)\exp[(\omega-\omega_0)/\omega_D]^2\}$$

$$\times\left\{[1-(2/\pi^{1/2})(\gamma'/\omega_D)]+\frac{\gamma\gamma'[1+(2/\pi^{1/2})(\gamma'/\omega_D)]}{(\gamma')^2+(\omega-\omega_0-\Delta)^2}\right\}^{-1}. \quad (3)$$

The unsaturated gain is G_0, the loss is L, and K is a constant factor. The frequency of the SWOF field is ω, and the exact resonance frequency of the atomic transition is ω_0. The Doppler width is $\omega_D=\omega_0(\bar{v}/c)$, in which $\bar{v}$ is the most probable atomic velocity; γ, γ', and Δ have been defined.

4. RESULTS

The data are fit by computer to the expression[8]

$$I(X')=A[1-(B)\,e^{(X')^2}]$$

$$\times\left\{[1-(2C/\pi^{1/2})]+\frac{C^2D[1+(2C/\pi^{1/2})]}{C^2+(X'-E)^2}\right\}^{-1}, \quad (4)$$

in which $A=\text{const}$, $B=L/G_0$, $X'=(\omega-\omega_0)/\omega_D$, $C=\gamma'/\omega_D$, $D=\gamma/\gamma'$, and $E=\Delta/\omega_D$; A, B, C, D, and E are varied for a best fit. The fit of the data to $I(X')$ is excellent. Differences between theory and experiment in the vicinity of the dip do not exceed 1%. Such a comparison is made in Fig. 2(a).

Both γ and γ' are required to fit the data properly. This may be shown clearly by setting $D=1$ ($\gamma=\gamma'$) and then attempting to fit data with the resulting modified expression for $I(X')$. The fit, in this case, is poor,

[8] Although not indicated in Ref. 4, in fact, an expression identical to that given by Eq. (4) in the text above was fit to the data for Ref. 4. Hence, the results for γ, γ', and Δ in Ref. 4 are appropriate to Eq. (4) above. A. Szöke (private communication).

especially in the vicinity of the dip. An example of this is given in Fig. 2(b).

The dependences of γ and γ' on total pressure are given in Fig. 3. The error accompanying individual datum points is due largely to uncertainties concerning pressure. In fitting the set of straight lines to the data optimally, the constraint is imposed that all four extrapolations to zero pressure have a common intercept, from which it follows that $\gamma_N = 13\pm12$ MHz. The results are summarized in Table I. These values are unchanged from those given earlier by us.[6]

Data for Δ are given in Fig. 4. As for γ and γ', a linear pressure dependence is assumed with the result that $d\Delta/dP = 2.3\pm0.6$ MHz/Torr. The displacement Δ is toward increasing frequency.

Using the numbers given in Table I, it is possible to compute cross sections for both Ne*-Ne and Ne*-He collisions. For this purpose, see Eq. (46) of Ref. 4.[9] The values one gets for Ne*-Ne are negative. This is really not so startling since the calculated cross sections are extremely sensitive functions of the input values for $d\gamma/dP$ and $d\gamma'/dP$. Accordingly, cross sections are not determined by us on account of the 20–30% errors associated with the results of Table I. Reduction of errors to a level of 5% or less seems required. A much more difficult experiment is implied by this.

Equation (3) is the result of a calculation carried to third order in the atomic polarization. Others considered

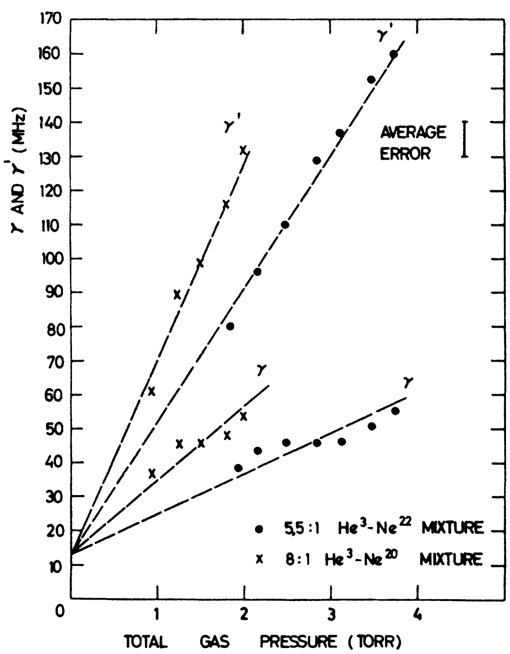

FIG. 3. Dependence of γ and γ' on P is given for both mixtures.

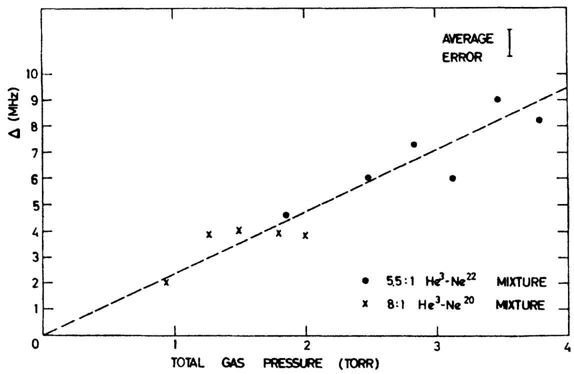

FIG. 4. Dependence of Δ on P is given for both mixtures.

terms of fifth order,[10] which increase in importance as the level of operation of the laser is increased progressively above threshold; but these calculations do not include the effect of collisions. A simple way to check the adequacy of our theory is to examine γ and γ', at fixed P, for any systematic dependence of γ and γ' on the level of discharge excitation. No such dependence was found.

5. DISCUSSION

An experiment very similar to ours was done earlier by Smith.[11] However, the data are fit to an expression for the power output of the form

$$I(\omega)=K\{G_0-(L)\exp[(\omega-\omega_0)/\omega_D]^2\}\times\left[1+\frac{(\gamma'')^2}{(\gamma'')^2+(\omega-\omega_0)^2}\right]^{-1}.$$

This does not account for (1) line-shape asymmetry, (2) terms of order $[(\omega-\omega_0)/\omega_D]^2$ in the integration of χ'' over atomic velocities, and (3) the two line-shape parameters γ and γ'.

Bennett *et al.*[12] have measured the frequency dependence of spontaneous emission at 6328 Å for varying discharge conditions. A comparison of a Lorentz width $\frac{1}{2}(\Delta\nu)_L$, and our γ is made regarding their dependence on pressure. A disagreement of one order of magnitude is cited with γ less than $\frac{1}{2}(\Delta\nu)_L$. Much better agreement with Smith is cited. We have discussed Smith's work. Further, in a pure Ne discharge, at 0.1 Torr, $\frac{1}{2}(\Delta\nu)_L=81$ MHz. This is very much larger than 15 ± 2.5 MHz obtained from an inverted Lamb-dip effect by Lee and Skolnick.[13] Finally, Lisitsyn and Chebotaev[14] recently

[9] There is a misprint in the equation for γ. The mean relative velocity in the second term should be $\bar{v}_{\text{Ne-Ne}}$ and not $\bar{v}_{\text{Ne-He}}$ as given. Further, γ_N is not defined correctly. Both the upper and lower levels contribute to γ_N; i.e., $\gamma_N=\frac{1}{2}(T_a^{-1}+T_b^{-1})$, in which T_a and T_b are the radiative lifetimes of the upper and lower laser levels, respectively. A. Szöke (private communication).

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reported results which show a marked deviation from the earlier work of Bennett, Chebotayev, and Knutson.[12]

The recent theory of Gyorffy, Borenstein, and Lamb[5] is most relevant to the present experiment. Therein, an expression for the power output of the type given in Eq. (3) above is further clarified. Their parameters γ_1 and γ_2 correspond to our γ' and γ, respectively. However, the theory predicts a nonlinear dependence on pressure for γ_2. From our determination of γ, this nonlinearity is apparently small for the 6328-Å transition. Estimates of line-shape asymmetry are in qualitative agreement with our observations.

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Atomic Relaxation in the Presence of a Coherent Optical Field

Edward A. Sziklas
United Aircraft Research Laboratories, East Hartford, Connecticut 06108
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Atomic relaxation in the presence of a strong coherent optical field is examined. Contrary to popular practice, we find that the Bloch equations do not apply when the applied field is strong enough to saturate the transition. The component of the induced polarization in phase with the field lives longer than the out-of-phase component. One important consequence is that pulse durations for adiabatic inversion of atomic levels need not be short relative to the transverse relaxation time of the atom if the pulse amplitude is sufficiently large.

ADIABATIC inversion of atomic levels by optical pulses has been proposed[1] as a technique for studying relaxation effects in atoms and molecules. Previous experiments involving light pulses and atomic coherence (e.g., photon echo[2] and self-induced transparency[3]) require pulse durations which are short compared to the transverse relaxation time T_2 of the atoms. Usually T_2 is several orders of magnitude smaller than the lifetime T_1 of the atom. Hence, this can be a serious limitation.

We find that adiabatic inversion is not subject to the same limitation on pulse duration. If the amplitude of the pulse is sufficiently large, pulse durations need only be short relative to T_1, not T_2. This feature, which is analogous to field-dependent relaxation[4] or spin locking[5] in NMR, suggests a broader range of applicability for adiabatic inversion than previously anticipated.

To exhibit this property we consider a simple model consisting of a two-level atom coupled to a coherent em field. The atom relaxes by virtue of a weak interaction with an external thermal bath, representing, for example, collisions with other gas atoms. The atomic states, labelled $|a\rangle$ and $|b\rangle$ with energy spacing $\epsilon_a-\epsilon_b=\hbar\omega_0$, are connected by an electric-dipole transition. We choose $l=1$, $m_l=1$ for level $|a\rangle$, and $l=0$ for level $|b\rangle$, where l,m_l are the usual angular momentum quantum numbers. The levels $|a\rangle$ and $|b\rangle$ are assumed to be nondegenerate.

Both the coherent applied field and the incoherent thermal bath are treated classically. The bath is described by a weak electric field whose components fluctuate randomly in time. The coherent wave oscillates with the frequency ω and is plane polarized in the x direction.

The Hamiltonian for the interacting atom is written $H=H_A+V_c(t)+V_I(t)$, where H_A is the Hamiltonian of the bare atom, and $V_c(t)$ and $V_I(t)$ are the coherent and incoherent interactions, respectively. Using the eigenstates $|a\rangle$, $|b\rangle$ of H_A as a basis for a matrix representation, we choose

$$V_c(t)=pE\cos\omega t\begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},\qquad V_I(t)=p\begin{pmatrix}\delta E_a(t) & \delta E_-(t)\\ \delta E_+(t) & \delta E_b(t)\end{pmatrix},\tag{1}$$

where $p=ex_{ab}$ is the atomic dipole matrix element, E is the amplitude of the coherent field, $\delta E_\pm(t)=\delta E_x(t)\pm i\delta E_y(t)$ are the fluctuating field components

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