Massachusetts Institute of Technology
Cambridge, Massachusetts 02139

TITLE

Study of Various Aspects of Raman Scattering Using Gas Lasers

Semi-Annual Technical Summary Report #4

Period Covered: February 1, 1967 to July 31, 1967

under supervision of

Project Scientist: Professor A. Javan
Telephone: Un 4-6900, Ext. 5038

under

Contract Nonr 3963(22) Modification #1 and 2
Project # NR 014-209/7-27-66

ARPA Authorization Order # 306

Starting Date: August 1, 1965 Expiration Date: July 31, 1968

Amount of Contract $120,340

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AD 667742

APR 22 1968

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SUMMARY

The laser induced line narrowing effect, observed and utilized previously in precise isotope shift determination in Ne, is analyzed theoretically in considerable detail. It is shown that two types of multiple quantum processes play important roles and dramatically influence the lineshape of the laser-induced signal. An extraordinary manifestation of these processes is observed in an experiment on the 8446Å laser oscillation in the atomic oxygen fine structure transition, $3p\ ^3P_{0,2,1} - 3s\ ^3S_1$. This experiment relates to a "stimulated version" of the laser induced line narrowing effect observed earlier in spontaneous emission in Ne.

The above laser induced line narrowing effect is particularly useful in precise measurements of atomic energy level structure; it also involves novel radiative processes influencing dramatically the observed shapes. Experiments are in progress emphasizing the radiative aspects of the effect as well as the spectroscopic application.

An experiment is in progress in which a new gas laser mechanism is being explored as a possible mechanism in a previously observed laser oscillation in one of the visible Ne transitions and also in a similar laser transition in Argon. Furthermore, this effect is also being explored to obtain possible laser oscillation in a number of important atomic transitions, such as the hydrogen $H\alpha$ line. It relates to laser oscillation within two levels where the upper level has a broad velocity distribution at an elevated temperature while the lower level velocity distribution is at much colder temperature; this effect may lead to population inversion and gain at the

wings of the Doppler broadened transition and heavy absorption within the central portion of the line profile. It is shown that this process is dominant in atomic oxygen fine structure laser transition.

The experiment on the motional narrowing of the spontaneous Raman scattering in hydrogen molecule is being pursued actively. This effect was observed previously, however, the final measurements were considerably delayed because of technical problems involving breakdown of the homemade Argon laser used in the experiment. Several new laser designs have been constructed and tested. The final design utilizes cold cathode together with floating voltage graphite rings placed within the discharge tube. This design is completely successful and has resolved all the previous technical difficulties. The experiment is now expected to be completed within the next several months; the observations include study of the effect in ortho and para-hydrogen molecules and also in D_2.

Frequency Spectrum of Spontaneous and Stimulated Line Narrowing Effects Induced by Laser Radiation

M.S. Feld and A. Javan

In an earlier publication,[1] a laser-induced line narrowing effect was utilized in a precise determination of isotope shifts in two Ne transitions. A recent paper[2] also reports observation of this effect and its application in studying some linewidth parameters in Ne. These experiments study fluorescence arising from the lower level of a Doppler-broadened laser transition to a third level. [See Fig. 1(a).] Viewed along the laser axis, the broad fluorescence lineshape is dramatically influenced by the laser field. For a standing-wave field detuned from its atomic center frequency, the laser-induced change signals appear as resonant increases in intensity over two narrow intervals symmetrically located on opposite sides of the fluorescence center frequency. The fluorescence from the upper laser level [See Fig. 1(c)], similarly viewed, would exhibit narrow resonant decreases in its overall emission profile.

The overall features of this effect may be described by noting that the standing-wave laser field selectively interacts with atoms whose velocities Doppler-shift one of its travelling-wave components into resonance; this produces changes in the laser level populations--an increase in the lower level population and a decrease in the upper level population--over two narrow intervals symmetrically located about the center of the velocity distribution. A recent letter[3] has analyzed the line shape details

for the cascade case [Fig. 1(a)] in terms of two-quantum transitions from level 2 to level 1, and predicts differing widths for the two laser-induced change signals. A similar lineshape asymmetry, described below, would appear in the change signals from the upper laser level, [Fig. 1(c)]. Note that in the latter case, however, the $0 \rightarrow 1$ emission act is an inherently single-quantum event, requiring another description. The summary of a different treatment[4] based on the density matrix formalism has described the spontaneous emission profile in both cases. Lineshape behavior of similar origins has been encountered in the microwave region in experiments involving the interaction of two monochromatic, classical fields with a three-level system.[5,6] These considerations may be readily extended to the present case by including the Doppler effect, and noting that the spontaneous emission spectrum from either laser level [level 0 of Figs. 1(a) and 1(c)] follows the emission line shape stimulated by a weak monchromatic field tuned through that resonance when the population of level 1 is ignored. In fact, recent experiments have studied the response of a Doppler-broadened three-level system coupled to two classical fields. The line-shape theory describing these experiments is directly applicable to important special cases of the effect observed in spontaneous emission. This letter has three intimately related objectives: The first is to point out the direct relevence of the detailed analyses of Refs. 7 and 8 to this problem; these also predict that one of the laser-induced resonances will be narrower than the other. Secondly, we emphasize that experiments based on stimulated versions of this effect demonstrate the different characteristics of the two laser-induced resonances. Thirdly, this letter generalizes the theory in several ways, including important power-broadening effects.

The analysis of Ref. 7 describes the third-order interaction of two monochromatic fields with the folded Doppler-broadened system of Fig. 1(b) in which

levels 1 and 2 are assumed to be closely spaced. Note that the analysis of a cascade system [Fig. 1(a)] in which the middle level lies about halfway between the other two levels follows identically. For purposes of illustration, let us assume that both transitions exhibit gain. Consider a strong standing-wave laser field $E_2(\Omega_2)$ detuned by an amount $|\Delta_2| > \gamma_{21} + \frac{1}{2}\gamma_0$ from ω_2, the peak of the 2-0 Doppler response [Fig. 1(b)]; here, $\gamma_{ij} = \frac{1}{2}(\gamma_i + \gamma_j)$, with γ_j the natural decay rate of level j. As a weak monchromatic field $E_1(\Omega_1)$ is tuned through the 1-0 transition, it is shown that the broad Doppler gain profile is considerable modified by the presence of E_2. The resulting lineshape is obtained from eqns. (33d) and (33e)[9] of Ref. 7:

$$P(\Omega_1) = G(\Omega_1)\left[1 + \xi E_2^{o2}\,\mathrm{Im}\left\{\frac{1}{(\Delta_2+\Delta_1) + i(\gamma_{21}+\gamma_0)} + \frac{1}{(\Delta_2-\Delta_1) + i\gamma_{21}}\right\}\right], \quad (1)$$

$$\omega_1 \approx \omega_2,$$

in which $\Delta_j = \Omega_j - \omega_j$, ω_j = atomic center frequency of j-0 transition, and Ω_j and E_j^o are frequency and amplitude of $E_j(\Omega_j)$, respectively; $G(\Omega_1)$ is the linear Doppler response, a slowly varying function of Ω_1, and ξ is a constant factor. For Ω_2 above ω_2, (1) predicts narrow Lorentzian responses of widths $\Gamma_B = \gamma_1 + \gamma_2 + 2\gamma_0$ below ω_1 and $\Gamma_N = \gamma_1 + \gamma_2$ above ω_1, the latter being narrower by $2\gamma_0$ and independent of γ_0. In subsequent discussion we shall refer to the "broad" (Γ_B) and "narrow" (Γ_N) resonances, respectively.

The narrow resonance, Γ_N, predicted by (1) has been observed and fully verified[10] for $\Gamma_N \ll \Gamma_B$. In these experiments levels 1 and 2 are tunable Zeeman components of an upper laser level connected to a common lower level, 0. The monochromatic fields are two oscillating laser modes of fixed frequencies determined by the cavity length. The effect is observed as sharp decreases in the laser output as the 2, 1 level splitting is magnetically tuned. These methods, however, have not been applied to the observation of Γ_B, which requires absolute frequency control as in Lamb-dip

experiments. (See Ref. 7)

We report here an extraordinary manifestation of the broad resonance, observed in high-resolution studies of the $3p\,^3P_{0,2,1}$-$3s\,^3S_1$ atomic oxygen fine-structure laser oscillations at 8446 Å. The relevant fine-structure components of this system consist of two resolved and closely spaced Doppler-broadened transitions forming folded three-level systems of the type shown in Fig. 1(b). For reasons unrelated to the present discussion, [11] (i) the central portion of each of the fine-structure gain profiles is entirely depleted and appreciable gain exists only on the wings; and (ii) laser action most readily occurs on the high-frequency side of the strongest (2-0) fine-structure component. For each laser mode oscillating on the 2-0 transition, two Lorentzian holes of enormously different widths selectively deplete the 1-0 Doppler gain profile: the low-frequency hole, of width $2\gamma_0+\gamma_1+\gamma_2 \simeq 130$ Mhz, is about 15 times broader than the high-frequency hole, of width $\gamma_1+\gamma_2 \simeq 9$ Mhz. [13] Because of multimoding on the 2-0 transition, multiple hole pairs are burnt into the 1-0 transition. The broad low-frequency holes overlap, completely suppressing laser action below the 1-0 line center, and oscillation can occur between the narrow, nonoverlapping holes above the center frequency. The 8446 Å spontaneous emission and laser output have been studied photographically with a high-resolution Fabry-Perot interferometer with free spectral range of 0.8 cm^{-1} under a wide range of conditions. The laser oscillations were obtained in argon-oxygen and neon-oxygen mixtures, [14] using a 3-meter cavity with 50 Mhz mode spacing. The operating pressure was kept below 1 torr to minimize pressure broadening effects. To establish the frequency shifts, Fabry-Perot images of the laser oscillations and the spontaneous emission were superimposed upon the same glass plate emulsion. In absence of laser oscillation, the spontaneous-emission analysis shows, in order of increasing frequency: a well-resolved fine-structure

component (1-0) with a completely symmetrical profile, and the strongest fine-structure component (2-0), overlapped on the high-frequency wing by the third (weak) fine-structure component. Close to threshold, laser oscillation first occurs on the high-frequency wing of the 2-0 component, where maximum gain occurs, as explained in Footnote 11 (ii). Further above threshold the 1-0 transition also breaks into oscillation. Despite the observed symmetry of the 1-0 profile, this concurrent oscillation occurs above the 1-0 center frequency, an effect which must follow from the depletion of gain over the entire low-frequency wing, as described above. (It must also be pointed out that in the observations reported here the 1-0 oscillation was at all times close to threshold and considerably weaker than the 2-0 oscillation.[15])

Reference 7 treats the field interactions up to third order. We now outline a different approach,[8] formulated in terms of single- and double-quantum transitions, which includes power-broadening effects due to a strong laser field. The latter are of considerable interest: on the theoretical side, it is important to inspect whether or not the strikingly simple lineshape behavior is merely characteristic of a third-order calculation; on the experimental side, it is important to know the influence of power broadening on the observed lineshape. Consider the cascade system of Fig. 1(a). [The final result will be written in a form also valid for emission from the upper laser level, Fig. 1(c).] It is desired to calculate the 0-1 emission spectrum stimulated by the weak travelling-wave field $E_1(\Omega_1)$ in the presence of the strong standing-wave field $E_2(\Omega_2)$ coupled to the 2-0 transition. Specifically standing-wave effects are avoided by taking Ω_2 detuned ($|\Delta_2| > \gamma_{20}$); then oppositely propagating travelling-wave components of E_2 do not couple, and the interaction consists of these components independently coupled with E_1.

Consider an ensemble of atoms moving with given axial velocity v,

the $+z$ axis being defined by the propagation direction of the travelling-wave field E_1. In the atoms' rest frame the incident fields appear Doppler-shifted to frequencies $\Omega_1' = \Omega_1(1 - \frac{v}{c})$ and $\Omega_2' = \Omega_2(1 - \epsilon \frac{v}{c})$ in which $\epsilon = \pm 1$ indicates the travelling-wave component of E_2 propagating along the $\pm z$ direction, respectively. We now require a solution in the atoms' rest frame in which the three-level system is coupled to one of the travelling-wave components of the strong field E_2 at frequency $\Omega_2' \sim \omega_2$ and to the weak travelling-wave field E_1 at frequency $\Omega_1' \sim \omega_1$. The resonant interaction of two monochromatic fields with a three-level system was treated in Ref. 5 for the case of γ_j all equal. The perturbation method consisted of first obtaining a closed form solution to the Schrodinger equation for $E_1^o = 0$ and E_2^o arbitrary; and then using this result to generate a solution valid to first order in E_1^o. When the method is extended to the case of arbitrary γ_j,[16] the emitted power induced by E_1 at Ω_1' is:

$$8\hbar\Omega_1' \, |\beta_1|^2 \, \mathrm{Im}\left[n_2 \, |\beta_2|^2 \, \frac{(L_2 - 2\frac{\gamma_{20}}{\gamma_0}R)}{AB} - n_1 \frac{R}{B} - n_0 \left\{ \frac{R}{E} + |\beta_2|^2 \frac{(L_2 - 2\frac{\gamma_{20}}{\gamma_0}R)}{AB} \right\} \right]. \quad (2)$$

Here, $A = |L_2|^2 + \frac{4\gamma_{20}^2}{\gamma_0\gamma_2}|\beta_2|^2$ and $B = -RL_1^* + |\beta_2|^2$, with $L_j = (\Omega_j' - \omega_j) + i\gamma_{j0}$, $R = [(\Omega_1' + \Omega_2') - (\omega_1 + \omega_2)] - i\gamma_{21}$, $|\beta_j| = |\mu_{jo}E_j^o/4\hbar|$, and μ_{jo} is the electric-dipole matrix element connecting levels j and 0; n_j is the number of background atoms with velocity v in level j, i.e. the population in absence of the strong laser field. In Eqs. (2) the n_2 coefficient is obtained from the $2 \to 1$ transition rate due to double-quantum emission at Ω_1' and Ω_2'; the n_0 coefficient, in contrast, is obtained from the single-quantum emission rate, arising from $0 \to 1$ transitions as modified by the presence of E_2; the n_1 coefficient results from the reverse processes, namely, double-quantum $1 \to 2$ transitions and single-quantum $1 \to 0$

transitions.[17] (See Ref. 5.) As a check of the detailed algebra, Eq. (2) has also been obtained in an independent calculation using the ensemble-averaged density matrix to estimate the induced polarization at Ω_1', an approach equivalent to the one presented here.[8]

It is important to note that (2) is entirely valid for the case in which the γ_j's are interpreted as decay rates arising from hard collisions. The detailed features of the line shape predicted[5] by (2) for equal γ_j have been fully verified in the microwave region where Doppler effect is negligible and the linewidths are entirely due to collision effects.[6]

The complete emission spectrum is obtained by averaging (2) over the atomic velocity distribution for $\epsilon = +1$ and for $\epsilon = -1$. In the fully Doppler-broadened limit $\gamma/D \ll 1$ ($\gamma \sim \gamma_{ij}$ and D= Doppler width), and for $\omega_1 \geq \omega_2$,

$$P(\Omega_1) = G(\Omega_1)\left[1 + \frac{N_0 - N_2}{N_0 - N_1}\,\xi'\,E_2^{o2}\,\mathrm{Im}\left\{\frac{1}{(\Delta_1 + \sigma\frac{\omega_1}{\omega_2}\Delta_2) + i\frac{\Gamma_B}{2}} + \frac{1}{(\Delta_1 - \sigma\frac{\omega_1}{\omega_2}\Delta_2) + i\frac{\Gamma_N}{2}}\right\}\right], \quad (3)$$

in which $\frac{1}{2}\Gamma_B = \gamma_{10} + \frac{\omega_1}{\omega_2}\gamma_{20}Q + \frac{\gamma_0}{2}(Q-1)$ and $\frac{1}{2}\Gamma_N = \gamma_{10} + \frac{\omega_1}{\omega_2}\gamma_{20}Q - \frac{\gamma_0}{2}(Q+1)$; N_j is the total background population of level j, ξ' is a proportionality factor > 0, $Q^2 = 1 + \frac{4|\beta_2|^2}{\gamma_0\gamma_2}$, and $G(\Omega_1) \propto (N_0 - N_1)E_1^{o2}$ is the power emitted at Ω_1 by the Doppler-broadened 0-1 transition induced by E_1 in the absence of the laser field. Equation (3) shows the power broadening influence of the laser field, which enters in a remarkably simple way. Equation (3) has been written in a form valid for both cascade ($\sigma = -1$) and folded ($\sigma = +1$) cases, Figs. 1(a) and 1(c).[18]

As pointed out above and explained in 18, the spontaneous emission spectrum from the upper or lower laser levels, as viewed along the laser axis, is also given by Eq. (3) when N_1 is set equal to 0 and $G(\Omega_1)$ is interpreted as the usual Doppler-broadened spontaneous emission spectrum for $E_2 = 0$.

The discussions of Ref. 3 are consistent with weak-field limit of our treatment as it applies to spontaneous emission for the special case of $N_0 = 0$. Note, for instance, that for $|\beta|^2 \ll 1$ and in the limit of $\gamma_{21} \to 0$, the frequency dependence of the $2 \to 1$ transition rate, obtained from the n_2 coefficient of our Eq. (2),

would involve a δ-function, becoming $\delta(\Omega_1' + \Omega_2' - \omega_1 - \omega_2)\,|L_2|^{-2}$; the $\gamma_{21} = 0$ discussion of Ref. 3 is equivalent to averaging this distribution over velocities.

To emphasize the significance of the role played by N_0, the background atoms in level 0, consider a cascade system in which only level 0 is populated (i.e. $N_1 = N_2 = 0$). Then in the rest frame of an atom, an applied laser field at Ω_2' will diminish the transition rate at Ω_1', leading to two holes of width Γ_B and Γ_N superimposed upon the emission profile. (See eqn. (3).) As discussed earlier, a $0 \rightarrow 1$ transition is an inherently single-quantum event [5] and may not be described in terms of a double-quantum process as in a $2 \rightarrow 1$ transition.

As pointed out above, in all of the experiments involving oxygen[10,12] and xenon[10] Γ_N and Γ_B enormously differ. In the Ne spontaneous emission experiments reported in Refs. 1 and 2, however, they are expected to differ by only about 30%. The observation of this difference would require high finesse Fabry-Perot analysis and good laser stability and has not yet been achieved.

In averaging eq. (2) for the case of finite γ_{21} a number of cancellations occur in the fully Doppler-broadened limit ($\gamma/D << 1$), leading to a particularly simple expression. It is important to point out that such cancellations do not occur in higher orders of γ/D. For instance, the complete cancellation of γ_0 in Γ_N, which occurs in the case of $(\omega_1/\omega_2) \approx 1$, will not occur in the next order of γ/D.

A paper including complete algebraic details and additional discussions is being submitted for publication.[8]

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9. There are several relevant misprints in eqn. (33): in line (33e), E_1E_2 should read $E_1E_2^2$; line (33d) should read:
$+ (|\mu_{12}\mu_{31}|^2/\gamma_1)(N_3 - N_1)\frac{1}{2}E_1E_2^2 \; [\gamma - i(\omega_B - \nu_B)]^{-1}$.

10. H.R. Schlossberg and A. Javan, Phys. Rev. Letters 17, 1242 (1966); G.W. Flynn, M.S. Feld and B.J. Feldman, Bull. Am. Phys. Soc 12, 669 (1967).

11. For explanation of (i), see Refs. 8 and 12; (ii) is merely due to the presence of the weak fine-structure component with gain ($^3P_0 - {}^3S_1$), which overlaps the high-frequency wing of the 2-0 transition ($^3P_2 - {}^3S_1$). The 1-0 transition ($^3P_1 - {}^3S_1$), however, is completely symmetrical and free of overlap.

12. M S. Feld, B.J. Feldman and A. Javan, Bull. Am. Phys. Soc. 12 669 (1967); "Frequency Shifts of the Fine Structure Oscillations of the 8446 Å Atomic Oxygen Laser", Phys. Rev., to be published.

13. For Oxygen linewidths, see: W.L. Wiese, M W. Smith and B.M. Glennon, Atomic Transition Probabilities, Vol. 1, U.S. Department of Commerce, National Bureau of Standards, Wash. D.C.

14. W.R Bennett, Jr., W.L. Faust, R.A. McFarlane, and C.K.N. Patel, Phys. Rev. Letters 8, 470 (1962).

15. A study of the intensity of the 1-0 laser oscillation as a function of cavity length would be of interest.

16. In this case the time-dependent wave-function, Ψ, is obtained from a three-level Schrodinger equation to which radiative decay terms have been added. For $\Psi = \sum_{j=0}^{2} c_j e^{-iW_j t} u_j$, with u_j the eigenfunction of level j of energy $\hbar W_j$, the coupled equations are: $\dot{c}_i = \sum_j (a_{ij} - \frac{1}{2}\gamma_j \delta_{ij}) c_j$, in which $a_{ij} = -\frac{\mu_{ij} E(t)}{i\hbar} e^{i(W_i - W_j)t}$ and E(t) is the sum of the two travelling-wave fields as seen in the atoms' rest frame.

17. As an example, for an atom in level 0 at initial time t_0, $|c_j(t=t_0, t_0)| = \delta_{j0}$, and the $0 \to 1$ transition rate at a later time t is $\gamma_1 |c_1(t, t_0)|^2$. (See preceeding footnote.) Thus, the total stimulated power emitted by background atoms in level 0 is $\hbar\Omega_1' n_0 \gamma_0 \gamma_1 \int_{-\infty}^{t} |c_1(t, t_0)|^2 \, dt_0$.

18. In extending eqn. (2) to the spontaneous emission case, the population of level 1, n_1, should be set equal to 0 and the energy density of the weak probe field, $E_1^{o\,2}/8\pi$, should be replaced by $(\hbar\Omega_1'^3/8\pi^3 c^3)\, d\Omega_1' \, dS$, where frequency interval $d\Omega_1' \ll \gamma$ and dS is a small solid angle in the forward direction (+z - axis); E_2, the laser field, remains in its classical monochromatic form. Similar remarks apply to eqn. (3); note, in particular, that $G(\Omega_1)$ becomes the usual Doppler-broadened spectrum of the power emitted spontaneously into $d\Omega_1 \, dS$ with given polarization.

FIGURE CAPTIONS: The figure caption is contained in the figure.

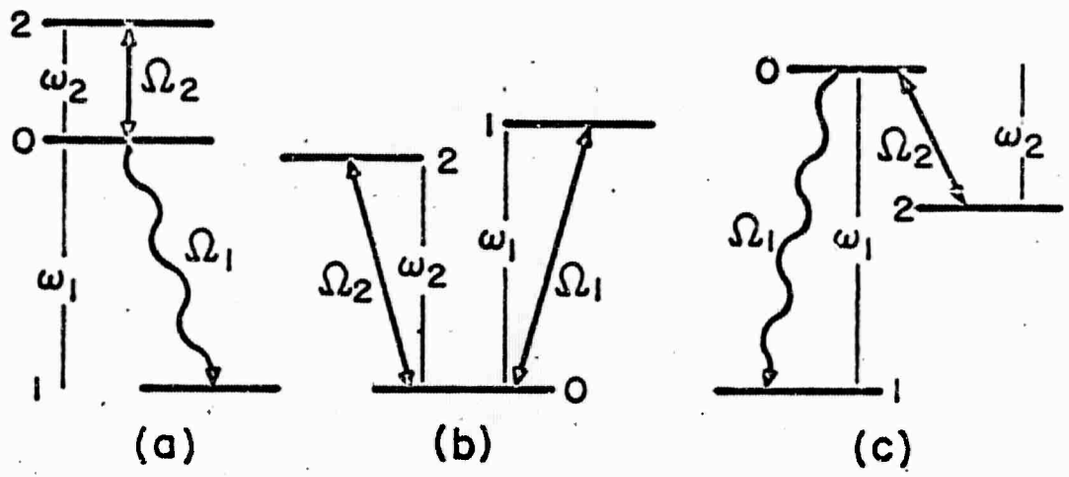

FIG. 1 ENERGY LEVEL DIAGRAM

"Inversion of Population Between a Kinetically Hot Upper Level and a Kinetically Cold Lower Level; a New Gas Laser Mechanism"

B. J Feldman, A. Javan

Consider an atomic transition consisting of two levels where the population of the lower level summed over all velocities is larger than that of the upper level. If both levels possess similar velocity distributions, than at any point on the velocity distribution the population of the upper state will be less than that of the lower state. Consider, however, the case where the velocity distribution of the upper level is excessively broadened[1]; population inversion may then occur at the wings of the velocity distribution. It is expected that this new type of inversion will have wide applicability as an effect for obtaining laser action. In particular, we report here the observation of this new type of inversion leading to laser oscillation in atomic oxygen and discuss the possibilities of two new important laser systems utilizing this effect.

In 1962 Bennett et al.[2] reported laser action on the 8446Å transition $(3_p\,^3P_2) \rightarrow (3s\,^3S_1)$ in atomic oxygen, observing an anamalous shift of laser radiation .07cm^{-1} off the peak of the flourescence leg line center. The upper levels of the 8446Å transition are populated by the dissociation of oxygen molecules upon collisions of the second kind with neon metastables, i.e. the dissociative excitation transfer process,

$$Ne^* + O_2 \rightarrow Ne + O + O(3p\,^3P) + k.e. \qquad (1)$$

They have confirmed the occurrence of this process by observing significant broadening of the $2p_1 \rightarrow 1s_2$ (5852Å) neon transition several hundred microseconds into the afterglow, indicating a $2p_1$ velocity distribution at three times room temperature. The $1s_2$ level is known to be heavily trapped. However, the breadth of the upper level in the afterglow suggests that inversion may occur at the Doppler wings during the afterglow period. The spontaneous lifetimes of 1.4×10^{-8} sec($2p_1$) and 5×10^{-9} sec ($1s_2$) are obviously favorable for laser action. A search of the literature reveals that Bridges and Chester [5] in a pulsed discharge tube 2 meters long and 3 mm. in diameter observed laser action at 5852Å with a gas mixture containing neon, helium, and traces of argon. At the present time, we are setting up such a pulsed discharge system to confirm the proposed mechanism. Using a high resolution Fabry-Perot interferometer, we shall study the position of the laser radiation with respect to the Doppler broadened spontaneous emission profile taking note of the time relation between the discharge pulse and laser pulse. If the laser is oscillating on the wings of the Doppler profile in the afterglow period of the discharge, this will confirm our prediction as to mechanism. Bridges and Chester [5] also observed pulsed laser oscillation on the $2p_1 \rightarrow 1s_2$(7503Å) transition in argon. We expect the same mechanism described for neon is also responsible in the case of argon.

We are also studying the possibility of utilizing population inversion at the wings of the velocity distribution to obtain laser oscillation

where the excess kinetic energy is shared by the oxygen atoms. Fabry-Perot measurements of the 3P flourescence taken in our laboratory indicate a breadth of $0.17 cm^{-1}$, five and one-half times that of the ground state. The lower level (^3S) is strongly connected to the ground state via ultraviolet transitions at about 1300Å, implying a large absorption cross-section so that for ground state densities exceeding $10^{13}/cm^3$, the resonance radiation is sizeably reabsorbed within one centimeter of discharge. Consequently, the population of the ^3S level is increased over the central portion of its velocity distribution. However, at the wings of the distribution there is little reabsorption due to the absence of high-velocity ground state atoms. A detailed experimental study has confirmed that laser action occurs only at the wings of the Doppler line, where the broad upper level still has sizeable population; but, the narrower lower level does not. (A paper discussing in detail laser action in atomic oxygen is presently being prepared and will soon be submitted for publication.)

At the present time we are studying the dissociative recombination process of Ne_2^+ molecules in the afterglow of a neon discharge as well as dissociative excitation transfer process in a hydrogen-neon discharge as possible sources of new important optical laser transitions utilizing the inversion effect described here. Connor and Biondi[4] have observed that in pure neon discharge afterglows at pressures from 2-75 mm. Hg. electrons decay predominantly through the dissociative recombination process,

$$Ne_2^+ + {}^e(\text{slow}) \rightarrow (Ne_2)_{\text{unstable}} \rightarrow Ne^* + Ne + k.\epsilon. \qquad (2)$$

where the kinetic energy is shared between the dissociated products.

in atomic hydrogen at 6563Å (H α line). Marshall[6] has reported that collisions between neon metastables and H_2 lead to the preferential population of the n=3 level of atomic hydrogen through the dissociative excitation transfer process,

$$Ne^* + H_2 \rightarrow Ne + H_{(n=3)} + H + k.e. \qquad (3)$$

Presently, we are using a high-resolution Fabry-Perot interferometer to ascertain the extent of broadening of the upper level. Due to the closeness of the energy match in (3), it may be necessary to liquid nitrogen cool the discharge tube so as to narrow sufficiently the ground state Doppler breadth to eliminate reabsorption of the 2p→1s ultraviolet photons at the wings. The enormous spectroscopic importance of this transition makes these attempts worthwhile.

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Study of the Laser Induced Line Narrowing in Neon

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Herein we report the progress made in our continuing study of the lineshape of spontaneous emission, at 6096-angstrom, originating from the lower level of the helium-neon laser transition at 1.15-micron. We begin this report with a very brief review of the changes in the lineshape, at 6096-angstrom, owing to the laser field. The ensuing discussion relates the current attempt to study the lineshape in greater detail in order to verify interesting and important radiative effects predicted recently from a rigorous theory applied to the problem in question.

First, let us briefly review the principal features of the laser-induced effect of interest. If spontaneous emission originating from the lower laser level (level-2) and terminating at some third level (level-3) is viewed in the direction of laser propagation (+z direction), the normal or Gaussian frequency dependence of the intensity is observed to be significantly modified. If the laser, operating in a single mode, is tuned to oscillate at frequency ω_0', corresponding to the peak of its gain profile, this modification takes the form of an increase in intensity at ω_0', the line center frequency of spontaneous emission at 6096-angstrom. The lineshape of this change or increase in intensity is Lorentzian and it has a width associated with it which is much narrower than the Doppler width normally associated with spontaneous emission. For the condition

of tuning cited, the signal is due to strong coupling to the laser field of atoms moving perpendicular to the +z-direction, with a consequent increase in the population of level-2 for atoms with velocity $v_z \simeq 0$. These features have been verified fully by us and an application of this effect to precise determinations of isotope shifts for neon spectra has been made.[1]

Let us now relate the exact nature of the problems being pursued currently. Some introductory remarks are necessary for purposes of clarity. There are important features in the lineshape of the spontaneously emitted signals involved in our experiment which follow from a detailed theory[2] but which have not been verified experimentally by us as yet. Let the laser oscillate at some frequency ω away from ω_o. Then two sets of atoms with velocities $v_z \simeq \pm c(\omega - \omega_o)/\omega_o$ are strongly coupled to the laser field, and at 6096-angstrom there occur two increases in intensity within narrow intervals of frequency symmetrically displaced about ω_o'. (This has also been verified by us earlier--see reference 1.) In a derivation of the expected widths of these signals from theory, if one considers only changes in level populations induced by the laser field, both high- and low-frequency signals are identical in that each is a Lorentzian with a half-width given by $\gamma_{23} + \omega_o' \gamma_{12}/\omega_o$, in which, for example, $\gamma_{12} = (1/2)(1/T_1 + 1/T_2)$, where T_1 and T_2 are the lifetimes of the upper and lower laser levels, respectively. These conclusions are consistent with our past observations within the precision implied therein.[1] A more rigorous theoretical analysis of the lineshape has now been done,[2] and it

has been shown that it is not sufficient to consider changes in level populations alone, or phrased somewhat differently, it is not sufficient to include only single-quantum events in the calculation. Coherent two-quantum events are important.[2] (The discussion given here is rather loose in that in a density-matrix calculation of the lineshape it is not possible to clearly separate one- and two-quantum contributions to the linewidth--indeed, it is an interference of these processes that accounts for the very interesting effects to be mentioned immediately below--see reference 2 for further detail.) The principal result of usch a more refined calculation, stated in the simplest terms, is that the widths associated with emission from atoms coupled to oppositely propagating running waves in the resonator are unequal. The emission from atoms with $v_z > 0$ and coupled to the running wave in the +z direction has a half-width $\gamma_{23} + \omega_o' \gamma_{12}/\omega_o$; i.e., the same as given earlier. The emission from atoms with $v_z < 0$ and coupled to the running wave in the -z direction has a half-width $\gamma_{23} + \omega_o' \gamma_{12}/\omega_o - \gamma_2$; i.e., the width is reduced by γ_2 from that given earlier.

At the present time we are attempting to verify these interesting and important radiative effects. There is a good reason why our earlier work failed to reveal the half-width difference in question. The laser was filled to a total pressure of 2 Torr. Collisions broaden both high- and low-frequency signals uniformly and equally, thus, at high pressure, mask the strictly radiative effects of interest cited above. The obvious,

but not necessarily the best, way to circumvent this difficulty is to build a laser to operate at a lower pressure--1 Torr is feasible for helium-neon but not much less. A less obvious, but much better, approach is to use a helium-neon laser filled to optimum pressure, albeit high, and to place within the resonator a very low pressure discharge of pure neon. One then views emission from the low pressure cell in which the relevant neon transition at 1.15-micron is saturated strongly by the laser field owing to the gain in the high pressure cell. In this way, owing to low pressure, the radiative effects of interest should be observable. The low pressure cell need not have an inversion of population for the transition of 1.15-micron. Signals from the high pressure cell will also be present but displaced in frequency from those of the low pressure cell because of an absolute pressure shift at 2 Torr.

We have now set up an apparatus very similar to that described earlier[1] except that the resonator has been lengthened to accomodate a second discharge tube. The high pressure gain cell is filled with an 8/1 He^3/Ne^{22} mixture to a total pressure of 2 Torr. The low pressure cell is filled with .03 Torr of pure Ne^{22}--the lowest pressure possible consistent with maintaining a discharge for some length of time. At this time it has been verified that the laser operates at 1.15-micron despite the addition of the second discharge (which attenuates at 1.15-micron) to the resonator. A preliminary search for the effects of interest is about to be made. The principal experimental difficulties encountered up to now have involved the construction of our discharge

tubes--this has always given us some difficulty. The tubes are very delicate and quite unique in design. Unfortunately, they are easy to break, and this has caused some delay.

Based on the lifetimes (in nanoseconds) $T_1 = 100$, $T_2 = 20$, and $T_3 = 20$, one expects a factor of two difference in the half-widths of the two signals, more specifically half-widths of $\simeq 20$ and 35 MHz. For an interorder spacing of 1500 MHz and a finesse of 70 (which we know is realizable from past work), the instrumental width contributed by the scanning Fabry-Perot interferometer will be $\simeq 10$ MHz. Collision broadening at .03 Torr will amount to only several MHz. Thus the two signals should have half-widths of $\simeq 30$ and 45 MHz, and this difference should be observable if one has a signal-to-noise ratio of about 5:1 or better.

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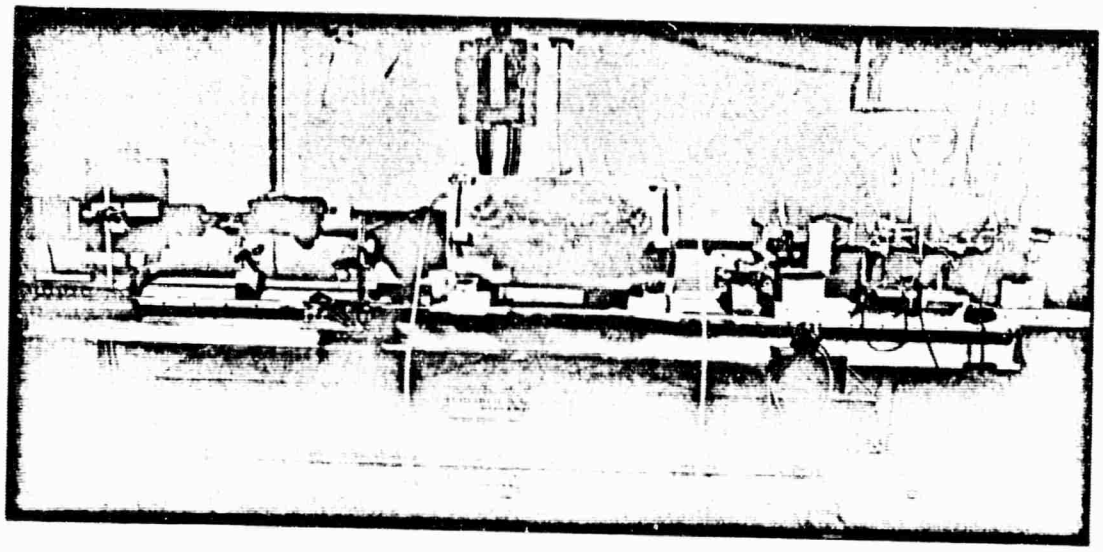

Figure 1

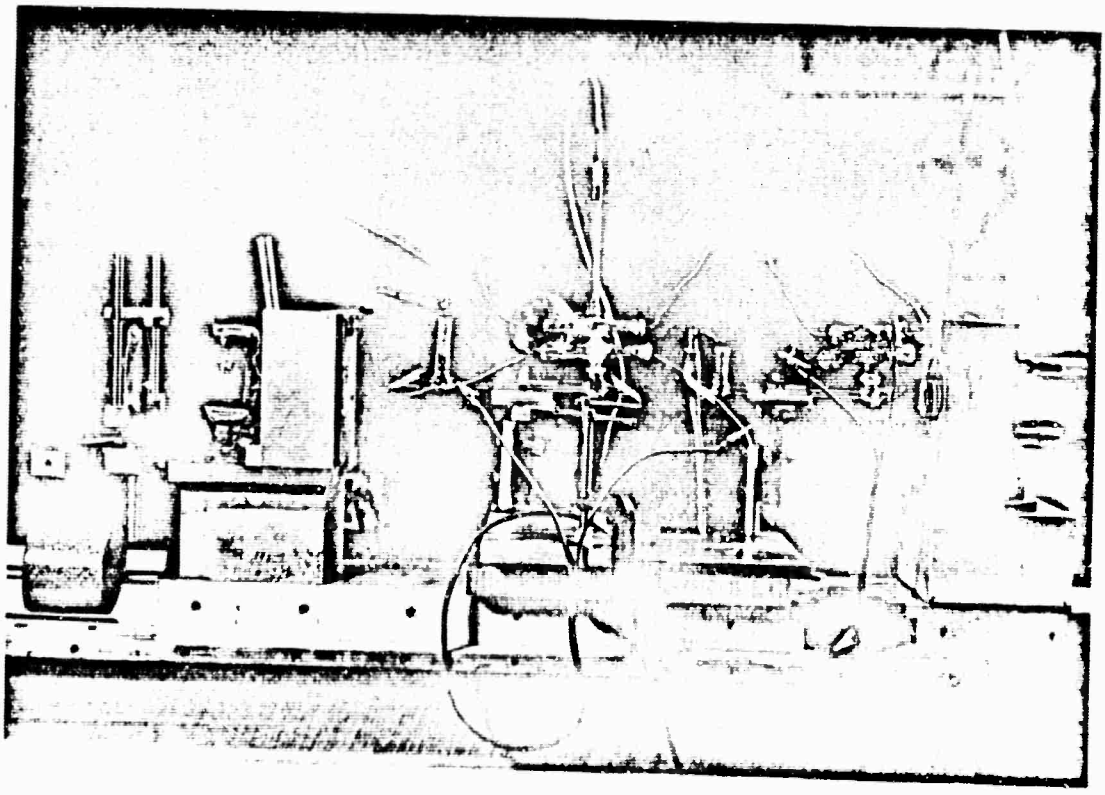

Figure 2

Pressure Narrowing Effect in H_2 and D_2

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The experiment or motional narrowing of the linewidth of spontaneous Raman scattering off hydrogen molecule is continuing. The narrowing of the Doppler linewidth as a function of pressure was observed[1] and reported in our previous progress report. The emphasis is now directed towards study of the effect in ortho- and para-hydrogen and in D_2 molecules.

The apparatus used a single mode Argon ion laser as a light source. Severe problems in the reliability of this source have forced a temporary redirection of the effort into improving the laser discharge tube. The quartz discharge tube previously employed has been eliminated, and a segmented graphite tube introduced. The new discharge tube confines the argon discharge to the necessary small diameter (3.5 mm) by passing it through small apertures in a stack of graphite plates suspended on a quartz support structure in a gas tube. The plates are electrically isolated from one another so that electric conduction through the graphite from cathode to anode cannot occur. The present structure used 50 plates 1/4" thick and 1 1/2" diameter. A barium oxide impregnated tungsten tube is used as a self heated thermionic hollow cathode.[2] The laser oscillates on a single mode, frequency stabilized to an external Fabry-Perot resonator.

With the completion of the new laser tube, we have now begun reexamination of the line narrowing effect. We anticipate that this project will be completed by the summer of 1968.

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