Selective Reabsorption Leading to Multiple Oscillations in the 8446-Å Atomic-Oxygen Laser*

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Laser oscillation of atomic oxygen at 8446 Å occurs in four closely spaced lines with peculiar intensity ratios, all detuned from the atomic center frequencies of the three fine-structure transitions. These anomalies are caused by the selective reabsorption of resonance radiation from the lower laser level by ground-state oxygen atoms. The selectivity results from the fact that the velocity distribution of the laser levels is considerably wider than that of the ground state, because of the dissociative mode of production of excited oxygen atoms. Possible extension of this mechanism to the atomic-hydrogen system is discussed. New atomic-oxygen laser lines at 2.89, 4.56, 5.97, 6.86, and 10.40 μ are also reported and assigned.

INTRODUCTION

Although the 8446-Å atomic-oxygen laser was one of the earliest gas lasers to be developed,[1] its behavior has remained mysterious. It was noticed immediately that the laser line was displaced from the peak of the spontaneous-emission profile [Fig. 1(a)], and later, when sufficient gain was obtained, that four laser lines appeared[2,3] with peculiar intensity ratios, all detuned from the atomic center frequencies of the three fine-structure transitions [Fig. 1(b)]. This unique behavior results from the unusual combination of three properties: (i) the fine structure, which consists of pairs of closely spaced transitions sharing a common lower level, (ii) the radiation trapping of the lower level, which is optically connected to the atomic oxygen ground state, and (iii) widely different velocity distributions for the atomic ground state and the lower laser level due to molecular dissociation excitation processes, resulting in resonance reabsorption of radiation over only part of the velocity distribution. The analysis given here explains the occurrence of the four lines and their asymmetrical placement.[4]

In a series of separate but related experiments the coupling among these four laser lines has been investigated. The results and analysis of these experiments are the subject of the following paper.[5]

LEVEL STRUCTURE AND EXCITATION PROCESSES

The level structure of the $3p\ ^3P_{0,1,2}$-$3s\ ^3S_1$ atomic-oxygen laser transitions at 8446 Å is shown in Fig. 2(a), together with the ^3P ground state, which is strongly connected to the $^3S_1$ lower laser level by three fast uv transitions at 1300 Å. The upper laser levels have a radiative lifetime of 36 nsec and the lower level a lifetime of 2.6 nsec.[6,7] The observed fluorescence spectrum due to spontaneous emission from the three upper levels is shown in Fig. 3(a). This measurement confirms the data of Bennett[8] and Tunitsky and Cherkasov,[3] together with an accurate early

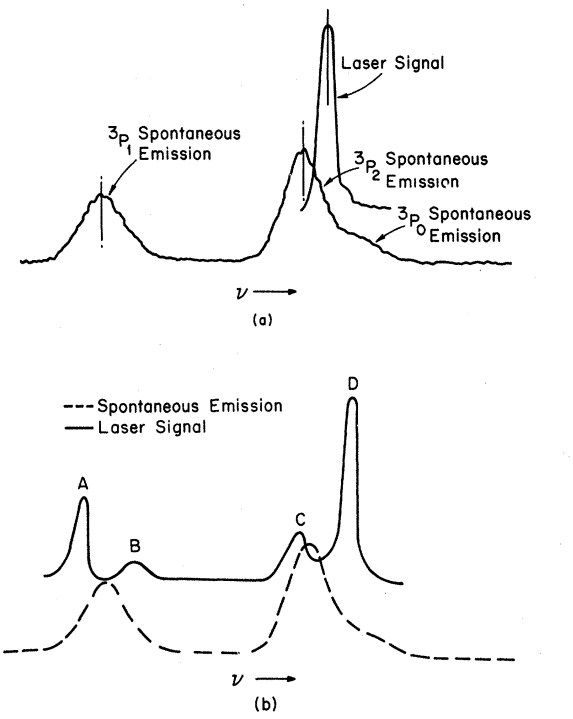

FIG. 1. (a) 8446-Å spontaneous-emission profile, showing initially observed laser line (after Bennett, Ref. 8). (b) 8446-Å spontaneous-emission profile, showing the relative positions of the four laser lines A, B, C, and D.

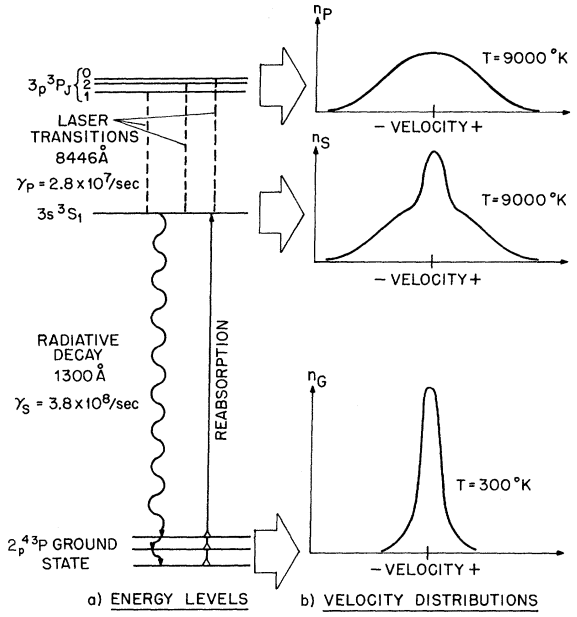

FIG. 2. (a) Relevant atomic-oxygen energy levels, and (b) corresponding velocity distributions, showing selective reabsorption.

study.[9] The $^3P_2$-$^3S_1$ transition is centered 0.557 cm^{-1} higher in frequency than the $^3P_1$-$^3S_1$ transition, and the $^3P_0$-$^3S_1$ transition is 0.159 cm^{-1} above the $^3P_2$-$^3S_1$ transition.[10] The spontaneous-emission intensities of the $J = 1, 2, 0$ components are in the ratios of their respective statistical weights, 3:5:1.

Although we are not directly concerned here with the details of the excitation mechanisms, it is necessary to consider their gross features. Low-pressure gas discharges of oxygen mixed with higher pressures of argon as in our experiments have been studied in detail by Bennett *et al.*[1,11] They surmise that oxygen molecules are dissociated in the discharge primarily by metastable argon atoms,

$$Ar^* + O_2 \rightarrow Ar + O + O^* + E_{K.E.}\ , \qquad (1)$$

where O* represents a metastable atomic-oxygen level, and following this, the oxygen metastables are raised to the upper laser levels by electron impact. The energy defect in reaction (1) is several electron volts, giving the excited oxygen atoms a velocity spread considerably wider than that of a room-temperature gas. Presumably, this distribution of velocities is maintained to some extent during the subsequent electron excitation.

Two recent developments in oxygen lasers raise the question of whether this mode of production is the dominant one. First, Tunitsky and Cherkasov have observed laser oscillation at 8446 Å in a pure oxygen discharge cooled to liquid-N_2 temperature.[12] Second, in collaboration with Professor George Flynn, now of Colombia University, we have observed and identified five new cw laser transitions at longer wavelengths in the same argon-oxygen discharge in which laser oscillation at 8446 Å occurs.[13] The energy levels associated with these lines lie considerably higher than the levels of the 8446-Å transition, and the upper level of one transition ($7\,^3D$) cannot easily be excited by electron collisions with the oxygen metastables ($2\,^1D$ and $2\,^1S$). These new transitions are tabulated in Table I.

SELECTIVE RESONANCE REABSORPTION

Whatever the mode of production, measurements of the ^3P-^3S fluorescence spectrum indicate Dop-

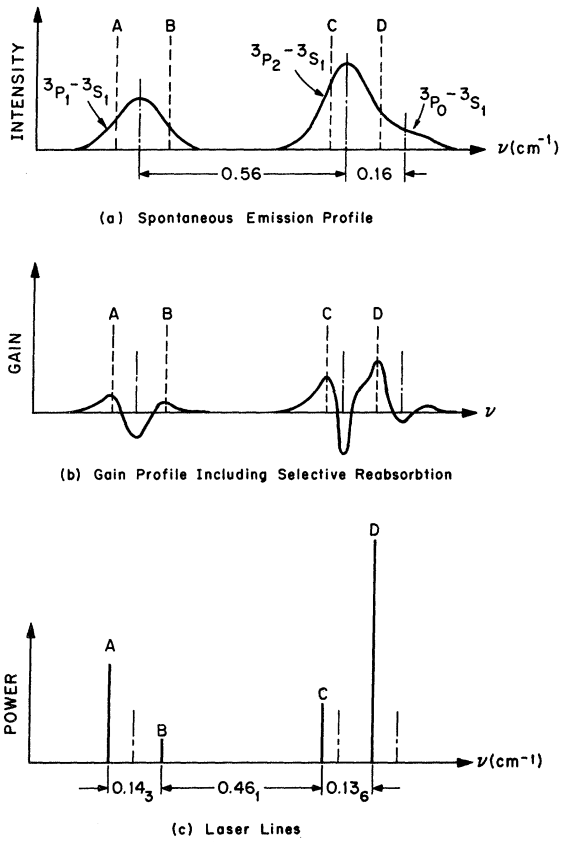

FIG. 3. (a) Spontaneous-emission profile of the three fine-structure transitions at 8446 Å and their measured frequency separations. (b) Sketch of the 8446-Å gain profile. The central portion of each transition is depleted due to selective reabsorption. The slight asymmetry of the $^3P_1$-$^3S_1$ profile is discussed in Ref. 5. (c) Location of the four laser lines relative to their spontaneous-emission profiles. Each line consists of a set of axial modes which extends over a range 0.01–0.05 cm^{-1}. The intensity ratios of the A, B, C lines are shown. The D line is off scale here.

TABLE I. Atomic-oxygen laser transitions.

Line (μ)	Observation: Vacuum wave numbers (cm^{-1})	Assignment: Vacuum wave numbers (cm^{-1})	Transition: Upper level	Transition: Lower level
0.8446	11 836.29[a] 11 836.16 11 835.70 ± 0.01 11 835.55	11 836.335[b] 11 836.176 11 835.619	$3p^3P_J$: $J=2$	$3s^3S°$ $J=1$
2.89	3 456.9 ± 3.0	3 454.90[c]	$4p^3P$	$4s^3S°$
4.56	2192.4 ± 0.9	2192.26	$4p^3P$	$3d^3D°$
6.86	1 458.4 ± 0.5	1 457.75	$5p^3P$	$5s^3S°$
10.40	961.4 ± 0.2	961.26	$5p^3P$	$4d^3D°$
5.97	1 674.9 ± 0.2[d]	1 671.40	$7d^3D°$	$6p^3P$

[a]See text for details.
[b]See Ref. 9.
[c]See Edlen, Ref. 10.
[d]This 0.2% discrepancy may be due to inaccuracy of Edlen's measurement.

pler widths of about 0.145 cm^{-1} full width at half-maximum, corresponding to a velocity distribution of over 9000 °K. In all likelihood the lower laser level also has a broad velocity distribution. In contrast, almost all ground-state oxygen atoms, produced by a variety of collision processes in equilibrium with the walls of the discharge tube, have a room-temperature velocity distribution.

Since resonance photons from the $^3S_1$ lower laser level are heavily reabsorbed, as we shall estimate, within a distance smaller than the tube diameter, atoms in the lower laser level with velocities that are also present in ground-state atoms are effectively inhibited from decaying. As a result, atoms in the central portion of the broad $^3S_1$ velocity distribution have effective lifetimes longer than those in the wings, because of the trapping of their resonance radiation. Consequently, the center population builds up considerably compared to the wings [Fig. 2(b)]. Since the laser gain depends on the population difference between the P and S levels, the final result is to deplete the central portion of the gain profile of each of the three laser transitions over a frequency range determined by the velocity distribution of ground-state atoms.[14] We shall refer to this velocity-dependent process as *selective reabsorption*.[15,16]

Including the fact that radiative decay from the upper laser level goes only to the lower laser level, simple rate equations for the populations of the upper laser n_P and the lower laser levels n_S yield, in the steady state,

$$\frac{n_P}{n_S} = \frac{\gamma_S/\gamma_P}{1+R_S/R_P} \quad , \tag{2}$$

where γ_P and γ_S are the decay rates and R_P and R_S are the net excitation rates. (Note that R_P is most likely much larger than R_S.) At pressures of interest γ_P is approximately A_p, the Einstein spontaneous-emission coefficient.[17] But γ_S is equal to A_S only at the wings of the n_S velocity distribution; at the low velocities where the ground state is heavily populated, resonance photons are trapped and γ_S decreases considerably. Thus γ_S becomes velocity dependent, causing the population inversion to vary across the velocity profile [Eq. (2)]. The corresponding gain profile will be depleted near the atomic-center frequency, and may switch into the absorption phase there. It is even possible, depending on the details of atomic density and temperature, for laser oscillation to occur between two levels whose over-all population inversion, summed over velocities, is zero or negative! This mechanism may well be operative in other molecular dissociation lasers.

A complete treatment of radiation trapping between levels of different velocity distributions is not presently available. Nevertheless, a reasonable estimate which demonstrates the importance of the radiation trapping effect can be obtained from the theory of Holstein,[18] which treats the case of equal velocity distributions. For this purpose the velocity distribution of the $^3S_1$ level may be divided into three regions with respect to u_G, the average thermal velocity of oxygen atoms at room temperature. Over the central portion ($v \ll u_G$), where the ground state is heavily populated, the Holstein theory should predict γ_S with reasonable accuracy. At the wings ($v \gg u_G$) where there are no ground-state atoms, γ_S is given by its untrapped value A_S. In the intermediate region ($v \sim u_G$), γ_S should range between these two values in a slowly varying fashion. A complete theory would, of course, be needed to predict the detailed behavior of γ_S in the intermediate region. The present approach, however, adequately demonstrates the importance of the radiative trapping effect.

According to the Holstein theory, the effective decay rate in the presence of trapping in a long cylinder radius R is given by

$$\gamma_S = 1.60 A_S (n_G \sigma R)^{-1} [\pi \ln(n_G \sigma R)]^{-1/2} \, , \tag{3}$$

where $n_G \sigma R \gg 1$, n_G is the ground-state density, and σ is the absorption cross section for resonance radiation. The cross section is evaluated from

$$\sigma = \frac{g_S}{g_G} \, \pi \lambda_0^2 \, \frac{A_S}{k_0 u_G/\sqrt{\pi}} \, . \tag{4}$$

Here $k_0 = 1/\lambda_0$ is the propagation constant of the radiation at line center, $k_0 u_G$ is the Doppler width (in units of circular frequency) of ground-state atoms with mass M and most probable speed $u_G = (2kT_G/M)^{1/2}$ and temperature T_G, which we take to be 300 °K. The statistical weights are $g_S = 3$ and $g_G = 9$ in our case. Taking $u_G = 5 \times 10^4$ cm/sec, $\lambda_0 = (1/2\pi)$ 1300 Å, and $A_S = 380 \times 10^6$/sec,[6] we find

TABLE II. Splittings (cm^{-1}) between adjacent pairs of the four oxygen laser lines, as reported by various authors. See text for details.

	A-B	B-C	C-D
Patel *et al.*[a]	0.126	0.464	0.130
Feld[b]	0.11	0.48	0.09
Tunitsky and Cherkasov[c]	0.1	0.46	0.1
Present work, pulsed and cw	0.143	0.461	0.136

[a]See. Ref. 2. [b]See Ref. 16. [c]See Ref. 3.

$\sigma \approx 1.2\times10^{-13}$ cm^2.

From this value of σ and the knowledge of the ground-state atomic-oxygen density we can estimate the effect of trapping at the atomic line center. Although we were unable to accurately measure the oxygen pressure in our quasiflow system we estimate it to be at least 100 mTorr. (See discussion in Ref. 5.) Other workers give an optimum value of about 36 mTorr.[1,14] Taking the latter value as a conservative estimate, and assuming as little as 5% dissociation[19] so that $n_G \geq 10^{14}$ cm^{-3}, Eq. (3) with $R = 0.6$ cm yields a decay rate of $\gamma_S \leq 3\times10^7$/sec for low velocity atoms. This quantity increases to the untrapped value of $\sim 4\times10^8$/sec at the wings. We have, then, a lower-laser-level velocity distribution of width ~9000 °K (assuming its Doppler width to be the same as the measured width of the upper laser level) whose central portion is considerably built up over the background population by means of selective reabsorption. Since the upper-level decay rate[6] $\gamma_P = \sim 3\times10^7$/sec, $\gamma_S \leq \gamma_P$ over the region of reabsorption. Accordingly, the n_P/n_S ratio of Eq. (2) is reduced to near or less than unity there, killing the gain. In contrast, at the wings $n_P/n_S \simeq 14$, assuming $R_S \ll R_P$. Thus, the gain profiles of the three 8446-Å transitions will not be proportional to the spontaneous-emission profiles of Fig. 3(a), but instead will have depleted regions at the three line centers, as shown roughly in Fig. 3(b).

INFLUENCE OF SELECTIVE REABSORPTION ON THE OXYGEN FINE-STRUCTURE LASER OSCILLATIONS

Taking into account the overlap of the $^3P_2$-$^3S_1$ and $^3P_0$-$^3S_1$ gain curves, which produce a large combined peak (so that the laser oscillation there is produced by a superposition of gain from the $^3P_0$-$^3S_1$ and $^3P_2$-$^3S_1$ transitions), five gain peaks occur. Oscillation has been observed on all except the smallest ($^3P_0$-$^3S_1$) peak.

The line labeled D in Fig. 3(c) falls on the largest peak and is normally the strongest line. This line, which we find to be displaced 0.12 cm^{-1} above (higher frequency) the $^3P_2$-$^3S_1$ spontaneous-emission peak, was first observed by Bennett, Faust, McFarlane, and Patel,[1] who suggested that the shift was due to an unidentified ozone transition.[20] Shortly afterward, Patel *et al.*[2] observed a quartet of laser lines at 8446 Å in an argon-bromine discharge which was later identified by Tunitsky and Cherkasov[3] as being due to an oxygen impurity and was, therefore, the first observation of the four oxygen lines. Labeling the two $^3P_1$-$^3S_1$ lines as A and B and the remaining line on the low-frequency side of $^3P_2$-$^3S_1$ transition as C [Fig. 3(c)], four lines are observed, spaced as follows: A-B: 0.143 cm^{-1}, B-C: 0.461 cm^{-1}, C-D: 0.136 cm^{-1}. For each line, laser oscillation is observed to extend over a width (excluding the instrumental width of our Fabry–Perot interferometer) of between 0.01 and 0.05 cm^{-1}, the extent of the D line being greatest. This wide range of oscillation is a consequence of the exceptional breadth of the Doppler gain profiles due to the mode of production of excited oxygen atoms, and indicates that a large number of axial modes are simultaneously oscillating. This observation is supported by the fact that a succession of beatnotes between axial modes—out to the response-time limit of our photomultiplier—could be observed on a spectrum analyzer.

The usual relative intensities in a pulsed discharge are as indicated in Fig. 3(c), with the A line second strongest to the D line. A cw discharge produces slightly different ratios, but the spacing between the four lines remains the same within the resolution of our Fabry–Perot interferometer, indicating that at a given pressure the effects of selective reabsorption are not very different for pulsed and cw discharges.

As can be seen from Fig. 3(c), line A is always stronger than line B. Furthermore, it is found that lines A and B are placed slightly asymmetrically about the $^3P_1$-$^3S_1$ spontaneous-emission profile. These small asymmetries are discussed further in Ref. 5.

If our explanation of the depleted central region is correct, the width of this zone and therefore, the laser line spacing, should be sensitive to the oxygen partial pressure through the n_G dependence of Eq. (3). Higher oxygen pressures should enlarge the depleted region. Indeed, our data were taken at a considerably higher O_2 pressure than that of Patel *et al.*[2] and, as shown in Table II, our line splittings change relative to theirs in the expected direction: A and B are further apart, B and C are slightly closer together, and the C-D separation, which undergoes influences in opposite directions from the $^3P_2$-$^3S_1$ and $^3P_0$-$^3S_1$ transitions, increases, but less so than A-B. Tunitsky and Cherkasov[21] observed a 3-GHz shift in the position

of the D line by varying the O_2 pressure over a decade.

POSSIBLE APPLICATIONS TO OTHER SYSTEMS

The mechanism which leads to laser oscillation in atomic oxygen has wide potential applicability in other gas-laser systems. What is required is that the velocity distribution of the upper laser level be broader than that of the lower level so that with appropriate level lifetimes, population inversion can occur at the wings of the distribution even though inversion is not possible at the center. A possible system in which gain may be produced in this way is the H_α line of atomic oxygen at 6563 Å[22] in a H_2-Ne discharge cooled to liquid-N_2 temperature. In this case "hot" hydrogen atoms are produced in the $n=3$ state by the reaction

$$H_2 + Ne^* \rightarrow Ne + H + H_{n=3} + 0.05\ eV\ , \qquad (5)$$

with a cross section of $\sigma = 1.4\times10^{-15}$ cm^2.[23,24] As explained above, cooling the discharge should narrow the velocity distribution of the $n=2$ level without affecting the corresponding width of the $n=3$ level.

We have studied the hydrogen system utilizing standard absorption techniques to measure the neon metastable population as a function of gas pressure and discharge current, while at the same time monitoring the spontaneous-emission intensity of the H_α, H_β, and H_γ lines. The details of the experiment will be prepared for publication at a later time. The results indicate that in a 1-Torr neon discharge with 20 mTorr of H_2 at low currents (~12 mA), so as not to dissociate too much H_2 by electron collisions, a population inversion of about 4×10^7/cm^3 occurs at the wings of the $3P_{5/2}$-$2P_{3/2}$, H_α transition. The gain of the system is estimated to be about 0.5% per meter, so that for laser oscillation a discharge tube several meters long would be required.

Other possible laser systems based on this effect are also under consideration.

ACKNOWLEDGMENTS

We would like to thank Professor Victor George for a useful discussion on plasma parameters. This paper is dedicated to the memory of Jerry Adler, whose enthusiasm and energetic assistance gave the experiment an effective start.

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[20]Other experimental data, reported in Ref. 5, indicate the existence of a second shift, smaller than that due to the selective reabsorption effect discussed here. The shift is possibly attributable to the presence of O^{18} in natural abundance. (See Ref. 5 for details.) It is emphasized, however, that the shifts and line placements described here are predominantly due to selective reabsorp-

tion and overlap, as discussed above.

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$$Ne^* + H_2 \rightarrow Ne + H_2^{+\nu} + e,$$

$$H_2^{+\nu} + e \rightarrow H^* \ (n=3,4,5) + H + E_{K.E.}.$$

This mechanism is consistent with the recently observed "superradiant" laser oscillations at 4861.2 and 4340.6 Å tentatively assigned to the H_β and H_γ lines. See G. J. Dezenberg and C. S. Willett, IEEE J. Quantum Electron. QE-7, 491 (1971). Similar processes may also play an important role in producing atomic-oxygen laser oscillations observed in a Ne-O_2 discharge.

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Interactions among Multiple Lines in the 8446-Å Atomic-Oxygen Laser*

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The atomic-oxygen laser oscillates at four closely spaced frequencies on the gain profile of the 8446-Å fine-structure transitions. A set of experiments is reported which studies the competition among these four lines by suppressing one of them. The results are explained on the basis of the interaction of laser lines oscillating on Doppler-broadened transitions sharing a common lower level.

I. INTRODUCTION

The atomic-oxygen laser oscillates at four closely spaced frequencies on the atomic gain profile of the 8446-Å fine-structure transitions ($3p^3P_{1,2,0} - 3s^3S_1$). In the preceding paper[1] it was explained that because of *selective reabsorption* of uv resonance radiation emanating from the lower laser level, the gain is depleted at the central portion of each fine-structure transition, and laser oscillation occurs only at the *wings* of the two fine-structure transitions with highest gains. Figure 1 illustrates again the positions of the four laser lines relative to the spontaneous-emission profile of the three fine-structure transitions. These lines are labeled A, B, C, and D in order of increasing frequency, with A and B falling on opposite sides of the $^3P_1 - {}^3S_1$ transition, and C and D falling on opposite sides of the $^3P_2 - {}^3S_1$ transition. Line D is largest due to overlap of the high-frequency wing of the $^3P_2 - {}^3S_1$ transition and the low-frequency wing of the $^3P_0 - {}^3S_1$ transition.

The present paper reports a set of experiments which studies the interactions among the four laser lines by suppressing one of them (in a manner to be described below) and observing the intensity changes of the other three. It is found[2] that suppressing the strongest line D causes line C to greatly increase in intensity, line B to increase somewhat, and line A to decrease. This is illustrated in Figs. 2(a) and 2(b), which show scanning Fabry–Perot interferometer traces of the four laser lines. Similarly, blocking line C causes line A to increase in intensity and line B to de-

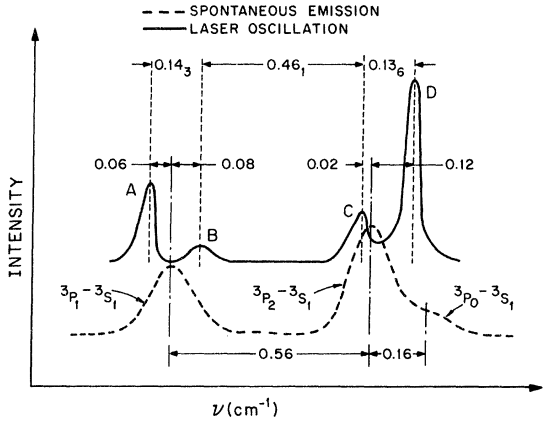

FIG. 1. The four laser lines near 8446 Å showing positions relative to the spontaneous emission of the three fine-structure transitions.