quasi-forbidden by much the same circumstance which makes its excitation unlikely [point (f)].

With regard to the possible relevance of the ± classification to atoms other than He, the complete degeneracy of the series limit is, of course, peculiar to two-electron atoms or ions. However, all states with two highly excited electrons will resemble the corresponding states of He in the same sense as all states of one-electron excitation belonging to Rydberg series resemble the H states. Doubly excited states may arise by mechanisms other than optical excitation. In particular, bombardment of neutral atoms with electrons of energy comparable to the ionization threshold might often result in the capture of the incident electron to form a two-electron state with an excited atomic electron. Evidence for this process is seen in the recent discovery of a resonance in elastic scattering by He at 19.3 eV,[4] in the theoretical prediction of similar resonances for H,[5] and in the behavior of the polarization of electron-induced light at a few volts above threshold, as interpreted by Baranger and Gerjuoy.[6]

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ISOTOPE SHIFT AND SATURATION BEHAVIOR OF THE 1.15-μ TRANSITION OF Ne†

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This Letter gives a summary of the results of an experiment with He-Ne optical maser in which the isotope effect together with the saturation behavior of one of the Doppler-broadened optical transitions of Ne, the $2p_4 - 2s_2$ transition[1] at 1.15 μ, are studied. An accurate value of the isotope shift due to Ne^{20} and Ne^{22} isotopes is measured, and the saturation parameters related to pressure effects and spontaneous radiative decays are determined.

The power output and the exact oscillation frequency of an optical maser depend on the saturation behavior of the atomic transition. These dependences may, hence, be used to study the details of the atomic line shape. This technique was adapted to the study of the 1.15-μ transition of Ne atoms. From the early stages of the experiment, it became evident that the slight distortion of the Doppler line shape due to the presence of 9% Ne^{22} with a small isotope shift introduced a considerable amount of ambiguity in the interpretation of the observed effects. In order to measure the isotope shift and to determine its effect on the over-all saturation of the resonance, two separate He-Ne optical masers were constructed containing isotopically enriched samples of Ne^{20} and Ne^{22}, respectively. The value of the isotope shift was determined as follows. Each maser was allowed to oscillate on a single axial mode. The oscillation frequency of each maser was then adjusted to coincide with the center of its respective Doppler-broadened transition. To achieve this, use was made of the saturation behavior to be described below. The outputs of the two masers were then heterodyned on the photocathode of a photomultiplier as described previously.[2] The difference frequency was measured by analyzing the frequency of the beat note at the output of the photomultiplier using a radio receiver. The value of the isotope shift was thus determined to be 261 ± 3 Mc/sec at 0.1 mm Ne, 1.0 mm He partial pressures, with the heavier isotope lying on the high-frequency side. In view of some unknown pressure shifts which are still subject to further studies, the quoted error is a conservative estimate.

The optical field within the maser resonators used in our experiments has the form of a stand-

ing wave. An atom may move a distance covering several wavelengths while it interacts coherently with this field. If the nonlinear effects due to saturation are ignored, the gain (or attenuation) of an applied standing optical field as a function of frequency has the usual Gaussian form of a Doppler-broadened line. This form is the same as in the case where atoms are subjected to a running wave. However, in the case of a standing wave, the atomic resonance saturates more readily at the peak frequency of the Doppler response.[3] As a result of this, in a gas maser operating on a single standing-wave mode, the peak frequency of the Doppler line does not correspond to maximum power output. Instead, a "dip" occurs in the power output versus oscillation frequency response which is centered at the peak frequency of the Doppler line.[4] The shape of the "dip" is determined by the relaxation processes and is related to the radiative lifetime of the levels and the details of the collision mechanism. Thus, a detailed study of the power dip as a function of various parameters, such as pressure, gives information on the saturation behavior and the relaxation processes of the active atoms.

Lamb has given a general formulation of the interaction of standing waves with atoms subject to thermal motions. His treatment applies rigorously at the limit of very low pressure. However, under our operating conditions, certain pressure effects, to be discussed below, become considerable and appreciable discrepancies from observations will result if they are not allowed for. The derivation given by Lamb may be adapted with slight modifications to include such effects. The resulting expression for the power gain of a Doppler line subject to a standing-wave optical field may be written, to the first order of dependence on the field amplitude, in the form

$$G = A\{\exp[-(\omega-\omega_0)^2/\Delta\omega^2]\} \times\left[1-\beta^2E^2\left(1+\frac{\gamma_{ab}'\gamma_{ab}}{\gamma_{ab}'^2+(\omega-\omega_0)^2}\right)\right].$$

In the limit when $E^2=0$, the above equation reduces to the usual expression for the unsaturated gain presented by a Doppler-broadened resonance. The parameter A is proportional to the population differences, ω_0 is the center frequency of the atomic resonance, and ω is the frequency of the standing optical wave. In the saturation term, $2\gamma_{ab}=1/T_a+1/T_b+h$ and $2\gamma_{ab}'=2\gamma_{ab}+s$, where T_a and T_b are the radiative lifetimes of the upper and lower levels; the parameters h and s are proportional to pressure and they arise due to atomic collisions to be described later below. The parameter β^2 is proportional to the square of the matrix element connecting the two levels and is also a function of the lifetimes of the two levels. In the present experiment, β^2 and A enter as scale factors and are not measured directly.

The steady-state oscillation of the optical maser corresponds to a value of E^2 for which the above expression for gain equals the losses of the cavity, φ. Thus, an expression for the power output, P, as a function of the oscillation frequency, ω, may be obtained by solving the equation $G=\varphi$ for E^2 and noting that P is proportional to E^2.

The tracings in Fig. 1(a) represent the power output versus the oscillation frequency of a He-Ne maser containing an isotopically enriched sample with 99% Ne^{20}. The maser was made to oscillate on a single mode and its length was varied continuously using magnetostriction.[5] Each tracing in this figure corresponds to a different excitation level of the discharge tube, hence, different values of the population difference, n_a-n_b. The tracings in Fig. 1(b) were recorded similarly except the maser contained a sample of Ne with normal isotopic abundance ($Ne^{20}=91\%$ and $Ne^{22}=9\%$). The abscissas in these tracings are proportional to the tuning current and were calibrated in terms of the frequency by tuning the interferometer length over at least three consecutive resonances of the interferometer. The known length of the interferometer, D, was then used to obtain the necessary calibration using the equation $\nu_{n+1}-\nu_n=c/2D$, where c is the velocity of light and $\nu_{n+1}-\nu_n$ is the frequency separation of two consecutive interferometer resonances. This was done only to within an accuracy of a few percent, sufficient for the purposes of this experiment.

The masers with pure Ne isotopes were used for determination of the saturation parameters, γ_{ab}' and γ_{ab} in accordance with the above theory. The experiment was repeated at several pressures of the gas mixture, keeping the ratio of the He and Ne partial pressures at a fixed value of ten to one. At each pressure, the tracings similar to those in Fig. 1(a) were recorded over a wide range of discharge excitation. At each pressure the parameters γ_{ab}' and γ_{ab} were determined by obtaining a fit of the observed tracings with the theoretical power versus oscillation frequency response. It was found that the resulting values

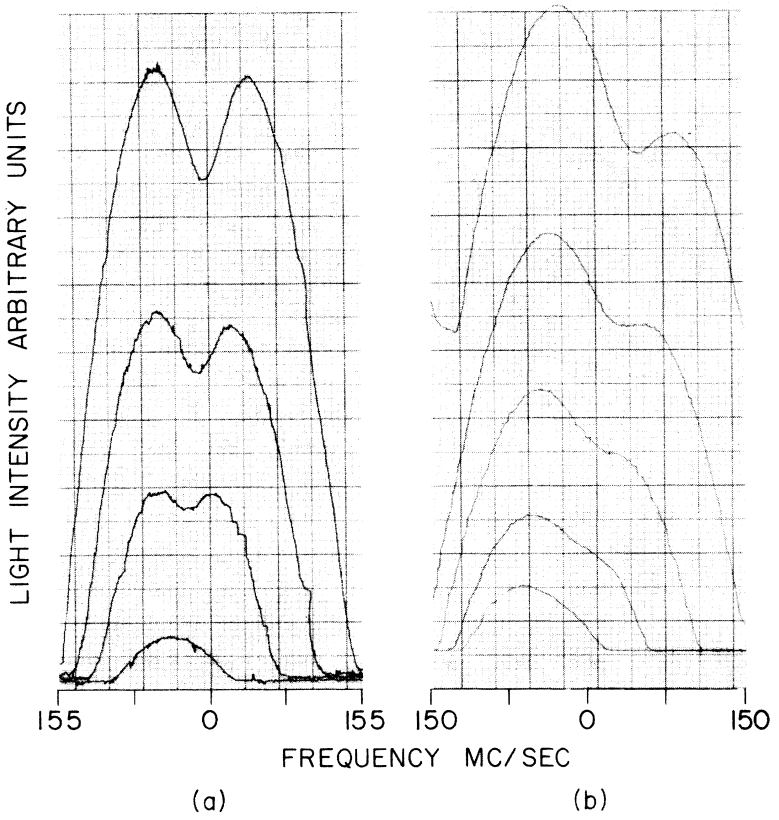

FIG. 1. Power output versus the oscillation frequency of He-He optical maser at 1.15 μ at varying excitation levels of the discharge tube. Figure 1(a) refers to a maser in which an isotopically enriched sample of Ne^{20} with 99% abundance was used. Figure 1(b) refers to a maser with normal isotopic abundance (91% Ne^{20}, 9% Ne^{22}). The total pressure is around 1 mm Hg with ten-to-one ratio of He to Ne partial pressures. Frequencies decrease to the right. The center frequency of the power dip in Fig. 1(a) is independent of the excitation level of the discharge tube. For normal isotopic species of Ne, Fig. 1(b), there is no characteristic frequency independent of power level.

of γ_{ab}' and γ_{ab} were rather insensitive to the choice of the Doppler width $\Delta\omega$. A value of 470 Mc/sec, corresponding to the calculated Doppler width at room temperature, was therefore adopted for $\Delta\omega$.

Figure 2 gives a plot of the measured γ_{ab}' and γ_{ab} as a function of the total He-Ne pressure. The pressure dependence of γ_{ab} and γ_{ab}' have several sources. The hard collisions during which the phase of the interacting Ne atom is completely interrupted contributes to the pressure dependence of γ_{ab}, which is represented by the parameter h introduced in the above. In the case of γ_{ab}', further dependence on the pressure is expected to exist due to a different type of collision. Consider small-angle scattering during which the wave function of the excited Ne atom remains essentially unaffected. In this collision, the velocity component along the axis of the optical wave suffers a small change leading to a small frequency shift due to change in Doppler effect. Such types of soft collisions are responsible for the presence of the parameter s in the expression for γ_{ab}'. The corresponding cross section may be obtained from the slope of γ_{ab}' versus pressure in Fig. 2. If the dominating mechanism is assumed to be due to Ne-He collisions, one obtains a value of 10^{-15} cm^{-2} for this cross section. The presence of the two parameters, γ_{ab} and γ_{ab}', in the saturation behavior is somewhat analogous to the presence of different T_2-type relaxation mechanisms which are encountered in radio-frequency spectroscopy.

The values of γ_{ab} and γ_{ab}' extrapolated to zero pressure converge to the same limit. From Fig. 2, this limit is $\gamma_{ab}=\gamma_{ab}'=10^8$ sec^{-1}. This is determined by the radiative rate of decays of the upper and lower maser levels. To within the limit of the accuracy of our measurements, this value is the same as the known[6] total rate of de-

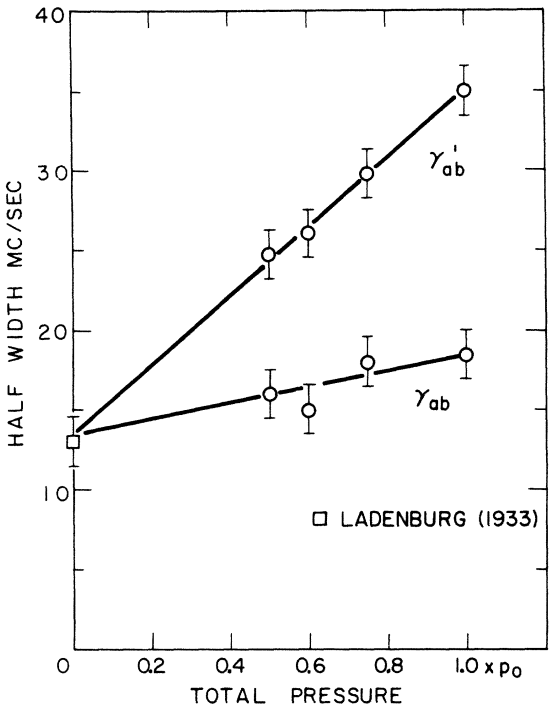

FIG. 2. Linewidth parameters at room temperature for the Ne $2p_4 \leftarrow 2s_2$ transition as a function of the total pressure of He-Ne mixture. The ratio of the partial pressures of He and Ne is kept constant, and P_0 corresponds to 1.0 mm Hg of He and 0.1 mm Hg of Ne.

cay of the lower maser level, $2p_4$. The total rate of decay of the upper maser level, $2s_2$, is due to its decay to several $2p$ levels and its decay to the ground state of Ne. The partial rate of decay of the $2s_2$ to the $2p$ level is known[7] to be 1.4×10^7 sec^{-1} and its contribution to γab is within the limit of our error. The rate of decay of the $2s_2$ to the ground state is not known. The above extrapolation of the low-pressure limit of γab gives an upper limit of around 10^8 sec^{-1} for this quantity.

The major difference in the expression for G appearing in the above and that given by Lamb[3] is the presence of the two linewidth parameters, γab and $\gamma ab'$ in the saturation term. At elevated pressures, the Lorentzian factor in this term is expected to take a more complicated form.

In the determination of the isotope shift, the center frequency of the resonance in each maser was reproduced by adjusting its length so that the corresponding power output coincided with the minimum of the power dip. The measured isotope shift together with the known normal abundance of Ne^{20} and Ne^{22} may be used to obtain an expression for the power versus the oscillation frequency of a normal Ne maser, in accordance with the above theory. The agreement of the observed tracings in Fig. 1(b) with theory using the saturation parameters determined above is excellent.

It should be noted that in the case of masers with pure isotopic species of Ne, the frequency corresponding to the minimum of the power dip is independent of the operating power level and hence independent of the excitation level of the discharge tube. The frequencies of the two maxima, however, depend strongly on the power level. As a result of this, in masers with normal Ne sample, the slope of the power versus the oscillation frequency response may not be used to define a characteristic frequency independent of the excitation level of the discharge. In this case, good accuracy in the frequency reproducibility may be obtained only if the maser is made to operate extremely close to the oscillation threshold. In masers with pure isotopic Ne samples, however, the center frequency of the dip may be reproduced independent of the excitation level of the discharge. Such a maser is, hence, more suited for its use as standard of length.

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[1]Paschen notation. The configuration for $2s_2$ is $2p^5(^2P_{1/2}^\circ)4s(J=1)$, and for $2p_4$ it is $2p^5(^2P_{1/2}^\circ)3p(J=2)$. For notation, see C. E. Moore, Atomic Energy Levels, National Bureau of Standards Circular 467 (U. S. Government Printing Office, Washington, D. C., 1949), Vol. 1.

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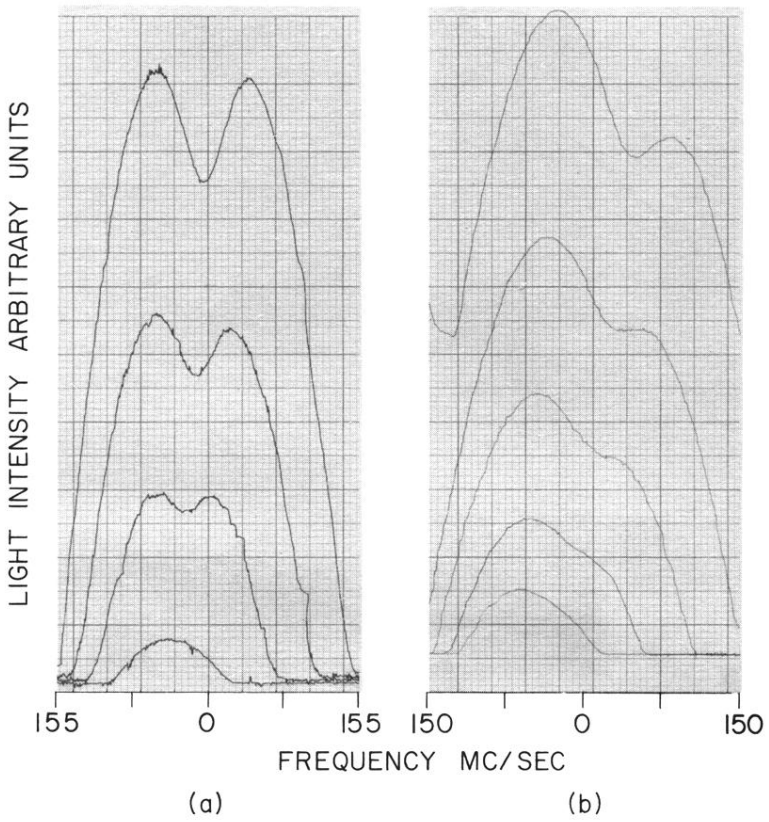

FIG. 1. Power output versus the oscillation frequency of He-He optical maser at 1.15 μ at varying excitation levels of the discharge tube. Figure 1(a) refers to a maser in which an isotopically enriched sample of Ne^{20} with 99% abundance was used. Figure 1(b) refers to a maser with normal isotopic abundance (91% Ne^{20}, 9% Ne^{22}). The total pressure is around 1 mm Hg with ten-to-one ratio of He to Ne partial pressures. Frequencies decrease to the right. The center frequency of the power dip in Fig. 1(a) is independent of the excitation level of the discharge tube. For normal isotopic species of Ne, Fig. 1(b), there is no characteristic frequency independent of power level.