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Determination of Atomic Temperature and Doppler Broadening in a Gaseous Discharge with Population Inversion

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The Doppler linewidth of the $5d[3/2]_1^0-6p[3/2]_1$ transition of xenon (Racah notation) at 2.026 μ in a helium-xenon discharge has been measured directly. Earlier, this transition has been shown to exhibit large optical amplification. The Doppler linewidth determination has been accomplished by measuring the optical gain in a helium-xenon discharge tube as a function of frequency. A short maser oscillator containing the same gas mixture is used as a tunable source of monochromatic radiation. The tuning has been accomplished by changing the length of the optical cavity and thus sweeping a cavity resonance across the Doppler-broadened gain curve. The source oscillator has been operated on a single longitudinal and transverse mode of the cavity to ensure as monochromatic source signal as possible. The measurements have yielded a Doppler linewidth of (210±10) Mc/sec corresponding to an average atomic temperature of 515°K±10%. It is seen that the actual line shape measured coincides very closely to a pure Doppler broadening with a Doppler width of 210 Mc/sec. This indicates that for the $5d[3/2]_1^0-6p[3/2]_1$ transition of xenon, the primary source of broadening is Doppler broadening. From this, an upper limit has been assigned to the line broadening due to finite lifetime and due to isotope shift effects.

INTRODUCTION

THE Doppler width of an atomic transition in a gaseous discharge is a very important parameter since it is a direct consequence of the thermal motion of the atoms, and thus indicates the atomic temperature. When one measures the Doppler width, one actually determines the average atomic temperature. We report here the direct determination of the Doppler broadening of the atomic line of xenon at 2.026 μ corresponding to the $5d[3/2]_1^0-6p[3/2]_1$ transition which has been reported earlier to have exhibited optical gain in pure xenon and in helium-xenon discharges.[1,2] From the high optical gain measured in the helium-xenon discharge, the Doppler-broadened linewidth is expected to be about 185 Mc/sec for the above transition for an assumed atomic temperature of 400°K. We have measured the Doppler-broadened linewidth to be (210±10) Mc/sec and a corresponding atomic temperature of 515°K±10%. The measurements indicate that for various levels of rf excitations of the discharge the measured values of the Doppler width and the atomic temperature shows no variation, as long as population inversion and the consequent optical gain at the 2.026 μ is maintained.

To be able to determine linewidth of 200 Mc/sec at a wavelength of 2 μ requires a resolution in excess of one part in a million. With the conventional use of spectrometers and interferometers such a resolution is not impossible to achieve but is certainly by no means easy. But with gaseous discharge media such as xenon where optical gain has been exhibited on at least one of the atomic transitions, the determination of the Doppler linewidth, and of the atomic temperature is possible with an entirely novel technique of measurements which bypasses the need of high-resolution spectrometers and interferometers in the conventional meaning of the word. The method of measurement described below also makes it possible to determine the lineshapes of the atomic transitions, especially in the infrared region beyond 2 μ where a number of atomic transitions exhibiting population inversion have been reported.[3] The broadening of the atomic transition in a low-pressure discharge is due to two major causes: the Doppler broadening due to the thermal motion of the atoms, and the natural broadening due to radiation damping. Since the Doppler broadening is proportional to the frequency of the transition, at wavelengths longer than about 2–3 μ, the natural broadening will make a significant contribution to the total linewidth of a transition. At lower wavelengths, as also in the measurements described here, the Doppler broadening is much larger than the natural broadening and, hence, the contribution of natural broadening to the total broadening is small. The line-shape measurement on xenon at 2.026 μ substantiates this statement as we shall see. At longer wavelengths, this technique of measurements will allow one to determine the line shapes and, consequently, the relative importance of the two sources of line broadening.

EXPERIMENTAL TECHNIQUES AND RESULTS

Figure 1(a) shows the Doppler-broadened curve for an atomic transition along with the cavity resonances which are separated by an amount equal to the interorder spacing of the optical resonator.[4] Since the cavity resonances are much narrower than the gain curve, the oscillation frequencies in such a maser would be determined primarily by the cavity resonances. Oscillations take place at each of the cavity resonances where the single pass gain exceeds the single pass loss in the oscilla-

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[1] C. K. N. Patel, W. R. Bennett, Jr., W. L. Faust, and R. A. McFarlane, Phys. Rev. Letters **9**, 102 (1962).

[2] C. K. N. Patel, W. L. Faust, and R. A. McFarlane, Appl. Phys. Letters **1**, 84 (1962).

[3] W. L. Faust, R. A. McFarlane, C. K. N. Patel, and C. G. B. Garrett, Appl. Phys. Letters **1**, 85 (1962).

[4] W. R. Bennett, Jr., Phys. Rev. **126**, 580 (1962).

tor. By making the cavity short, it is possible to obtain oscillation at a single frequency, i.e., only one of the cavity resonances would fall within that part of the gain curve where the net gain is greater than zero,[5] as shown in Fig. 1(b). The position of cavity resonance with respect to the gain curve maximum is, of course, determined by the cavity length. By changing the cavity length by small amounts, it will be possible to sweep the cavity resonance across the gain curve. This will lead to oscillations at the frequency of the cavity resonance as long as the net gain is above zero. Thus, it is possible to tune the oscillator-frequency over a part of the atomic transition and obtain a frequency-tunable source of monochromatic coherent radiation. The output of the maser oscillator will, of course, not be constant as we tune the cavity resonance across the transition. With this source one can measure the optical gain in another gas discharge tube as function of frequency, leading to a determination of the width of the atomic transition.

Figure 2 shows the sketch of tunable maser oscillator used in these experiments. The maser consists of an external mirror scheme with spherical mirrors. The maser tube is 25 cm long and 4–5 mm i.d. The mirrors forming the cavity are separated by 30 cm. The interorder spacing of the cavity resonances for this separation is 500 Mc/sec. An iris of about 2-mm diam is inserted within the cavity to allow oscillation only the fundamental transverse mode of the cavity.[5] Tuning of the maser is accomplished by keeping one of the mirrors fixed in position and moving the other one by small fractions of a wavelength. The fine motion of this mirror is obtained by way of magnetostriction.[6] Since the linear expansion of the magnetostrictive rod is linear with current over a small region of current, a voltage proportional to the current provides a frequency sweep signal from the device. The frequency calibration on the current scale is very simple since one knows the mirror separation. The frequency separation between two consecutive maxima in the power output corresponds to the interorder spacing of the cavity, which, here is 500 Mc/sec. A frequency tunability of the maser oscillator of about 350 Mc/sec has been obtained. The output from the maser is in a single transverse and longitudinal mode. The maximum output is approximately $\frac{1}{10}$mW.

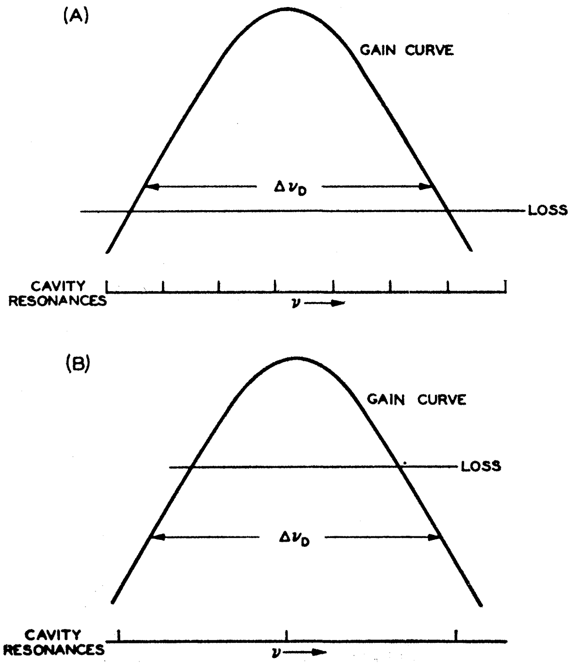

Fig. 1. Doppler-broadened gain curve and relative positions of cavity resonances for two cavity lengths.

Fig. 2. Sketch of the tunable maser oscillator used as the source.

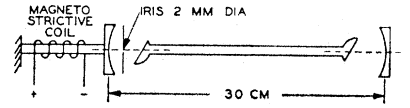

Figure 3 shows the experimental arrangement for measurement of gain as function of frequency. The optical-power level at A and at B is measured and is compared as a function of frequency. The instrumentation is arranged to give the ratio of B to A or the optical gain in decibels. The quantity $\ln(B/A)$ is automatically plotted against the current in the magnetostrictive tuning coil as the current is varied. As mentioned in the previous paragraph, this plot gives the optical gain of the helium-xenon discharge tube as a function of frequency. The gain measurements are valid only for that region of the tuning over which the oscillations are possible, i.e., over about 350 Mc/sec. Since the gain is plotted in decibels, the full width of the gain curve when the negative absorption coefficient is down by a factor of 2 is given by the frequency separation between the half-height points on the plot. Figure 4 shows a typical curve of gain versus frequency. On the same figure we have shown a Doppler-broadened curve with a full width of 210 Mc/sec.

Table I. Experimental results on Doppler-width measurements for a number of gas discharge conditions.

Gas pressures in Torr		Peak gain	Doppler width
Xe	He	(dB/m)	(Mc/sec)
0.035	4	2.25	206
0.04	4	3.28	200
0.035	4	3.55	209
0.05	4.8	4.14	215

The linewidth measurements were carried out at various rf excitation levels of the amplifier tube and also for various gas proportions in the discharge tube. The results are given in Table I which shows the average

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[5] H. W. Kogelnik and C. K. N. Patel, Proc. IRE **50**, 2362 (1962).

[6] W. R. Bennett and P. J. Kindlemann, Rev. Sci. Instr. **33**, 601 (1962).

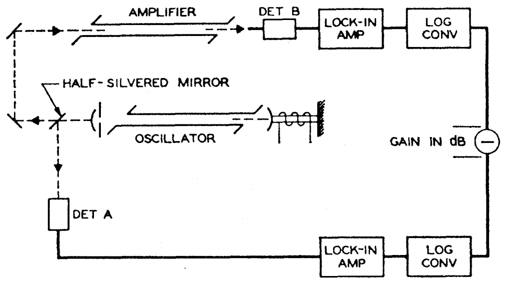

FIG. 3. Experimental arrangement used in the linewidth measurements.

of a number of measurements for each experimental condition. The maximum deviation of individual measurement from the reported average was less than ±5%. It can be seen from the results that the linewidth measured is independent of the excitation conditions.

DISCUSSION OF THE RESULTS

As mentioned earlier, there are two major contributions to the total linewidth measured. In Fig. 4 is plotted also the theoretical Doppler-broadened line with a Doppler full width of 210 Mc/sec. It can be noted that the experimentally measured gain variation as a function of frequency matches very closely a pure Doppler-broadened line. Figure 4, thus, substantiates our earlier statement as to the natural broadening being a very small contribution to the total linewidth. The natural linewidth contribution, however, will show up at frequencies far removed from the resonance—where the present method cannot be used. Other methods, such as the time dependence of the radiated transition can be used to determine the natural broadening. The linewidth measurements reported in Table I, hence, represent the Doppler broadening of the $5d[3/2]_1^0-6p[3/2]_1$ transition of xenon. Hence,

$$\Delta\nu_D=(210\pm10)\text{ Mc/sec.} \tag{1}$$

Now,

$$\Delta\nu_D=\frac{2\nu}{c}\left(\frac{2kT}{m}\ln2\right)^{1/2}, \tag{2}$$

where ν = frequency, c = velocity of light, k = Boltzmann's constant, T = average atomic temperature, and m = atomic mass. Taking the average atomic mass of xenon to be 131.3 amu, we obtain

$$T(\text{av})_{\text{atomic}}=515^{\circ}\text{K}. \tag{3a}$$

The uncertainty in the calculation of the average atomic temperature, if any, would arise from the fact that relative abundance of the various isotopes in the sample of xenon used in these measurements was not known. The error due to this uncertainty however, would be less than ±3%, which is within the experimental accuracy. Thus,

$$T(\text{av})_{\text{atomic}}=515^{\circ}\text{K}\pm10\%. \tag{3b}$$

It should be pointed out that we have seen from Fig. 4 that the measured line shape very closely matches the theoretical line shape assuming a full Doppler width of 215 Mc/sec and a pure Doppler broadening. This also eliminates possibility of error in interpretation of the experimental results due to isotope shift of the xenon levels in various natural isotopes. The very close match of the two curves in Fig. 4 indicates that the contribution of the isotope shift of various isotopes is negligible compared to the Doppler broadening. The maximum contribution of the possible isotope shift and of the natural linewidth to the total linewidth reported will be of the order of the experimental error limits.

The close match seen in Fig. 4 between the measured line-shape and an assumed pure Doppler-broadened line for $\nu-\nu_0\lesssim\Delta\nu_D$, where $\nu-\nu_0$ is the frequency deviation from the center of the line, allows us to assign an upper limit to the extent of natural linewidth of the $5d[3/2]_1^0-6p[3/2]_1$ transition of xenon. Referring to the calculations of a Doppler-broadened line with small natural broadening as given by Mitchell and Zemansky[7] we can see that the compound broadened line deviates from a pure Doppler-broadened line in the region of $\nu-\nu_0\leq\Delta\nu_D$ by less than 5% even when the quantity 'a' is as large as 0.05, i.e.,

$$\frac{\Delta\nu_n}{\Delta\nu_D}(\ln2)^{1/2}\leq0.05, \tag{4}$$

where $\Delta\nu_n$ = natural linewidth. Thus,

$$\begin{aligned}&\Delta\nu_n\lesssim0.06\Delta\nu_D,\\&\Delta\nu_n\lesssim13\text{ Mc/sec.}\end{aligned} \tag{5}$$

This upper limit of 13 Mc/sec seems quite reasonable when one compares it with estimates of $\Delta\nu_n$ for neon maser transitions.

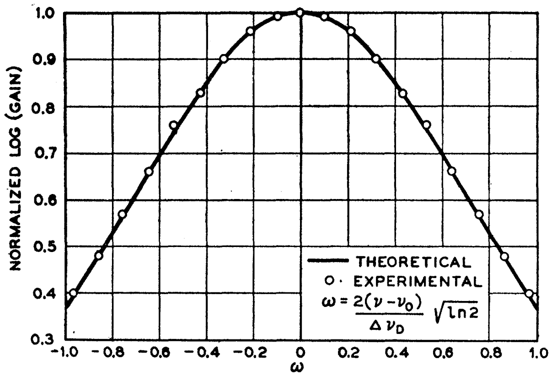

FIG. 4. Typical experimental results with a theoretical Doppler-broadened line with $\Delta\nu_D=210$ Mc/sec plotted as a function of $\omega=[2(\nu-\nu_0)/\Delta\nu_D](\ln2)^{1/2}$, where ν_0 = center frequency of the atomic transition and ν = frequency of measurement.

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[7] A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (Cambridge University Press, New York, 1961), Chap. III.