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and August 1, 1966 to January 31, 1967

Title

Study of Various Aspects of Raman Scattering  
using Continuous Wave Optical Masers

under supervision of

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Telephone: Un 4-6900, Ext. 5088

Contractor:

Massachusetts Institute of Technology  
Cambridge, Massachusetts

under

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The progress report is in two parts:

Part 1 contains a report of the continuation of our earlier experiments on motional narrowing of the linewidth of Raman scattering in hydrogen molecules. This narrowing, which has now been observed in spontaneous Raman scattering is being studied in detail. The report contains also details of the instrumentation of the Raman spectrometer which utilizes a single mode, frequency stabilized, high power Argon laser.

Part 2 reports, for the first time, development and application of a new spectroscopic technique to ultra-high resolution studies of atomic line centers. It involves a line narrowing effect induced by laser radiation and the measured quantities are precise values of isotope shifts in levels of Ne.

## MOTIONAL NARROWING OF RAMAN SCATTERED LINEWIDTHS IN $H_2$

J. Murray and A. Javan

This report gives further details of our recent observations on motional narrowing of the linewidth of Raman scattered emission of  $H_2$ . In this experiment, a single mode Argon laser is used and the line narrowing of spontaneous Raman scattering is observed using a precision Fabry-Perot interferometer. We have improved considerably the overall instrumentation of our Raman spectrometer. These improvements have included the use of feedback circuits to achieve frequency stabilization of the single mode Argon laser, and improvement of the overall detection sensitivity. The accuracy of our measurements has improved considerably compared to our initial observation of this effect. Our existing instrumentation is expected to allow determination of additional related effects and the exact shape of the pressure narrowed line.

The distribution in velocity of molecules in a gas causes Doppler broadening of an emission line. If the radiating molecule collides with other molecules during its radiation, the Doppler width is affected: It can be shown that if the molecule is confined by collisions to displacements on the order of a wavelength or less during its radiation the Doppler broadening is eliminated. <sup>(1)</sup> This is illustrated in Figure 1.

Hydrogen gas is known to have small pressure broadening in both its vibrational and rotational transitions, so it is a good candidate for detection of collision narrowing. These lines are electric quadrupole transitions and thus very weak in direct spectroscopy. The present experiment detects collision narrowing in spontaneous Raman scattering from the Q(1) rotation-vibration transition in hydrogen at  $4155 \text{ cm}^{-1}$ .

#### Apparatus

Figure 2 shows the apparatus used in this experiment. A  $4880\text{\AA}$  argon ion laser capable of a multimode power output of about 1w or a single mode output of about 100 mw is used to excite the Raman spectrum. The laser beam is directed by mirrors into a high pressure scattering cell. Raman scattering in the backward direction<sup>(2)</sup> is collected by a lens and passed through a spectrometer consisting of a pressure-scanned Fabry-Perot interferometer and a set of bandpass interference filters to pass the Q(1) Raman scattered line at  $6130\text{\AA}$ . The output of the spectrometer is focussed onto the very small (2.5mm dia) photocathode of an ITT Fw-130 photomultiplier. A lock-in detection system or a simple pulse integrator can be used as a detector: for the conditions of this experiment they give much the same results. The spectrum is presented on a strip chart recorder. Integration times are typically 3 seconds, and the signal to noise ratio at 10 atmospheres pressure and about 30mw laser

output (multimode) is typically 10:1. The signal to noise ratio is largely limited by laser amplitude fluctuations at this pressure. Photon statistics become important at lower pressures. A signal to noise ratio of at least 5:1 is required for any reasonable linewidth measurements. When well aligned, this apparatus provides about one hundred photoelectrons per second in the Q(1) line at a laser power of 30 milliwatts and a H<sub>2</sub> pressure of 10 atmospheres. Alignment is rather critical.

The scattering cell (Figure 3) has been described in a previous progress report but will be mentioned briefly here. It is a conventional high pressure cell containing a .5mm I. D. quartz capillary 30 cm long. The laser beam is directed through the capillary, and the Raman scattered radiation at small angles in the forward or backward direction is reflected at grazing incidence on the walls of the capillary to emerge at the ends. This design allows a large scattering volume while maintaining the small cross-sectional area needed for imaging the light through the spectrometer with high efficiency.

#### Laser Linewidth

The resolution of the apparatus is limited by the argon laser linewidth. A four mirror Michelson interferometer resonator allows a long resonator to have much higher frequency selectivity than the standard Fabry-Perot resonator<sup>(4)</sup> (Figure 4). Very briefly, the laser oscillates only when a mode of the 80cm resonator coincides with a mode of the 8cm resonator, and other modes of the 80cm resonator are suppressed strongly.

The laser of this experiment is now operating in such a resonator. A few tens of milliwatts are available on a single mode of the 80 cm resonator, giving a linewidth  $\leq 200$  MHz (allowing for jitter due to vibrations). At higher power outputs (up to  $\sim 120$  mw) the laser operates on a group of two or three adjacent modes of the 80 cm resonator, giving a width  $\leq 600$  MHz. Oscillation on the wings of the  $4880\text{\AA}$  line at the next coincidence of the two resonators can also be seen, but it is of low-power and not a serious problem. This resonator has been seen to operate on the  $5145\text{\AA}$  line of argon as well, with lower output power, but on no other lines.

At the present stage of development of more sophisticated FM laser single mode techniques, this system can compete fairly well, putting about 10% of the  $4880\text{\AA}$  output into a bandwidth on the order of 300 MHz, as compared to about 50% of the  $5145\text{\AA}$  output into 300 MHz with FM techniques and a rather more expensive and complicated system. <sup>(4)</sup> The FM technique has not been sufficiently well developed to operate on  $4880\text{\AA}$  as of the latest publications in the field. It is likely that the FM laser system will ultimately be better.

The laboratory in which this laser is operating is rather noisy, hence frequency jitter and drift caused by mechanical vibrations is a serious problem, though the system can be used for many purposes without stabilization. Such jitter makes the rather bad amplitude fluctuations normally present in the laser worse. Stabilization will be discussed in a later section of the report.

Preliminary Results <sup>(5)</sup>

In Figure 5 are shown the linewidths measured in the apparatus for three different runs under different conditions of laser power output and different Fabry-Perot resolutions in the spectrometer. Run 1 was made with a high power laser (500 mw) giving a laser linewidth  $\sim 4.5$  GHz and a Fabry-Perot interorder spacing  $\sim 48$  GHz, finesse 15-20. Run 2 was made under conditions of smallest linewidth consistent with reasonable output power ( $\sim 30$  mw) in a multimode laser, giving a laser linewidth of  $\sim 2.5$  GHz, and the same Fabry-Perot as above. Run 3 was made with an unstabilized Michelson-type single mode laser with linewidth .6 GHz if low frequency jitter runs are selected (power output  $\sim 40$  mw) and a Fabry-Perot of 24 GHz, interorder spacing, finesse 15-20.

Figure 6 shows these data plotted on a log-log-scale for ease in comparison with the results recently announced by Lallemand and others <sup>(6)</sup> is stimulated Raman scattering from hydrogen and with a simple theoretical mode of collision narrowing. The data are plotted as taken, and also with the instrumental linewidth determined by the plot of Figure 5 subtracted.

There is fair agreement with both Lallemand data and the theoretical curve (parameters adjusted to fit Lallemand's points).

It is possible to say from these results that the collision narrowing effect does indeed exist and bears some similarity to what theoretical treatments to date predict, but the results are not good enough to answer any questions in detail. Better instrumental resolution and noise performance adequate to go to somewhat lower pressures are required.

### Stabilization of the Laser in Frequency and Amplitude

The problems arising from the laser power drift and frequency jitter have been described earlier. Stabilization systems are being developed to reduce these effects.

Frequency stabilization to the tolerances required by this experiment (on the order of 100 MHz) is not difficult: One must exert some damping effect on vibrations and compensate for any thermal drifts or mechanical creep present in the Michelson resonator. A system which serves this function is pictured in Figure 7. It performs well within the necessary tolerances. A simple optical frequency discriminator based on a sealed Fabry-Perot with invar spacer provides an error signal which controls a piezoelectric element positioning one of the mirrors.

Amplitude stabilization presents technical difficulties. The laser has two general types of amplitude noise, fast ( $\sim 60$ hz) variations due to plasma situations, ripple on the power supply, and vibrations; and slow (seconds) drifts. The fast variations can be as much as 30% of the output power. The slow drifts are typically 10-15% in minutes, with about 5% short term (few seconds) jitter. The Michelson resonator system is similar but somewhat worse. The fast fluctuations do not affect this experiment. They do complicate any feedback regulation, however: if they are corrected by the regulator the active range of the regulator must be much larger, and if they are not to be corrected, problems arise in the design of the feedback loop. These are two ways in which an amplitude



control can be applied to the laser, either by regulating the laser discharge current or by controlling the output beam with a variable attenuator. Large powers must be handled by a laser current regulator (typically  $\sim 100V$  at 30A), which is a difficult technical problem, so the variable attenuator approach has been selected, (Figure 7) using a Kerr cell as the variable element. Unfortunately this causes losses which reduce the useful power output. The technical problems with this regulator have not all been solved, but it should be operating soon.

#### Future Plans

The signal to noise ratio and resolution of the apparatus will be sufficient to get good data on the collision narrowing of the Q(1) line as soon as the regulation systems are working well. There have been some delays in equipment delivery and laser discharge tube failures which have slowed the experiment. If the signal appears to be adequate, we then plan to look at rotational lines in  $H_2$  and perhaps at lines in Deuterium. The spectrum may be examined in the forward direction if the resolution is adequate.

A detailed theoretical treatment of collision narrowing in Raman scattering must be developed as it has not been treated and is rather more complex than the simple cases considered so far. (1)

REFERENCES

1. R. Dicke, Phys. Rev., 89, 472 (1953); L. Galatry, Phys. Rev., 122, 1218 (1961); and Rautian and Sobelman, Lebedev Institute (preprint).
2. In the backward direction one expects a Doppler shift  $\frac{\Delta\nu_B}{\nu_B} = \frac{v}{c}$  where  $\nu_B = \nu_{\text{laser}} + \nu_{\text{Stokes}}$ ; whereas in the forward direction one has  $\frac{\Delta\nu_F}{\nu_F} = \frac{v}{c}$  where  $\nu_F = \nu_{\text{laser}} - \nu_{\text{Stokes}}$ , giving for this experiment a Doppler width 9 times less. Thus much higher resolution is required. Also, the laser beam impinging directly on the spectrometer causes background problems.
3. P. Smith, IEEE, JQE, 1, 343 (1965).
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5. J. Murray and A. Javan, Bull Am. Phys. Soc., Series II, 12, 1 (1967).
6. Lallemand, Simova and Bret, Phys Rev. Letts., 17, 1239 (1966).

# COLLISION NARROWING

Theoretical Treatments  
simple treatment-

R. H. Dicke Phys. Rev. 89, 472 (1953)

more detailed treatments -

L. Galatry Phys. Rev. 122, 1218 (1961)

Rautian & Sobel'man of the Lebedev Institute

- 10 -

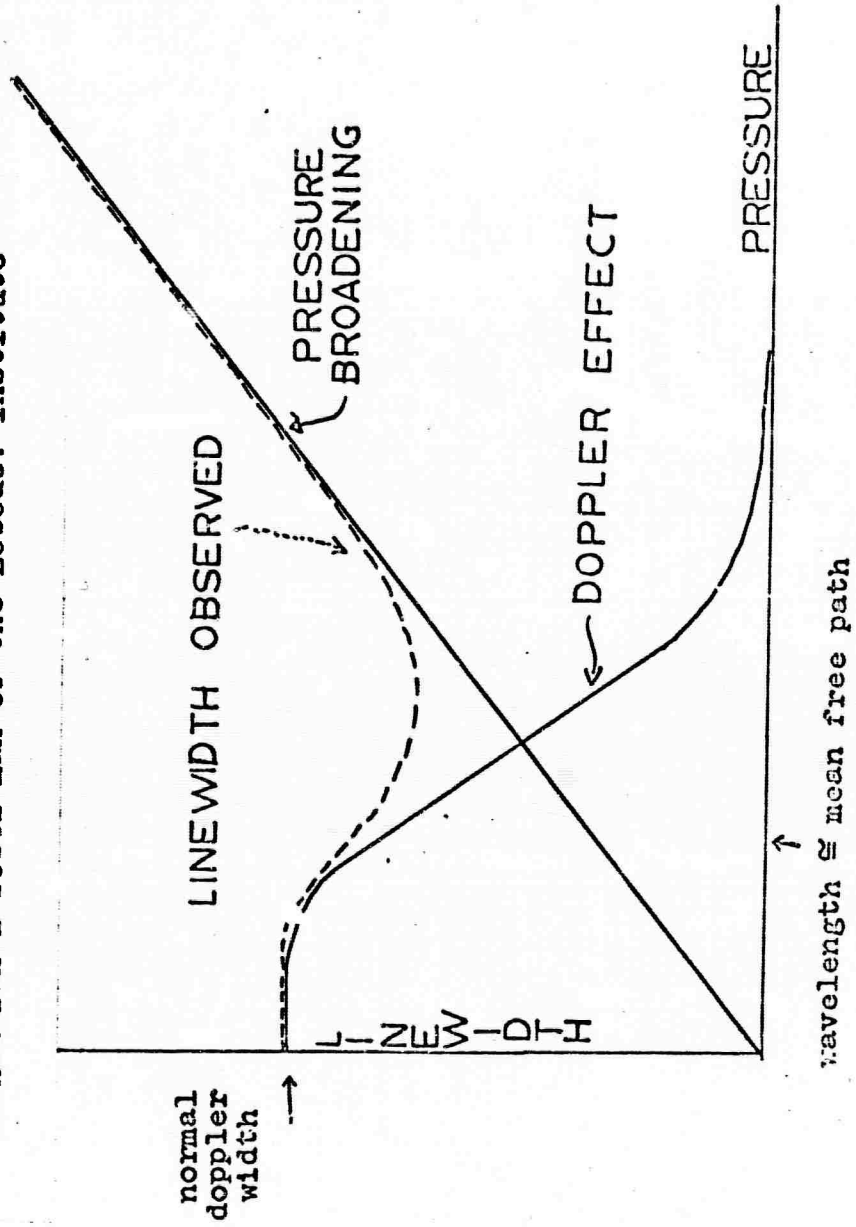
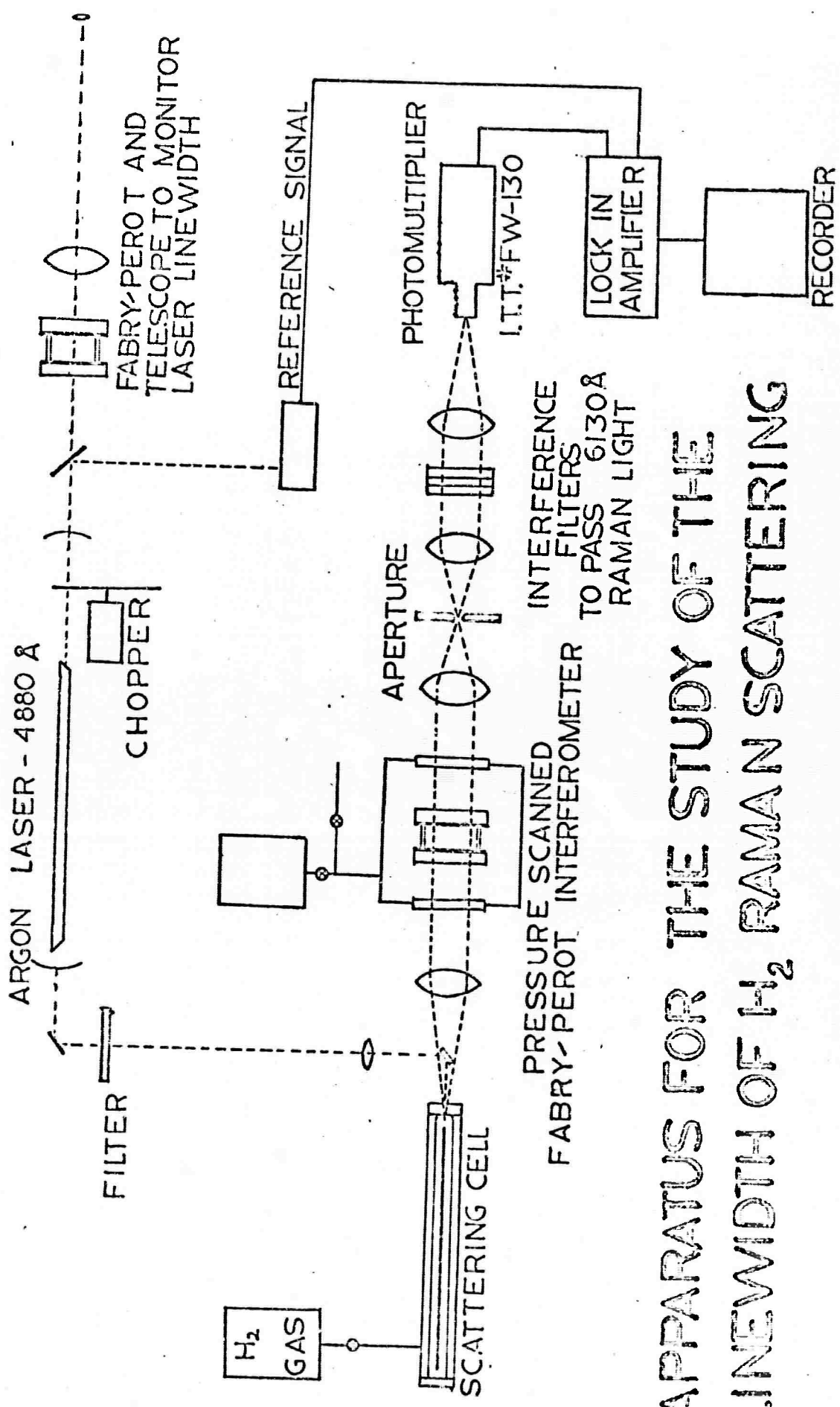
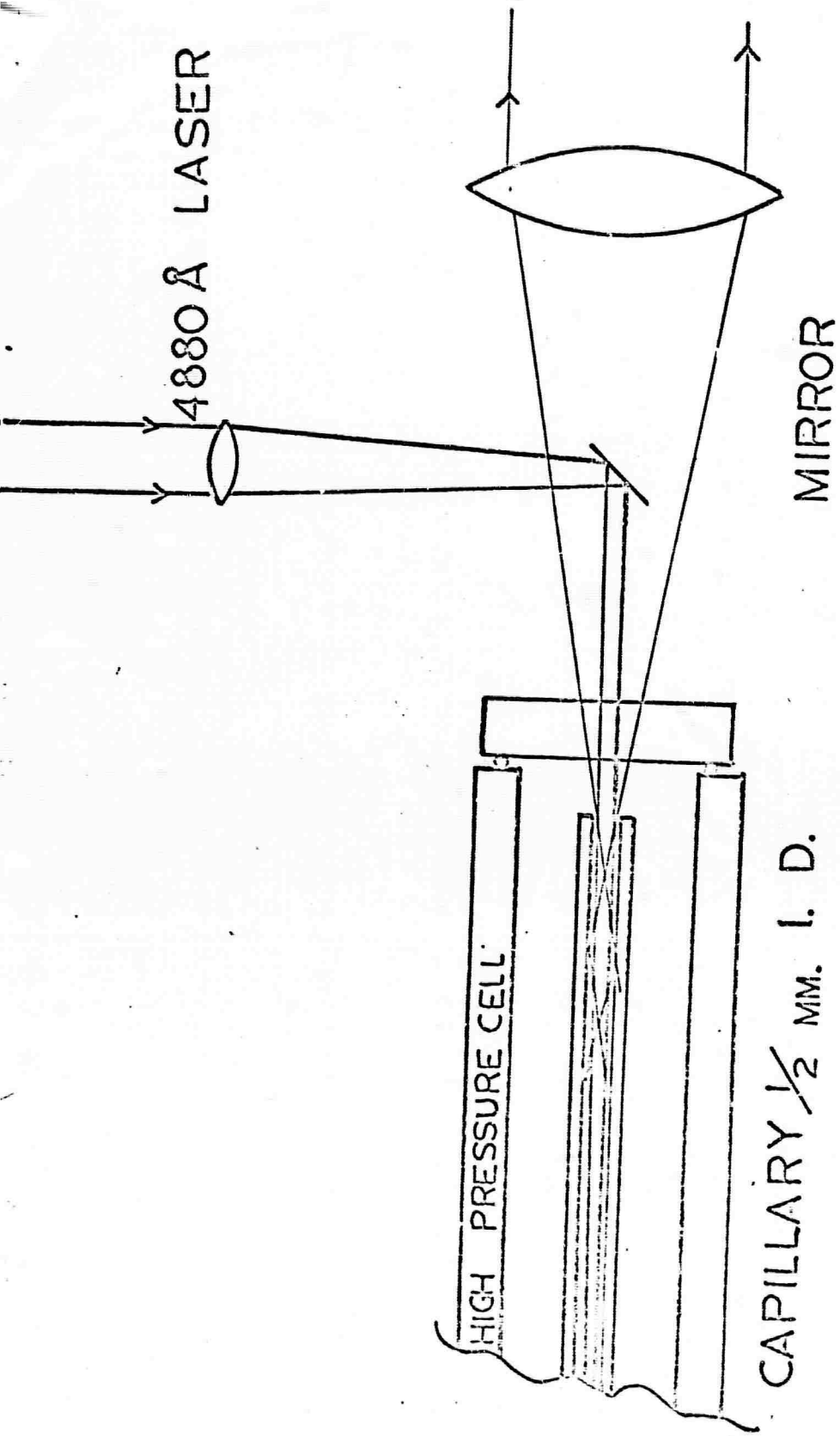


FIG. I



# APPARATUS FOR THE STUDY OF THE LINEWIDTH OF H<sub>2</sub> RAMAN SCATTERING

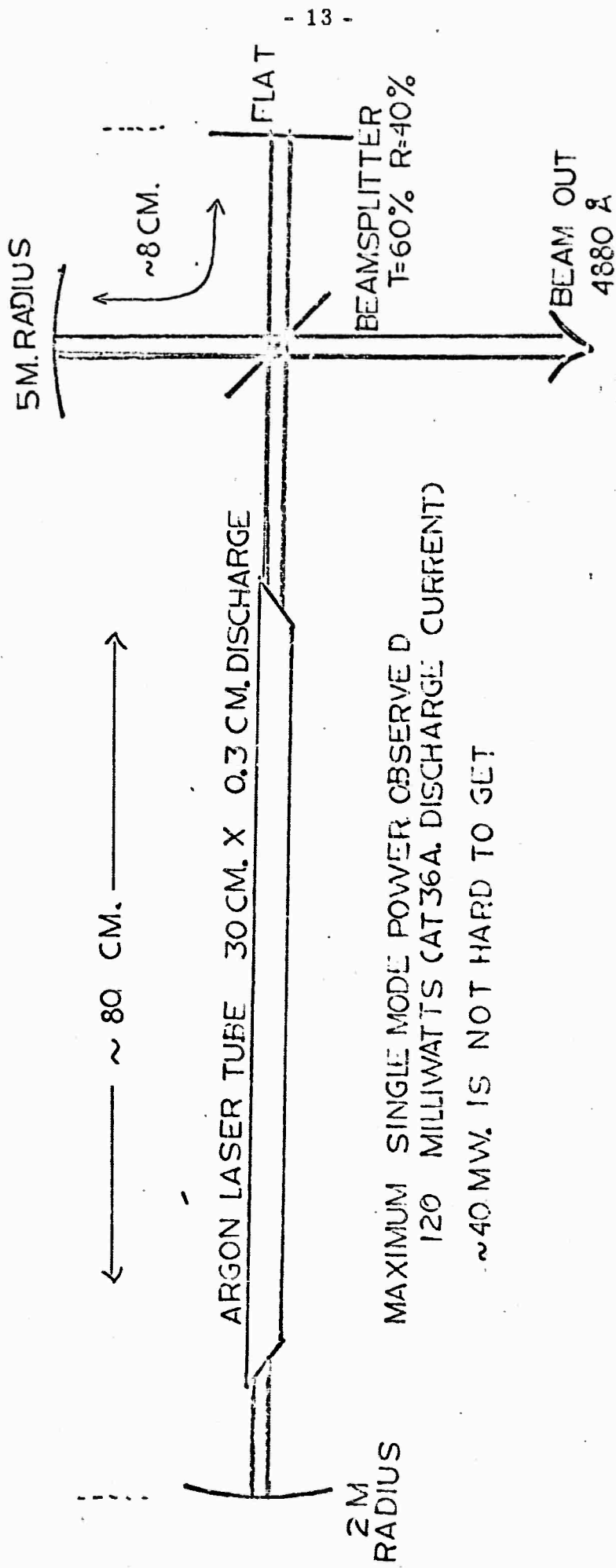
FIG. 2



DETAIL OF SCATTERING CELL

RAMAN SCATTERED LIGHT IS REFLECTED  
AT GRAZING INCIDENCE ON INSIDE WALL  
OF CAPILLARY

FIG.3



## SINGLE AXIAL MODE ARGON LASER

IN A "MICHELSON" INTERFEROMETER RESONATOR

[SEE: P. SMITH, J. QUANTUM ELECTRONICS 1, 343 (1965)]

FIG. 4

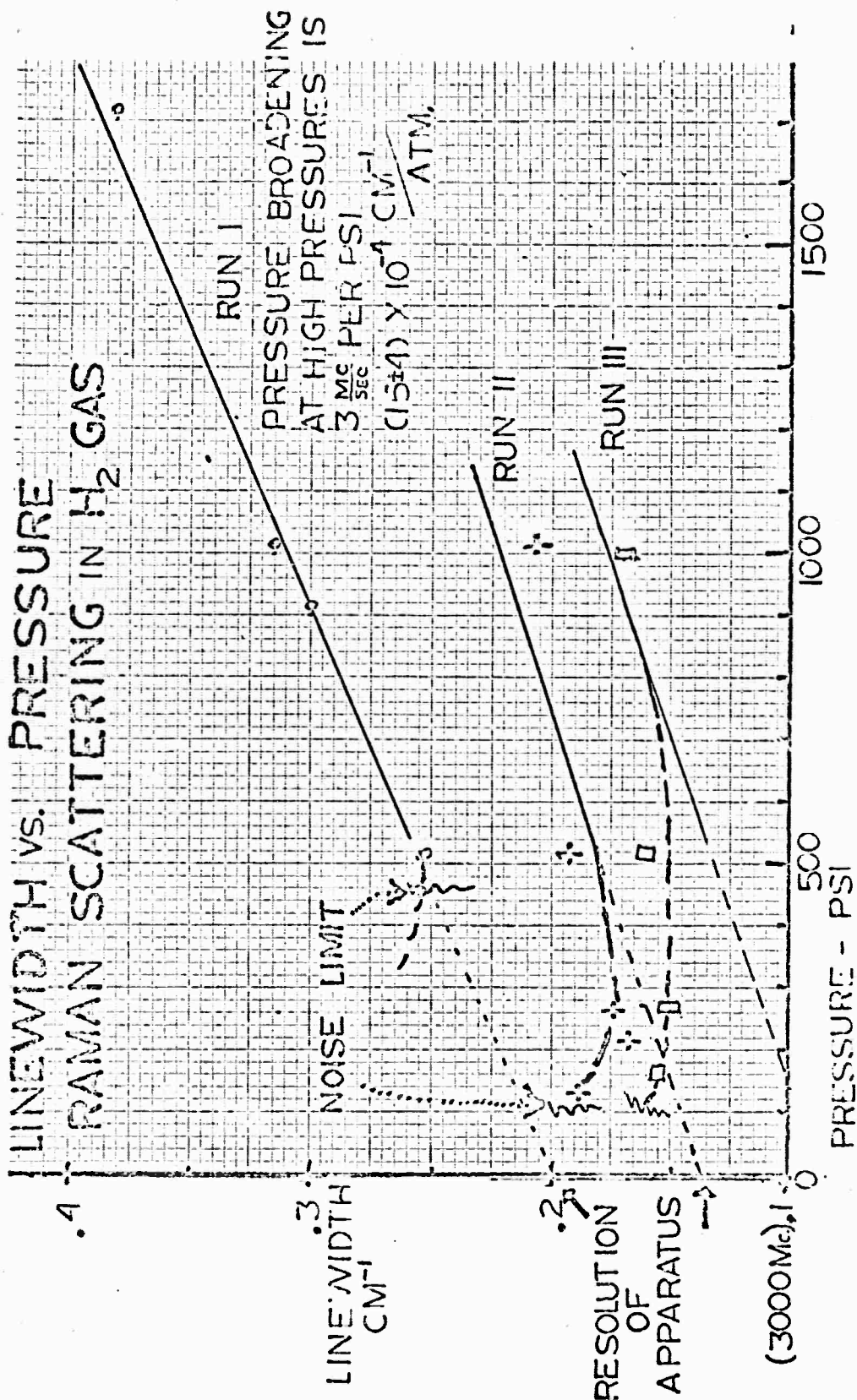


FIG.5



DATA COMPARED TO:

RESULTS OF LALLENAND, SINOVA, & BRET (PHYS. REV. LETT. 17, 1239 (19 DEG '66))

SOLID CURVE - BEHAVIOR PREDICTED BY GALATRY (PHYS. REV. 122, 1218 (1951))

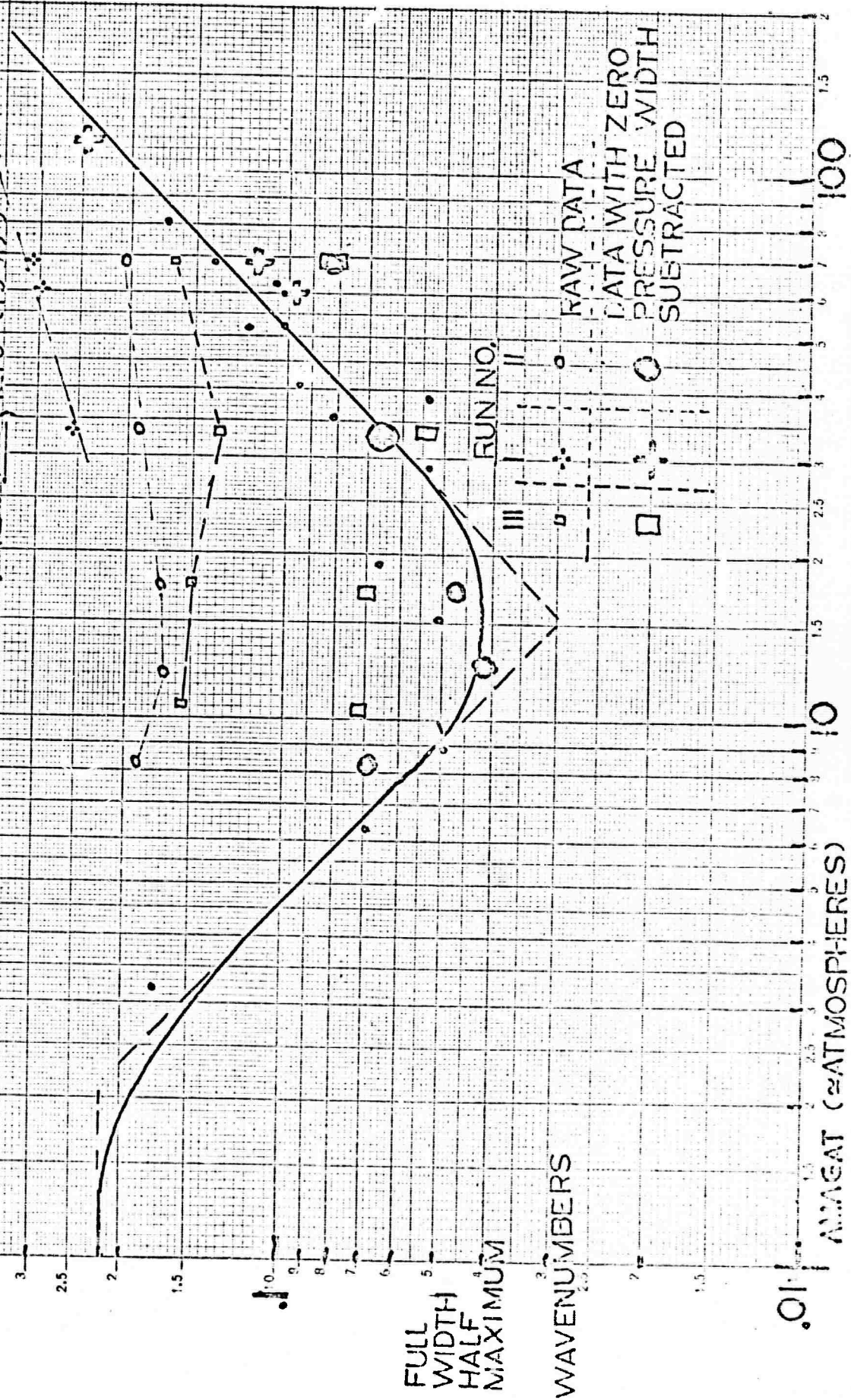
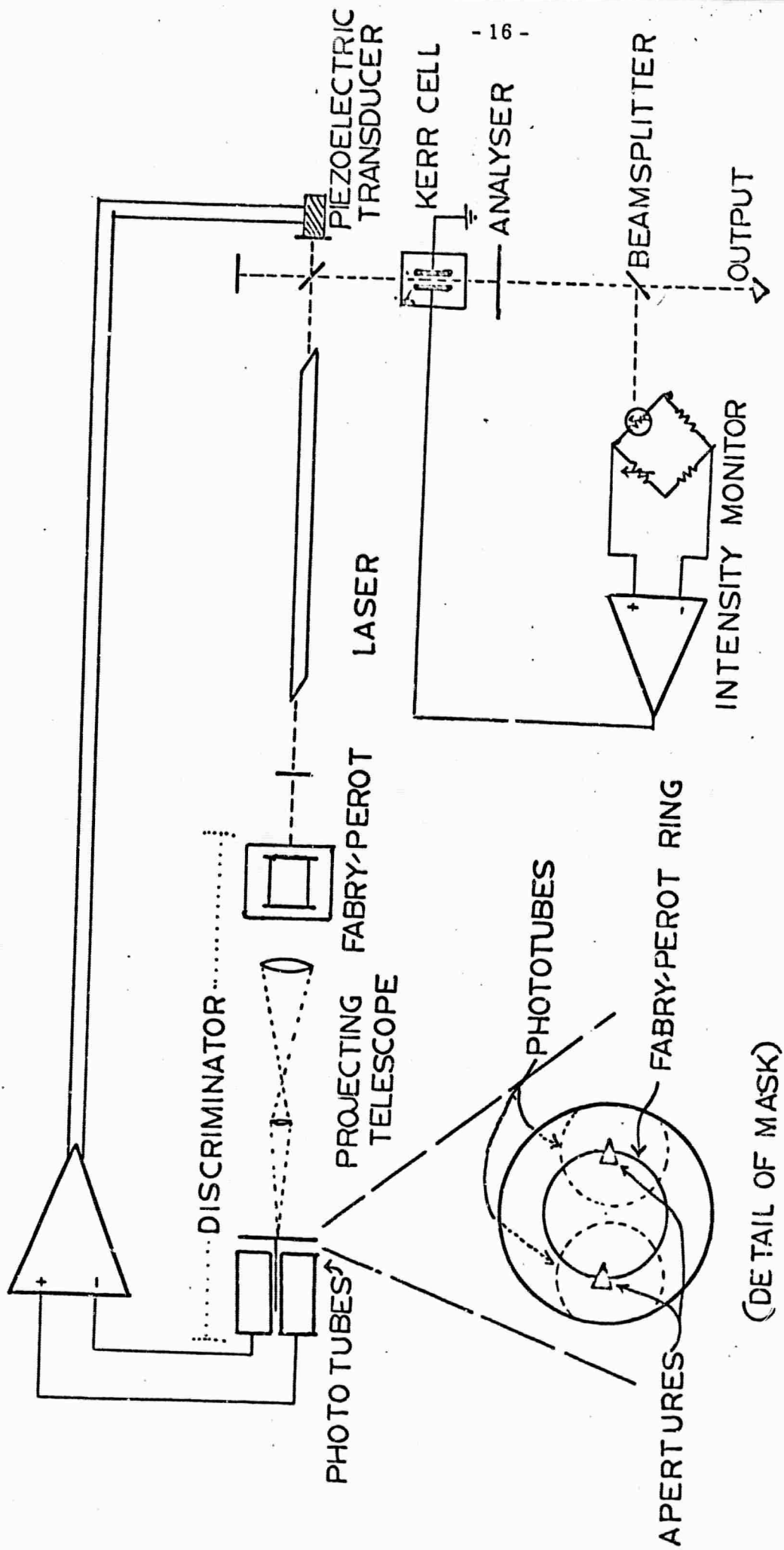


FIG. 6





FREQUENCY AND AMPLITUDE STABILIZATION

FIG.7

NARROWING OF THE DOPPLER LINEWIDTH INDUCED  
BY LASER RADIATION: APPLICATION TO PRECISE  
ISOTOPE SHIFT MEASUREMENTS

R. H. Cordover, P. A. Bonczyk, and A. Javan

Let us consider the spontaneous emission from an upper or a lower level of a Doppler-broadened gas laser transition. If such a spontaneous emission signal is viewed along the axis of laser propagation, its lineshape is changed considerably when the laser is allowed to oscillate. This change occurs over a narrow frequency interval which may be considerably less than the Doppler width. This report gives details of the observation of this effect and its application for the first time to ultra-precise measurements of isotope shifts for two optical transitions in Neon. The splittings due to the isotope effect are completely resolved. The measured linewidths are also analyzed.

In this experiment, a short Brewster-angle He-Ne gas laser was made to oscillate on a single mode of the  $1.15\mu$  transition ( $2p_4 \leftarrow 2s_2$ ). The spontaneous emission at  $6096\overset{\circ}{\text{A}}$  ( $1s_4 \leftarrow 2p_4$ ), which originates from the lower laser level, was observed through an end mirror. (In the following, we refer to the latter transition as the  $0.6\mu$  transition). The reflectance of the laser mirrors was designed to be transparent at  $0.6\mu$ . The signals were observed with a high resolution, pneumatically tuned Fabry-Perot interferometer and a photomultiplier with a pinhole for mode selection. The laser was switched on and off by a square-wave voltage applied to a transducer supporting one of the laser mirrors. Modulation of the transducer at an audio-frequency allowed narrow-band phase-sensitive detection of the signal. Fig. 1 gives a block diagram of the experimental arrangement.

Before discussing in detail the isotope shift measurements, let us review the observations for a single isotope. Suppose the laser is adjusted to oscillate at a frequency close to  $\omega_0$ , the frequency of the center of the Doppler-broadened  $1.15\mu$  transition. In this case, the linewidth of the  $0.6\mu$  transition, analyzed in the forward direction, consists of a narrow, nearly Lorentzian response superimposed on a broad, Gaussian, Doppler profile. The center frequency of the Lorentzian response coincides with the center frequency of the Gaussian profile. As the laser frequency is detuned, the Lorentzian response splits symmetrically about its center frequency (see Fig. 2a).

We now give a brief explanation of the origin of the above effect. The nearly Lorentzian signals arise from changes in the populations of the laser levels induced by the laser field. For a Doppler-broadened transition, these changes occur for atoms whose velocities  $v_z$  lie within a narrow range (where  $z$  is the direction of propagation of the laser radiation). This leads to a departure of the distribution of atoms with velocity component  $v_z$  from the usual Maxwellian velocity distribution for the population of each laser level. If the laser oscillates close to the peak of its Doppler response, the departure from the Maxwellian distribution occurs around  $v_z \approx 0$ . To first order in the laser field intensity, the functional dependence of this departure on  $v_z$  is a Lorentzian with half-width at half-maximum-intensity given by  $\Delta v = c \gamma / \omega_0$ , where  $c$  is the speed of light and  $\gamma = (1/2)(1/T_1 + 1/T_2)$ , with  $T_1$  and  $T_2$  being the radiative lifetimes of the two laser levels. This change in the Maxwellian distribution

leads to the observed behavior of the spontaneous emission signal viewed in the z-direction. The lineshape resulting from the change in population due to the laser field is a linear superposition of two Lorentzians both centered at  $\omega'_0$ , the center frequency of the  $0.6\mu$  transition. Their corresponding half-widths at half-maximum-intensity (in angular frequency units) are given by  $\gamma_{\pm} = \gamma' \pm \lambda \gamma$ , where  $\lambda = \omega'_0 / \omega_0$  and  $\gamma'$  is similar to  $\gamma$ , except referred to the two levels of the  $0.6\mu$  transition. The intensities of the two Lorentzians are proportional to their respective half-widths (at half-maximum-intensity). This result applies only when the usual Doppler width is considerably larger than  $\gamma_{\pm}$ . The lineshape is given by:

$$I(\omega') = \frac{\gamma_+}{(\omega' - \omega'_0)^2 + \gamma_+^2} + \frac{\gamma_-}{(\omega' - \omega'_0)^2 + \gamma_-^2}$$

Suppose now the laser frequency  $\omega$  is detuned from  $\omega_0$ . In this case changes in population occur within two narrow ranges of velocity centered about  $v_z^0 = \pm c(\omega - \omega_0) / \omega_0$ . The (+) and (-) signs arise from the standing wave nature of the laser field. This leads to the symmetrical splitting of the signals observed for the  $0.6\mu$  spontaneous emission. It may be seen that, due to the Doppler effect, the splitting at  $0.6\mu$  is given by  $2\omega'_0 \left| v_z^0 \right| / c = 2 \left| (\omega - \omega_0) \right| \omega'_0 / \omega_0$ , where  $\omega'_0$  is the center frequency of the spontaneous emission.

Let us now consider the effect described above as applied to the determination of isotope shifts. The measurements were done using a

He-Ne laser tube containing a (3:2)  $\text{Ne}^{22}$  -  $\text{Ne}^{20}$  mixture. The two isotopic components overlap for the  $1.15\mu$  transition as well as for the  $0.6\mu$  transition. The upper tracing of Fig. 2b shows the usual Doppler profile for the spontaneous emission at  $0.6\mu$ . Note that the isotope shift in this case is barely resolvable. The laser induced signals were observed with the  $1.15\mu$  oscillation set to a frequency close to the center of the Doppler response of the  $\text{Ne}^{20}$  isotope. Accordingly, the  $\text{Ne}^{20}$  transition at  $0.6\mu$  shows a single narrow response. However, because of the isotope shift at  $1.15\mu$ , the response due to  $\text{Ne}^{22}$  at  $0.6\mu$  is split. Furthermore, the isotope shift for the  $0.6\mu$  transition results in a shift of the center between the two peaks of the  $\text{Ne}^{22}$  signal away from the center of the signal due to  $\text{Ne}^{20}$ . This is seen in the lower tracing of Fig. 2b. Notice that in this case the isotopic splittings are completely resolved. Furthermore, this tracing contains information on the isotope shift for the  $1.15\mu$  transition as well as for the  $0.6\mu$  transition. The measured isotope shift at  $0.6\mu$  is  $(1706 \pm 30)$  MHz and the shift at  $1.15\mu$  is  $(257 \pm 8)$  MHz.  $\text{Ne}^{20}$  lies on the low frequency side for both transitions. The result of the  $1.15\mu$  measurement is in complete agreement with an earlier precise experiment done in our laboratory which involved the heterodyning of two lasers. It should be noted, however, that since the  $0.6\mu$  transition is not a laser oscillation, heterodyning techniques may not be applicable to this transition.

We have also studied the lineshape of the observed signals as a function of pressure. This lineshape, extrapolated to zero pressure, has

been analyzed approximately in terms of two Lorentzian responses as discussed above. Using the  $2s_2$  and  $2p_4$  radiative lifetimes determined from earlier "Lamb dip" measurements, we obtain a radiative lifetime of  $1.6 \times 10^{-8}$  sec for decay of the  $1s_4$  level to the ground state. In view of cumulative errors in this measurement the value given for this lifetime is accurate to within a factor of 2.

We would also like to point out that the above technique is applicable, in principle, to measurement of details of the structure of any transition which involves states populated directly or indirectly via radiative cascade from the laser levels. Furthermore, this technique may be used to precisely define the centers of Doppler-broadened atomic transitions and thus may be applied to problems related to achieving standards of length.

### FIGURE CAPTIONS

Figure 1. Block diagram of the experimental arrangement.

Figure 2. Spontaneous emission signals from lower laser level observed along laser axis. (a) Data for pure Neon isotope. The upper trace is the output of the photomultiplier and represents the normal Doppler-broadened spontaneous emission. The center trace is the output of the phase-sensitive detector when the laser frequency is set to the center of its Doppler profile. In the lower trace the laser frequency was set away from the center of its Doppler profile, and the signal splits into two components corresponding to both traveling waves. (b) Data for 3:2 mixture of  $\text{Ne}^{22}$  -  $\text{Ne}^{20}$ . The laser frequency was set to the center of the  $\text{Ne}^{20}$  Doppler profile. Note the barely resolved structure in the normal spontaneous emission signal in the top tracing.

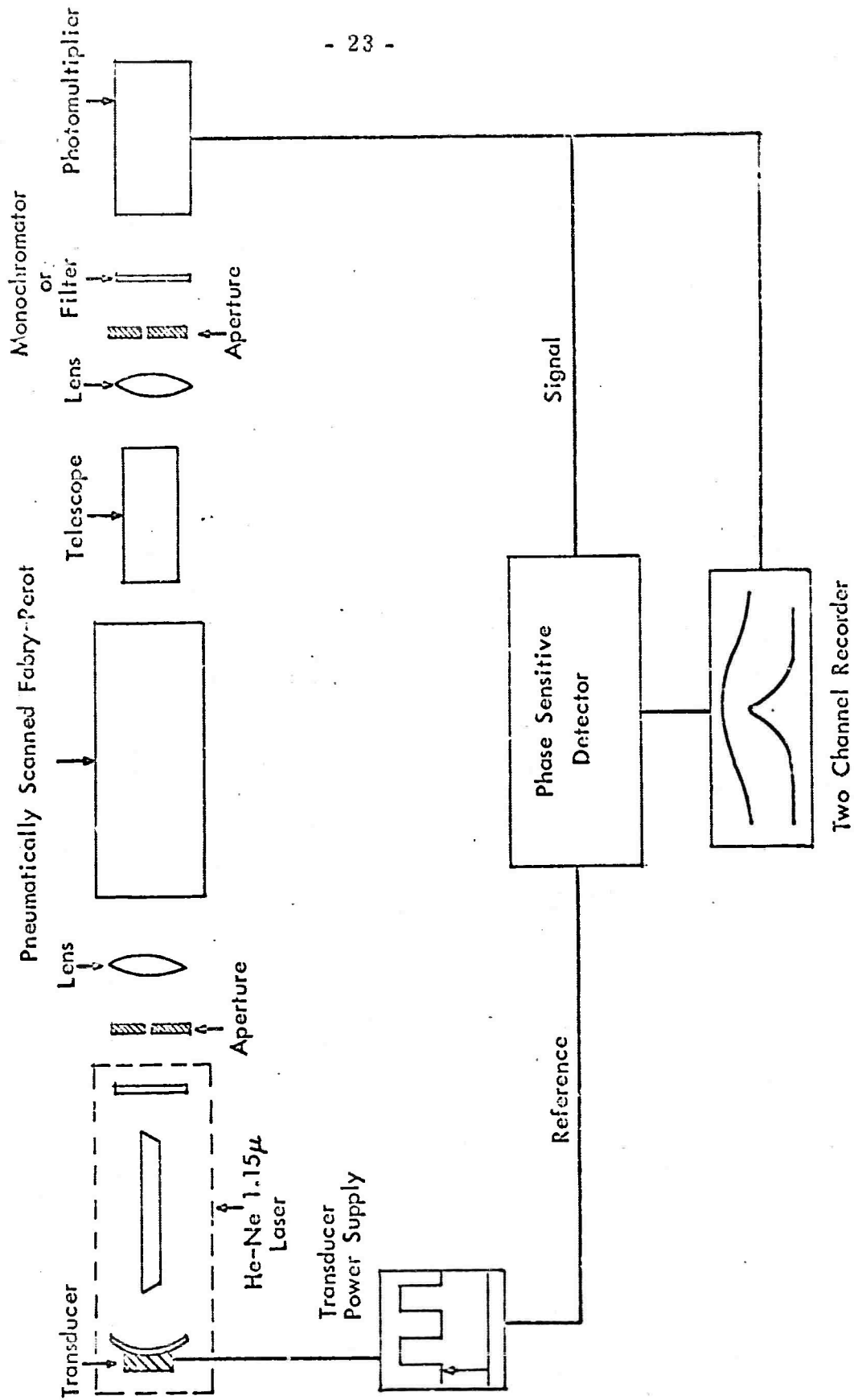
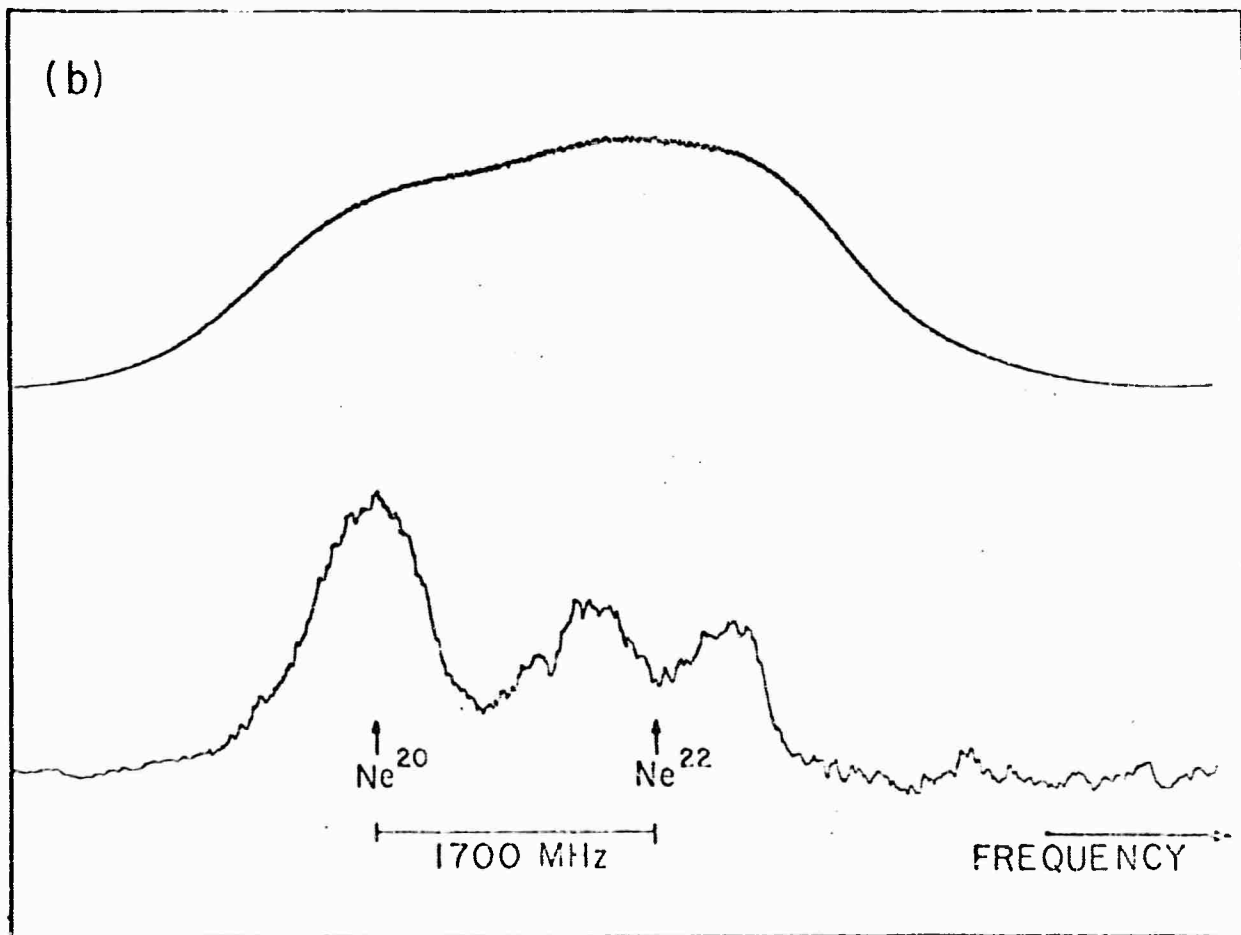
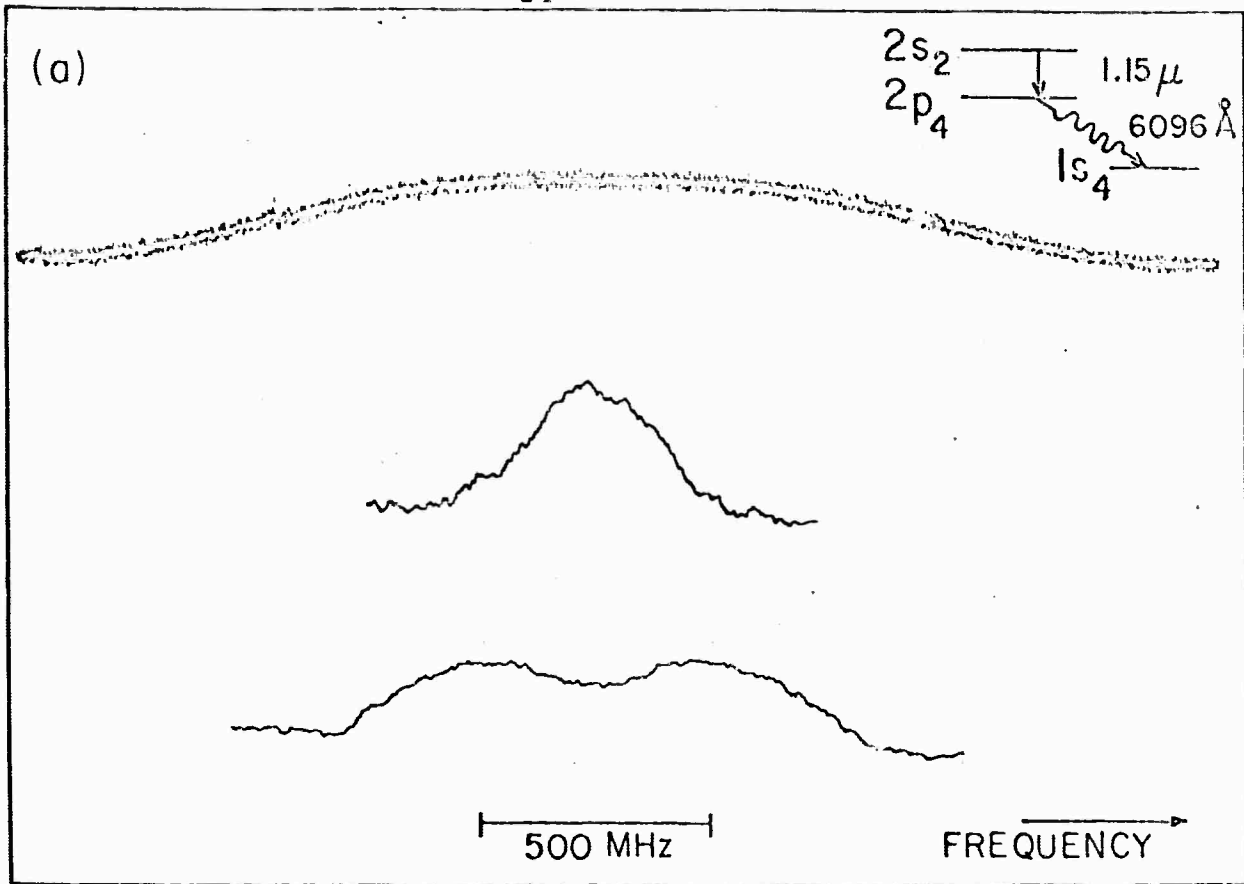


FIGURE 1





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**13. ABSTRACT:** Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as (TS), (S), (C), or (U).

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

**14. KEY WORDS:** Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.