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Authors
Winocur, Joseph
Pyle, Robert V.

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A SPECTROSCOPIC TECHNIQUE FOR PROBING AN IONIZED GAS

Joseph Winocur and Robert V. Pyle

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A Spectroscopic Technique for Probing an Ionized Gas*

Joseph Winocur

Department of Physics
University of California
Berkeley, California

and

Robert V. Pyle

Lawrence Radiation Laboratory
University of California
Berkeley, California

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ABSTRACT

Ionized strontium is studied by the method of resonant absorption of optical radiation. Because of an accidental frequency overlap, normal rubidium can be used in a lamp as the source of resonant light. Spectral profiles of absorption were made with a scanning Fabry-Perot interferometer. The $5s_{1/2}$-$6p_{1/2}$ line of RbI and the $5s_{1/2}$-$5p_{1/2}$ line of SrII are found to differ by 0.014 Å. Optical and Langmuir probe measurements of density compare favorably. Ion temperatures in a steady state and a decaying plasma were determined from the Doppler width. Two different sources of ionized strontium plasma are described. Negative results were obtained in an attempt to optically pump strontium ions in a plasma.

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I. INTRODUCTION

The measurement of neutral gas densities by the absorption of optical resonance radiation is a well-known technique.[1] With the development of scanning Fabry-Perot interferometers and electronic detectors this method has become useful for highly accurate density determinations.[2] We have applied the technique to measurements of density and temperature of an ionized gas.

Some advantages of this method over the conventional Langmuir probe and microwave techniques are that the plasma is not disturbed by the light probe, good spatial resolution is possible, effects of the magnetic field on the probe are well understood, only one level of a particular ion species is measured, ion temperatures can be measured, and density measurements accurate to ±10% are feasible. The method is limited to those elements that have a transition to the ground state at an experimentally convenient frequency, and an oscillator strength large enough to give large absorption at the desired gas density. Singly ionized alkaline earths have the same electronic configuration as the alkali metals, and most of them have a strong ns-np excitation frequency lying in the visible portion of the electromagnetic spectrum. Ionized strontium was chosen for this study because of an accidental coincidence of its first principal line with the second principal line of normal rubidium. This means that light from an easily constructed rubidium lamp can be used to probe the plasma. In general, light from a suitable arc discharge would be used. The $5s_{1/2}$-$5p_{1/2}$ transition in SrII and the $5s_{1/2}$-$6p_{1/2}$ transition in RbI are reported to have the same wavelength, 4215.524 Å.[3]

Additional information about the properties of the ions and their interactions with the plasma could be obtained by optically pumping the ions. This is considerably more difficult than pumping neutral rubidium because of the short relaxation times in the plasma and weaker lines of the available light sources. Our attempt to optically pump Sr^+ ions was unsuccessful, and is mentioned only briefly.

II. METHOD

When resonance radiation of initial intensity $I_0(\nu)$ passes a distance x through an absorbing gas of density N(x), the intensity becomes

$$I(\nu, x) = I_0(\nu, x_0)\exp[-\int_0^x k(\nu, x)dx]. \tag{1}$$

This equation defines the absorption coefficient $k(\nu,x)$, which is related to the gas density by means of the equation

$$\int k(\nu,x)\,d\nu = \pi r_0 cfN(x). \tag{2}$$

Here r_0 is the classical electron radius, c is the velocity of light, and f is the oscillator strength of the induced absorption. The absorption coefficient is determined experimentally by measuring optical absorption vs frequency, using a high-resolution Fabry-Perot interferometer. The quantity $\int k(\nu, x)d\nu$ is calculated by numerical integration. The density is computed from this and the known value of f. Kostkowski and Bass[4] have shown that the measured value for the integral $\int k(\nu)d\nu$ is not very sensitive to instrument width.[5] The oscillator strength of the $5s_{1/2}$-$5p_{1/2}$ transition in Sr^+ has been determined by the simultaneous measurement of total absorption and dispersion by Ostrovski et al. to be f = 0.39.[6] Because their method does not require knowledge of the concentration of absorbing atoms, a probable error of about 5 % is indicated in their measurement.

The principal source of ionized strontium line width in our plasmas is the Doppler effect, and the absorption coefficient may be written in the Gaussian form,

$$k(\nu,x) = k_0(x) \exp \{-4 \ln 2 [(\nu-\nu_0)/\Delta \nu_0]^2\}, \qquad (3)$$

where $\Delta\nu_0 = 2\nu_0 (2RT \ln 2/Mc^2)^{1/2}$ is the line width at half intensity. Here R is the universal gas constant, T is the temperature, and M is the molecular weight. Neutral and ionized gas densities in these experiments are too low to cause significant collision broadening or Stark broadening.

III. APPARATUS

Two strontium plasma devices were developed and studied. The first is a discharge using a heated tungsten filament as the cathode. Argon or helium gas is circulated through the tube to sustain the discharge and to control the diffusion rate to the walls. Metallic strontium is evaporated into the column of the discharge from an electrically isolated heated pan lying just above the anode (Fig. 1). The observation windows are mounted 10 cm from the discharge region to prevent their being coated by condensed strontium vapor. Typical operating conditions are in the range of neutral gas pressures from 0.05 to 10 torr, discharge currents between 0.1 and 1.0 Å, and anode potentials of 50 to 100 volts. A Langmuir probe can be introduced perpendicular to the axis into the same region that is scanned by the optical beam. The current can be operated dc or pulsed; in the latter case observations can be made in a decaying plasma. The Helmholtz coils provided the small magnetic field used in the optical pumping attempt.

The second plasma device, shown in Fig. 2, permits investigation of the behavior of a strontium plasma in a well-defined geometry with a strong axial magnetic field and low neutral-particle pressure ($\leqslant 10^{-3}$ torr). A 3-mm-diameter discharge, 10 to 40 Å, from a hollow cathode (Fig. 2) is confined to the axis of the cylinder by the magnetic field. Strontium can be introduced through the hollow cathode or can be evaporated into the discharge from a heated pan about 1 cm below the axis of the arc. The plasma can be run dc or it can be interrupted periodically either by electronic means or by mechanically intercepting the beam from the hollow cathode. The latter method is preferable at low repetition frequencies because the temperature of the hollow cathode source would drop appreciably if the arc were turned off for periods of the order of 1 second.

For the transmission attenuation measurements a narrow beam of electronically chopped (typically 70 cps) rubidium 4215 Å light, with a cross-sectional area determined by two apertures, is transmitted through the plasma, diffracted by the scanning Fabry-Perot interferometer, and focused onto the entrance slit of a monochromator. The output from the 1P21 photomultiplier tube of the monochromator is fed to a lock-in amplifier that is phase-locked to the lamp at the modulation frequency. The rectified output of the amplifier is recorded on a chart recorder. The lock-in amplifier is essential because it averages to zero the signal from random emission of Sr^+ 4215 Å radiation at the modulation frequency. The light at this wavelength that is emitted from the plasma may be an order of magnitude more intense than that from the rubidium source.

Most of the present measurements were obtained with a mechanically driven scanning Fabry-Perot interferometer with which the interference rings were swept across the monochromator slit. Better sensitivity and resolution were obtained from a central-spot-type scanning interferometer into which gas from a high-pressure source was introduced at a constant rate through an adjustable needle valve. The plates are polished flat to within $\lambda/50$, and the dielectric coating has a reflection coefficient of 0.87 at 4215 Å. All absorption measurements were performed with a 1-cm spacer, for which the mean spectral range is 500 mK and the instrumental width is about 40 mK.

Both systems were found to be quite linear; however, in the mechanical scanning method the fringe radii are proportional to $n^{1/2}$, where n is an integer.

IV. OBSERVATIONS

The Hot Cathode Discharge

A typical transmission measurement is shown in Fig. 3a. For ease in reducing data, and to improve accuracy, we scanned both the full-intensity transmitted beam and the partly absorbed beam within the same fringe by cyclically turning the discharge off and on at the rate of about one-half cps or less. The fringes were scanned at the rate of one every 4 or 5 minutes. The upper envelope in the figure is the transmitted rubidium radiation at 4215 Å with the mass-85 and -87 hyperfine components identified by arrows. The lower envelope is the line shape after resonance absorption by the plasma has occurred. Figure 3b is a plot of the absorption coefficient reduced from the curves of Fig. 3a. We see that the peak absorption by Sr^+ ions is displaced by about 80 mK (0.014 Å) from the weighted mean frequency of rubidium.

Density Measurements

The optically measured strontium ion density increases with neutral strontium vapor pressure reaching a maximum (for a given discharge current) that is unaffected by a further increase in neutral density. Because the ionization potential of strontium (6 eV) is considerably less than that of argon or helium, the discharge at saturation essentially runs on strontium.

In order to obtain the absolute density at any point in the discharge, it is necessary to know the radial variation of density. In the column of the hot cathode discharge this is presumably given by the Bessel function distribution $J_0(2.4\ r/R)$, which we verified with the movable Langmuir probe. To obtain the radial distribution by the optical absorption technique the interferometer was tuned to the frequency of maximum absorption and the absorption at this fixed frequency was measured as a function of radius. Assuming azimuthal symmetry the results were unfolded numerically, and the results fitted a Bessel function distribution quite well, with R = 3.75 cm, the effective radius of the discharge chamber in the region of observation. The absolute density at any radius could then be inferred. These results were compared with Langmuir probe measurements using the formula

$$N_+ = J_+(M/2kT_e)^{1/2}/0.4e, \qquad (4)$$

where J_+ is the saturated ion current density, M is the ion mass, k is Boltzmann's constant, T_e is the measured electron temperature, and e is the electronic charge. Optical and probe measurements of the ion density at the axis of the discharge tube are compared in Table I for two differing argon pressures and for zero and saturated strontium ion densities. The appropriate ion masses are used in formula (4) for the two cases. The optical and probe results are in somewhat better agreement than might be expected considering the uncertainties in the probe determination.

The rate of decay of ground-state strontium ions after the discharge is pulsed off can be determined by monitoring the transmitted light intensity, using a technique similar to that employed by Phelps and Pack for measuring the decay of metastable helium atoms.[7] A gated photomultiplier with variable gate width and a variable delay after the discharge is turned off samples the current over many discharge cycles. The Fabry-Perot is tuned to the peak absorption frequency, the lamp is operated CW, and

Table I. Typical axial density determinations in the positive column by optical absorption and Langmuir probe measurements. I=0.4A, $T_e \approx 1.5$ eV.

$N_{Sr^{\neq}}$ (absorption) ($10^{10}/cm^3$)	N_+ (Langmuir probe) ($10^{10}/cm^3$)	p(argon) (torr)
0	5.7 (Ar^+)	0.05
7.4	8.8 (Sr^+)	0.05
0	8.6 (Ar^+)	0.45
9.5	7.7 (Sr^+)	0.45

the amplifier is synchronized to the gater. Our measurements were limited by noise to two orders of magnitude loss in ion density, from about 10^{11} to 10^9 ions/cm^3. At low neutral gas pressures the decay rate should be determined by diffusion to the walls and at high neutral gas pressures and large ion densities the loss rate should be set by recombination processes. Our results are reasonably consistent with estimates of these two rates, but as no effort was made to determine the purity of the gas mixture it is not possible to obtain quantitatively useful numbers from the data.

Temperature Measurements

Ion temperatures were estimated from the Doppler-broadened absorption line widths in both steady-state and decaying plasmas. The correction for instrument width is taken from the calculations of Minkowski and Brück,[8] who assumed a Doppler line and an Airy instrument function. The observed strontium 4215 Å absorption line is 85 mK in the steady-state plasma, corresponding to a temperature of about 1100° K after correction for instrumental function, and is about 10 mK smaller in the decaying plasmas, indicating a temperature of about 800° K. These measurements were made at a helium pressure of 1 torr and a discharge current of 0.8 A. As far as we know they are the only direct measurements of ion temperature in the column of a low-current discharge, and are in agreement with the usual assumptions that are made about such discharges.

Optical Pumping Attempt

Attempts were made, by conventional means, to observe optical pumping[9] in the decaying strontium plasma of the pulsed positive column. Circularly polarized resonance radiation from a Rb electrodeless discharge source was transmitted through the positive column in the direction of the constant magnetic field and perpendicular to an applied rf field. The discharge was pulsed on for 0.1 msec and off for about 1 msec. The photodetector was gated synchronously to observe transmitted light only during the off time. Modulation of the rf field coupled with phase-sensitive detection was used to enhance signal to noise. No resonance-induced optical absorption was detected, under conditions such that the ratio of lamp intensity to noise was about 5×10^5. Optical pumping of neutral rubidium vapor was easily observed in the same apparatus.

Hollow-Cathode Arc Discharge

Experiments with the plasma diffusing across a magnetic field from the hollow-cathode source were limited mainly to temperature studies, and some radial measurements of density. A knowledge of the ion temperature is important in comparing the axial and radial density distributions with classical diffusion models.[10] In the magnetic field the $5s_{1/2} - 5p_{1/2}$ strontium transition is split into four components: $\nu_0 \pm 62$ mK/kG and $\nu_0 \pm 31$ mK/kG. At 500 G, where nearly all of the measurements were made, these appear as a single broadened line. The temperature was determined by comparing the observed absorption profile with the resultant of the four Doppler-broadened, equally weighted Zeeman components. Typical measurements are summarized in Table II; the ion temperature is seen to be a fraction of a volt, increasing with discharge current and decreasing neutral gas pressure. It was not possible to make reliable temperature measurements at larger magnetic fields because the magnetic field broadening dominates at these temperatures. However, in the core of the discharge, where the ion temperature is higher, it is in principle possible to go to considerably higher fields. In practice this could not be done because the intense radiation of the discharge core produced too much noise in comparison with the signal from the rubidium lamp. (Self-broadened emission lines show that the ion temperature is less than about 1 eV at the axis.)

V. CONCLUSIONS

The resonance absorption of light has been applied to the measurements of the density and temperature of a specific ion species in two laboratory discharges.[11] Further measurements of the disappearance rate in decaying plasmas under controlled purity conditions appear to be worth while. The pumping time in the optical pumping attempt was predicted to be about 0.3 sec, whereas the relaxation time due to ion-electron spin-exchange collisions was estimated to be of the order of 0.1 msec. With an optical absorption of about 3%, it was thought that an effect with as much as 5 to 1 signal to noise might be possible. We believe that an improvement in the light source might make the experiment feasible.

Table II. Strontium ion temperatures in the plasma diffusing from the hollow-cathode line source. B = 500 G.

Strontium ion temperature (eV)	Argon pressure (10^{-3} torr)	Arc current (A)
0.07 ± 0.07	2	12
0.2 ± 0.1	2	25
0.5 ± 0.2	2	40
0.5 ± 0.2	0.5	12
0.45 ± 0.2	0.5	25

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REFERENCES

1. Allan C. G. Mitchell and Mark W. Zemansky, Resonance Radiation and Excited Atoms (Macmillan Co., New York, 1934).
2. S. M. Jarrett, Spin Exchange Cross Section for Rb^{85}-Rb^{87} Collisions, Phys. Rev. 133, A111 (1964).
3. C. Moore, Atomic Energy Levels as Derived from Analyses of Atomic Spectra, Vol. II, NBS Circular No. 467 (US Govt. Printing Office, Washington D. C. 1948),
4. Henry J. Kostkowski and Arnold M. Bass, J. Opt. Soc. Am. 46, 1060 (1956).
5. For example, if the instrument function and line profile are Gaussian and in the ratio 2:1, the error in $\int k(\nu) d\nu$ will be only 2.5% when the measured line width is in error by 150% and the measured peak absorption coefficient is in error by 50%. In addition, these quantities are insensitive to the fraction of light absorbed.
6. Yu. I. Ostrovskii, and N. P. Penkin, Opt. Spectr. (U.S.S.R.) (English Transl.) 11, 307 (1961).
7. A. V. Phelps and J. L. Pack, Rev. Sci. Instr. 26, 45 (1945).
8. R. Minkowski and H. Brück, Z. Physik 95, 299 (1935).
9. A. Kastler, J. Opt. Soc. Am. 47, 460 (1957).
10. K. H. Berkner, R. V. Pyle, H. F. Rugge, J. W. Stearns, and J. Winocur, Diffusion From a Line Source in a Magnetic Field, in Proceedings of the VI International Conference on Ionization Phenomena in Gases, (S.E.R.M.A., Paris, 1964).
11. F. W. Hofmann, Phys. Fluids 7, 532 (1964) has recently reported the measurement of ion density in a barium plasma using resonance scattering of light from a Hg arc.

FIGURE CAPTIONS

Fig. 1. The positive column apparatus.

Fig. 2. The hollow cathode apparatus. The lamp and opposite mirror are simultaneously motor driven to permit scanning along different chords.

Fig. 3(a). Line shape of RbI 4215-Å light transmitted through the positive column apparatus. The upper and lower envelopes are respectively without and with absorption by strontium ions. p= 0.8 torr, I = 0.6 A.

Fig. 3(b). Curve of absorption coefficient vs wavelength, obtained from Fig. 3(a).

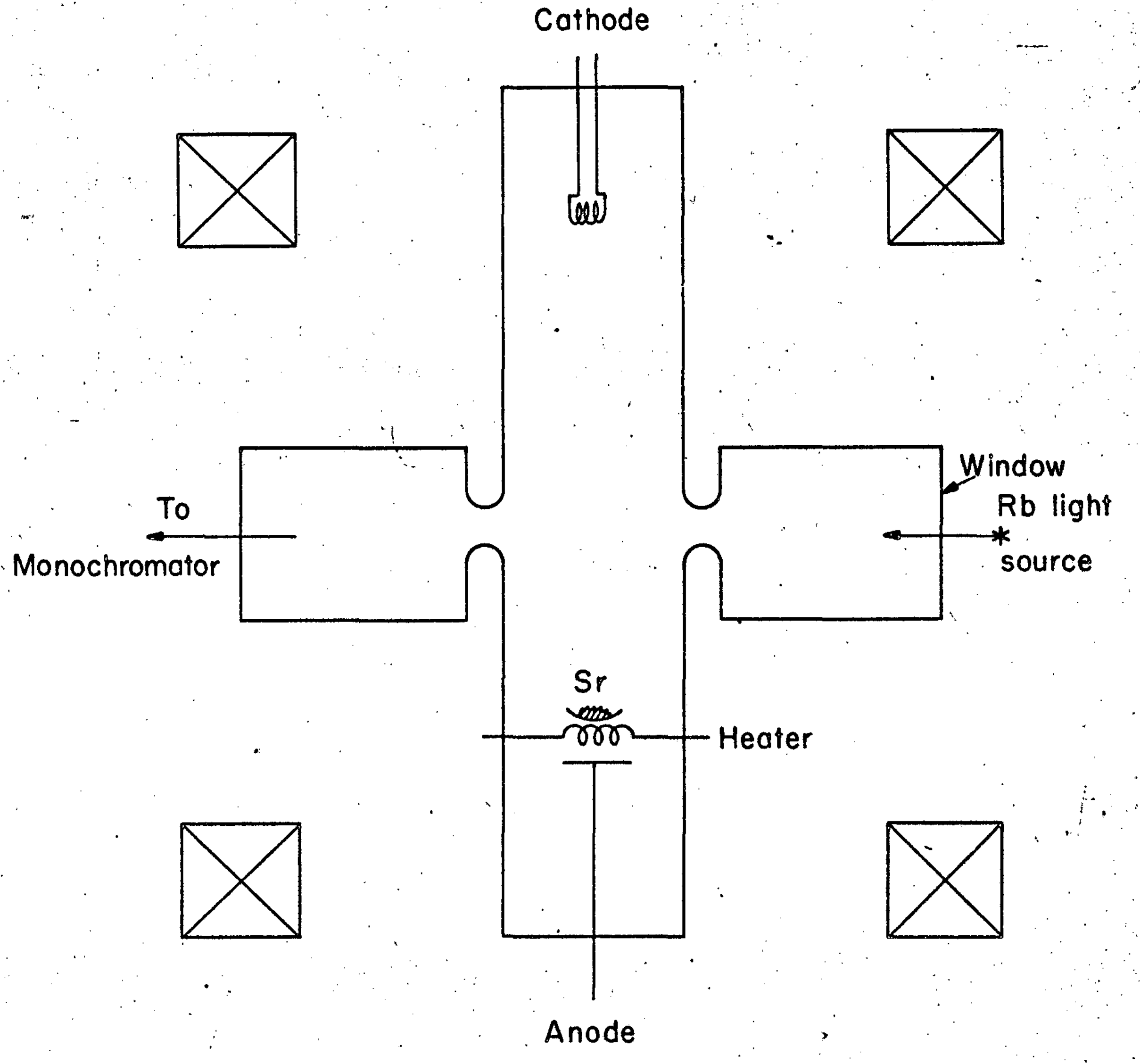
Cathode
Window
To
Monochromator
Rb light
source
Sr
Heater
Anode
MU-33628

Fig. 1

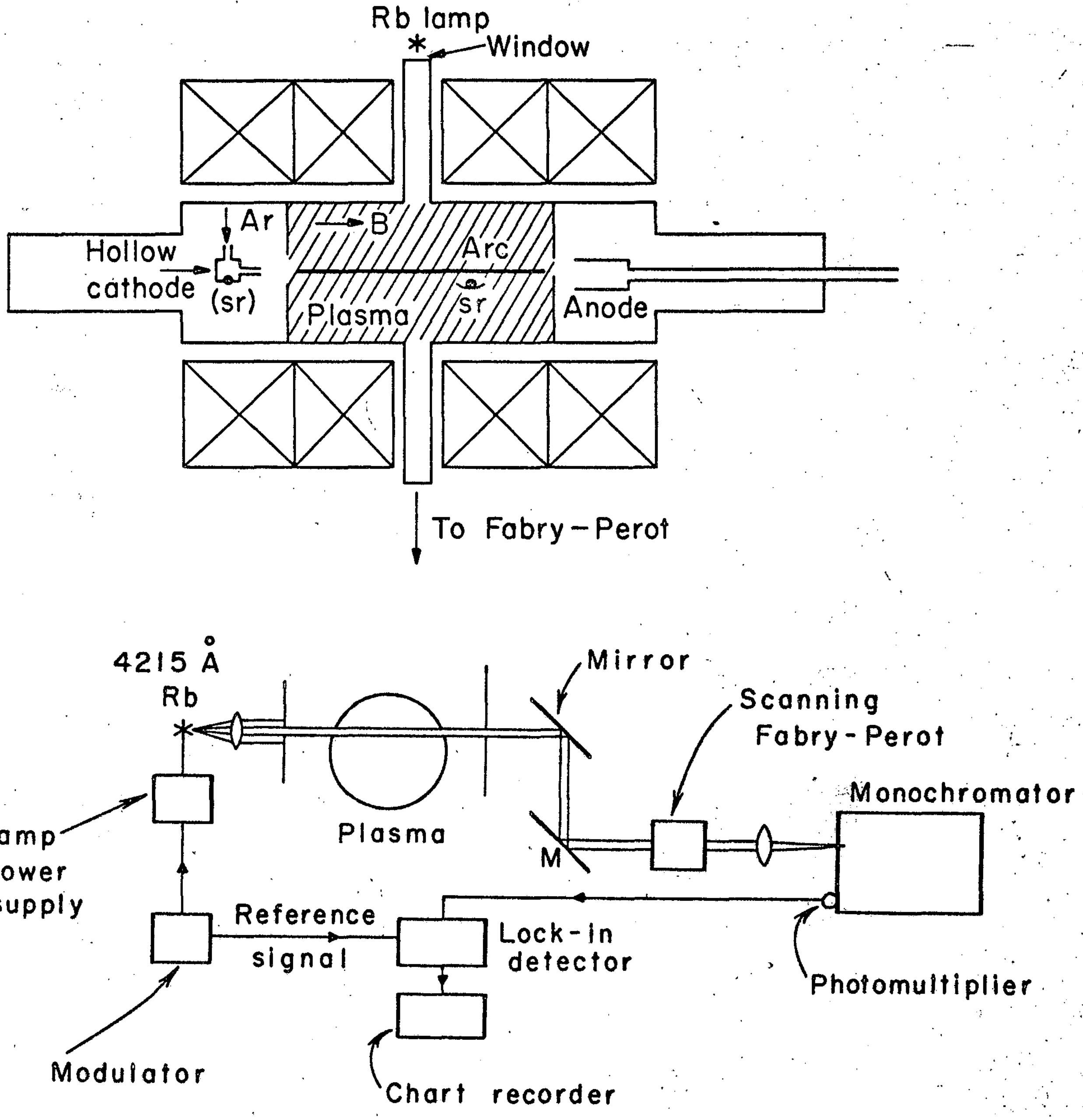

Fig. 2.

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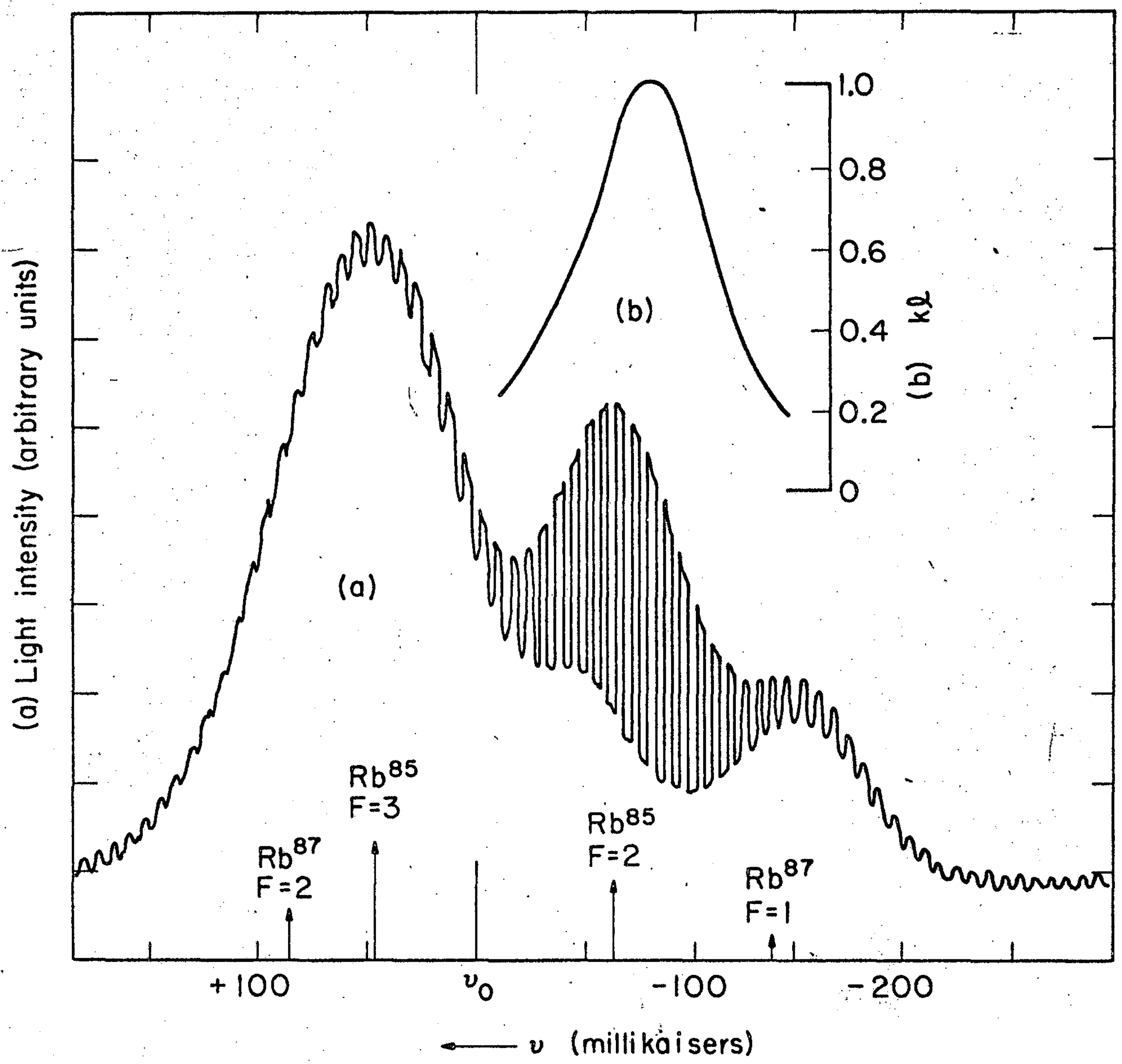

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Fig. 3