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BHABHA ATOMIC RESEARCH CENTRE
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60 Abstract : Deconvolution of atomic line profiles, recorded on PC interfaced recording Fabry-Perot spectrometer, into its Lorentzian and Gaussian component has been carried out. Effect of various parameters of hollow cathode discharge lamp (light source) such as discharge current, bath temperature and gas pressure on Lorentzian and Gaussian width has been studied. The value of the self-broadening coefficient for neon-neon atomic interaction for the transition $2p^5 4P - 2p^5 3s$ ($\lambda = 3472.571$ Å) has been determined.

70 Keywords/Descriptors : LINE BROADENING; LINE WIDTHS; SPECTRAL SHIFT; NEON; GAUSSIAN PROCESSES; DISTRIBUTION; DOPPLER COEFFICIENT; SPECTROMETERS; FABRY-PEROT INTERFEROMETER; ELECTRIC DISCHARGES; TEMPERATURE DEPENDENCE; EXPERIMENTAL DATA; DATA ACQUISITION SYSTEMS; MICROCOMPUTERS; CATHODES; PRESSURE DEPENDENCE; ATOM-ATOM COLLISIONS; GADOLINIUM; LIGHT SOURCES

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S. G. Nakhate, S. A. Ahmad, Pushpa M. Rao and G. D. Saksena

1. Introduction:

The study of atom-atom, atom-ion collisions is a highly interesting field in atomic physics. There are many theoretical model potentials formulated for these interactions, and experimental result can test the validity of these existing interaction model potentials. One of the techniques for this is the study of pressure broadening and shift of spectral lines which is a valuable source of information about interaction between the excited state and the ground state atoms. Many experimental investigations on broadening and shift of spectral lines have been reported in literature. Among these rare gases are of particular interest because of their extensive use as a buffer gas in discharge lamps and also because they present simpler test-system for theoretical modeling. The experimental investigations on spectral line broadening and shift of argon [1-3], neon [4-7] and helium [8-11] have been reported in literature. Recently we have initiated these types of studies in our laboratory. The existing Recording Fabry-Perot Spectrometer (REFPOS) designed and built in our laboratory [12] has been interfaced with a PC to acquire the data in digitized form for better data handling and computing. We report here the method of deconvolution adopted for separating Gaussian ($\Delta\nu_G$) and Lorentzian ($\Delta\nu_L$)component from observed line profile obtained using REFPOS. We have carried out a test of our deconvolution procedure by comparing the ratio of Gaussian width (i.e. full width at half maximum) of Neon and Gadolinium lines excited under identical conditions which according to theory is expected to be equal to their inverse mass ratio. We also report here the effect of outer environmental temperature (i.e. bath temperature), discharge current and discharge gas pressure on Lorentzian and Gaussian width. We have also determined the collisional self broadening coefficient for neon-neon atomic interaction for the transition $2p^5 4p - 2p^5 3s$ (λ = 3472.571 Å).

2. Experiment

a) The light source:

The light source consisted of a d.c. discharge hollow cathode lamp (HCL), cooled with liquid nitrogen, ice or water depending on the temperature requirement. The figure 1 shows schematically the hollow cathode discharge tube, details of which have been described elsewhere [12]. The HCL can be operated at discharge currents varying from 10 mA to 60 mA. The pressure of neon in the discharge was measured on line by means of calibrated oil manometer and was varied in the range 1-10 torr. The number density of the gas has been estimated from the gas pressure and the discharge temperature evaluated from the Gaussian widths of spectral lines.

b) The high-resolution system :

The system is described in full detail in reference [12], the schematic diagram of which is shown in figure 2. High resolution (10^6) was provided by a pressure-scanned Fabry-Perot etalon having plane surfaces ($\lambda/100$) coated with multilayer dielectric films (finesse, N_R = 30) for maximum reflectivity at λ = 3600 Å. The gas used for scanning was nitrogen, slowly leaked into the etalon chamber through a capillary which gave a linear scan of wavelength with respect to time. A two - metre grating spectrograph gave the predispersion. The intensity at the centre of the fringe pattern in a spectral line was monitored by photomultiplier tube (PMT). The PMT pulses were amplified further by an electrometer amplifier.

c) Data acquisition system :

The analog signal from electrometer amplifier was digitized using a digital multimeter (Keithley Model 197). The maximum sampling rate of this system is 3 samples/sec. This was coupled to PC/XT through GPIB bus. The interface software developed by Computer Division was prepared with following features:

- i) Remote initialisation of digital volt meter (DVM).
- ii) Transfer of DVM readings to PC disk files.
- iii) Graphic display of output profile with actual units.
- iv) Manipulation of graphic display of output profile; e.g. horizontal/vertical expansion and compression, cursor roll and time difference and average computations.

We found this digitized system is suitable for recording grating spectrum as well as Fabry-Perot fringe pattern.

3. Data Analysis

The analysis of line profile was carried out using essentially the procedure described by Ballik [13]. If the emitting atoms have a Maxwellian velocity distribution, the contribution of their motion to the line profile is a Gaussian distribution of width $\Delta\nu_G$. In the impact approximation which is valid at low number density, the natural and the pressure broadening have Lorentzian distributions, of widths $\Delta\nu_n$ and $\Delta\nu_P$ respectively. We recall further that the convolution of two Lorentzians (of width $\Delta\nu_{LX}$ and $\Delta\nu_{LY}$) is also a Lorentzian, with a width given by

$$\Delta\nu_{LXY} = \Delta\nu_{LX} + \Delta\nu_{LY} \qquad (1)$$

and the convolution of Lorentzian over Gaussian profile gives Voigt profile. Thus the intensity distribution of emitter at low density is a Voigt function.

The instrumental function of an ideal Fabry-Perot etalon is an Airy distribution, which can be written as a sum of an infinite number of displaced Lorentzian distributions spaced one spectral range (FSR) apart [14]; FSR = $(1/2t)$ cm^{-1},where t (in cm) is the separation between the etalons. Thus, each of the Lorentzians comprising the instrumental Airy distribution is convolved with the Lorentzian arising from natural as well as pressure broadening to produce a set of broader Lorentzians, still one FSR apart. This in turn is equivalent to an Airy distribution of increased width convolved with a Gaussian distribution.

For the ideal Fabry-Perot interferometer the numerically convenient expression for this convolution is given by Ballik [13] and it has the form:

$$I_T(\nu) = \frac{\left[\frac{1}{2} + \sum_{n=1}^{\infty} (Re^{-L})^n \; e^{-n^2 D^2/4} \; \cos 2\pi nz\right]}{\left[\frac{1}{2} + \sum_{m=1}^{\infty} (Re^{-L})^m \; e^{-mD^2/4}\right]} \qquad (2)$$

where $I_T(\nu)$ is the transmitted intensity of Fabry-Perot interferometer,

R is the reflectivity of Fabry-Perot etalons,

$$Z = \frac{\nu - \nu_o}{\Delta\nu}$$

$$L = \frac{\Pi \Delta\nu_L}{\Delta\nu} \qquad (3)$$

$$D = \frac{\pi \Delta\nu_G}{\Delta\nu \sqrt{\ln 2}} \qquad (4)$$

$\Delta\nu_L$ and $\Delta\nu_G$ are the Lorentzian and Gaussian half-widths, respectively, and $\Delta\nu$ is the FSR of the interferometer.

ν_O is the centre frequency of first order of interferogram and ν is the relative frequency from ν_O.

The line profile parameters $\Delta\nu_L$ and $\Delta\nu_G$ were determined by a least-square fit of the experimentally observed line profile data points to the equation (2) derived by Ballik [13].

4. Results and Discussions

We have done three types of studies on NeI line at 3472.571A excited in hollow cathode discharge lamp (HCL):

(a) Effect of hollow cathode discharge current on Doppler temperature of gas at constant bath temperature.

(b) Effect of HCL bath temparature on Doppler temperature of the gas at constant discharge current.

(c) Effect of Ne pressure on Lorentzian width at constant bath temperature and discharge current.

4.1 Test of deconvolution procedure:

The Gaussian width $\Delta\nu_G$ for a spectral line is given by the relation

$$\nu_G = 7.16 \times 10^{-7} \times \nu_o \sqrt{\frac{T}{M}} \qquad (5)$$

where T is temperature (in ^{o}K), M is mass (in amu) of the emitter and ν_o is the center frequency (in Hertz). As can be seen from equation (5) the Gaussian width is inversely proportional to square root of the mass of emitter at constant temperature.

We have recorded the neon line at 3472.571 Å and Gadolinium line at 3494.40 Å excited in same HCL. The cathode was coated with enriched isotope of Gd (A = 160, 96%). The evaluated Gaussian and Lorentzian widths for Ne and Gd after deconvoluting the line profiles along with the ratio of their mass is given in table 1. The theoretical fit of the experimental data points for both the lines is shown in figure 3 and 4. From table 1 we can see that the ratio of Gaussian width of Ne to Gd is nearly equal to the square root of ratio of inverse of their masses. This is in accordance to equation (5). This gives the validity of our deconvolution procedure.

4.2 Effect of discharge current on Doppler temperature of gas (Ne) at constant bath temperature

The Ne line at 3472.571 Å excited in hollow cathode discharge lamp (at Ne pressure of 2.5 torr) was recorded at various discharge currents varying from 10 mA to 60 mA. The bath was kept at liquid N_2 temperature through out the experiment. The line profile was deconvoluted into its respective Lorentzian and Gaussian component. Using equation (5) Doppler temperature of discharge was calculated from separated Gaussian width and is given in table 2 along with corresponding Lorentzian width for various discharge currents. The Doppler temperature (T_D) derived from Gaussian width $\Delta\nu_G$ was found to be more than the bath temperature (77°K) for all discharge currents including minimum at 10 mA. The difference between T_D and the bath temperature (T_B) is caused by the heating of gas due to the discharge current. Only at the extreme low discharge currents the gas temperature may approach the bath temperature. The plot of discharge current (I_D) versus calculated Doppler temperature (see figure 4) shows the linear increase of T_D with respect to I_D.

4.3 Effect of bath temperature on Doppler temperature of gas at constant discharge current

The above mentioned study was carried out on Ne line at 3472.571 Å to see the effectiveness of cooling on the Gaussian width and thus on Doppler temperature of gas. The hollow cathode lamp operated at discharge current 20 mA was cooled from outside with different coolants. The evaluated Gaussian half-width and the corresponding Doppler temperature along with Lorentzian half-width for different bath temperatures is given in table 3.

4.4 Effect of Ne pressure on Lorentzian and Gaussian width at constant discharge current and bath temperature

We have studied the Ne spectral line at 3472.571 Å arising from the $2p^5 4p$ - $2p^5 3s$ transition at discharge current 20 mA and constant bath temperature at 77°K. The values of the Lorentzian and the Gaussian width were evaluated as earlier and measured as a function of the Ne number density (or pressure). Figure 5(a) shows the Gaussian component of the line profile plotted against the Ne pressure. As can be seen, the Gaussian width of the line is practically independent of the perturbing gas pressure. The mean Doppler temperature (T_D) corresponding to these widths $\Delta\nu_D$ is found to be 273.50°K (corresponding to mean line width) which is on higher side of bath temperature (77°K) as mentioned earlier. In figure 5(b) the plots of Lorentzian widths $\Delta\nu_L$ against number density of Ne (N_{Ne})is shown. The (N_{Ne}) values were determined from the measured Ne gas pressure and the evaluated average Doppler temperature of the gas. The plot shows the linear dependence of $\Delta\nu_L$ on N_{Ne} which is according to the prediction of impact broadening theory. So that we can introduce the broadening coefficient according to equation.

$$\Delta\nu_L = \Delta\nu_L^{(0)} + \beta N \qquad (6)$$

where $\Delta\nu_L^{(0)}$ denote the "residual" Lorentzian width. In case of self-broadening (Ne^*- Ne) and for the ideal Fabry-Perot interferometer, $\Delta\nu_L^{(0)}$ should, in principle, correspond to the natural half-width of the line. However, for the real interferometer $\Delta\nu_L^{(0)}$ may also contain some instrumental contribution so that the observed value of $\Delta\nu_L^{(0)}$ usually differs from the true natural width of the line.

The other important information we can get from figure 5 is the collisional broadening coefficient which is found to be 6.30 X 10^{-20} cm^{-1}/atom/cm^3. This value was determined by the least-square fit to linear equation (6). Our β value (at Doppler temperature of 273.5 $^\circ$K) is close to the value determined by Bobkowski et al [6] (at Doppler temperature of 311 $^\circ$K) which is 2.47 × 10^{-20} cm^{-1}/atom/cm^3 for Ne line at 5341.1 A$^\circ$ arising from $2p^5 3p$ - $2p^5 4d$ transition. This difference in the β values can be accounted due to the difference in the Doppler temperatures and the cofigurations of the levels involved in the transitions studied.

The evaluated collisional broadening coefficient gives the information about the interaction between excited state and ground state atoms and helps in ascribing the interaction model potential between them. In the case of the Van der Waals' potential ($V(R) = -C_6 R^{-6}$) which is purely attractive type, the formula from the adiabatic impact theory for the broadening coefficient [15] is given by

$$\beta = 8.08\ (\Delta C_6/h)^{2/5}\ \bar{v}^{\,3/5} \qquad (7)$$

where $\Delta C_6 = C_6^u - C_6^l$ (u = upper, l = lower state index) is the difference of the Van der Waals' constant respectively for upper and lower state, $\bar{v}$ is the mean relative velocity = $(\frac{8KT}{\pi\mu})^{1/2}$, μ is reduced mass of the two atoms. The constant C_6 can be estimated using the approximate expression [15]

$$C_6 = \frac{1}{\hbar} e^2 \alpha \overline{r^2} \qquad (8)$$

Where $\overline{r^2}$ is the quantum mechanical average value of r for agiven state of the radiating atom, α is polarizability of the perturbing atom and e is the elementary charge. Thus a comparison of our experimental β values with the theoretical values which can be calculated using the above discussed equations will give an idea about the interaction potential existing between excited and ground state atoms.

We plan to undertake a systematic experimental as well as theoretical studies of broadening coefficient for the rare-gas systems.

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References

1. Wawrzyniski J and Wolnikowski J, Physica Scripta **33**, 113 (1986).
2. Wolnikowski J, Wawrzyniski J, Bielski A and Szudy J, Physica Scripta **35**, 153 (1987).
3. Wolnikowski J, Physica Scripta **42**, 220 (1990).
4. Bielski A, Dokurno W, Szudy J and Wolnikowski J, Physica **101C**, 113 (1980).
5. Bobkowski R, Bielski A, Lisicki E, Szudy J and Wolnikowski J, Acta Physica Polonica **A67**, 601 (1985).
6. Bobkowski R, Bielski A, Szudy J and Wolnikowski J, Acta Physica Polonica **A69**, 347(1986).
7. Bobkowski R, Bielski A, Lisicki E and Szudy J, Acta Physica Polonica **A72**, 709 (1987).
8. Kuhn H G and Vaughan J M, Proc. Roy. Soc. Lond. **A277**, 297 (1963).
9. Vaughan J M, Proc. R. Soc. Lond **A277**, 164 (1966).
10. Malvern A R, Pinder A C, Staley D N and Thompson R C, Proc. R. Soc. Lond **A371**, (1980).
11. Su J F and Nicol J L, J. Phys. B: At. Mol. Opt. Phys. **23**, 215 (1990).
12. Saksena G D and Ahmad S A, Bhabha Atomic Research Cetre (India), Report No. BARC **290** (1967).
13. Ballik E A, Applied Optics **5**, 170 (1966).
14. Bayer-Helms F, Z. angew. Phys. **15**, 330 (1963).
15. Sobelmann I I, Vainshtein L A and Yukov A, *Excitation of Atoms and Broadening of Spectral Lines*, Springer, Berlin (1989).

Table 1: Evaluated Gaussian ($\Delta\nu_G$) and Lorentzian ($\Delta\nu_L$) width for Ne (A = 20) and Gd (A = 160) line excited in hollow cathode lamp operated at current 40 mA and at bath temperature 77°K (Ne pressure = 2.5 Torr).
(1 mK = 10^{-3} cm^{-1} = 30 MHz).

Element	Wavelength	$\Delta\nu_D$	$\Delta\nu_L$	$\frac{(\Delta\nu_D)_{Ne}}{(\Delta\nu_D)_{Gd}}$	$\sqrt{\frac{M_{Gd}}{M_{Ne}}}$
	(Å)	(mK)			
Ne	3472.571	118.381	11.96	2.57	2.79
Gd	3493.40	46.033	29.183		

Table 2: Evaluated Gaussian ($\Delta\nu_G$) and Lorentzian ($\Delta\nu_L$) width for Ne line at 3472.571 A° excited in hollow cathode with different discharge currents (I_C). The corresponding Doppler Temperature (T_D) is also given (Ne pressure = 2.5 torr, Bath temperature = 77°K).

I_C	$\Delta\nu_G$	$\Delta\nu_L$	T_D
(mA)	(mK)		(°K)
10	77.185	25.079	282.83
20	87.568	21.262	364.05
28	94.837	21.098	426.99
34	102.797	19.636	501.68
42	110.758	17.711	582.39
44	111.45	16.601	589.69
47	111.45	18.030	589.69
60	124.257	8.634	733.0

Table 3: Evaluated Gaussian width ($\Delta\nu_G$) and corresponding Doppler temperature and Lorentzian ($\Delta\nu_L$) widths for Ne line at 3472.571 A excited in hollow cathode lamp (operated at constant discharge current of 20 mA) for different bath temperatures.

Bath Temp. T_D (°K)	$\Delta\nu_G$ (mk)	Doppler Temp. (°K)	$\Delta\nu_L$ (mk)
77 (Liq N_2)	85.5	347	19.3
270 (Ice+salt)	103.5	508.5	17.2
285 (Water)	108.7	560.8	16.6
300 (Room temp)	111.4	589.7	14.7

Table 4: Gaussian and Lorentzian width for Ne line at 3472.571 **A** excited in hollow cathode discharge lamp, operated at constant bath temperature (77 K°) and discharge current of 10 mA with different Ne pressure.

Ne pressure (Torr)	$\Delta\nu_L$ (mK)	$\Delta\nu_G$ (mK)	T_D (K°)	Ne number density* (atoms/cc)
3.0	22.74	75.63	271.56	1.061×10^{17}
4.0	27.63	78.22	290.48	1.415×10^{17}
5.0	28.50	77.53	285.37	1.768×10^{17}
6.0	30.61	71.64	243.66	2.12×10^{17}
7.0	31.30	76.84	280.32	2.48×10^{17}
8.0	35.52	78.31	291.15	2.83×10^{17}
9.0	37.42	73.26	254.81	3.18×10^{17}
10.0	47.40	75.80	272.78	3.54×10^{17}
		75.90± 2.39	273.50	

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*Ne number density has been calculated assuming average Doppler temperature as 273.5°K corresponding to average Gaussian width of 75.90 mK. (See Figure 6).

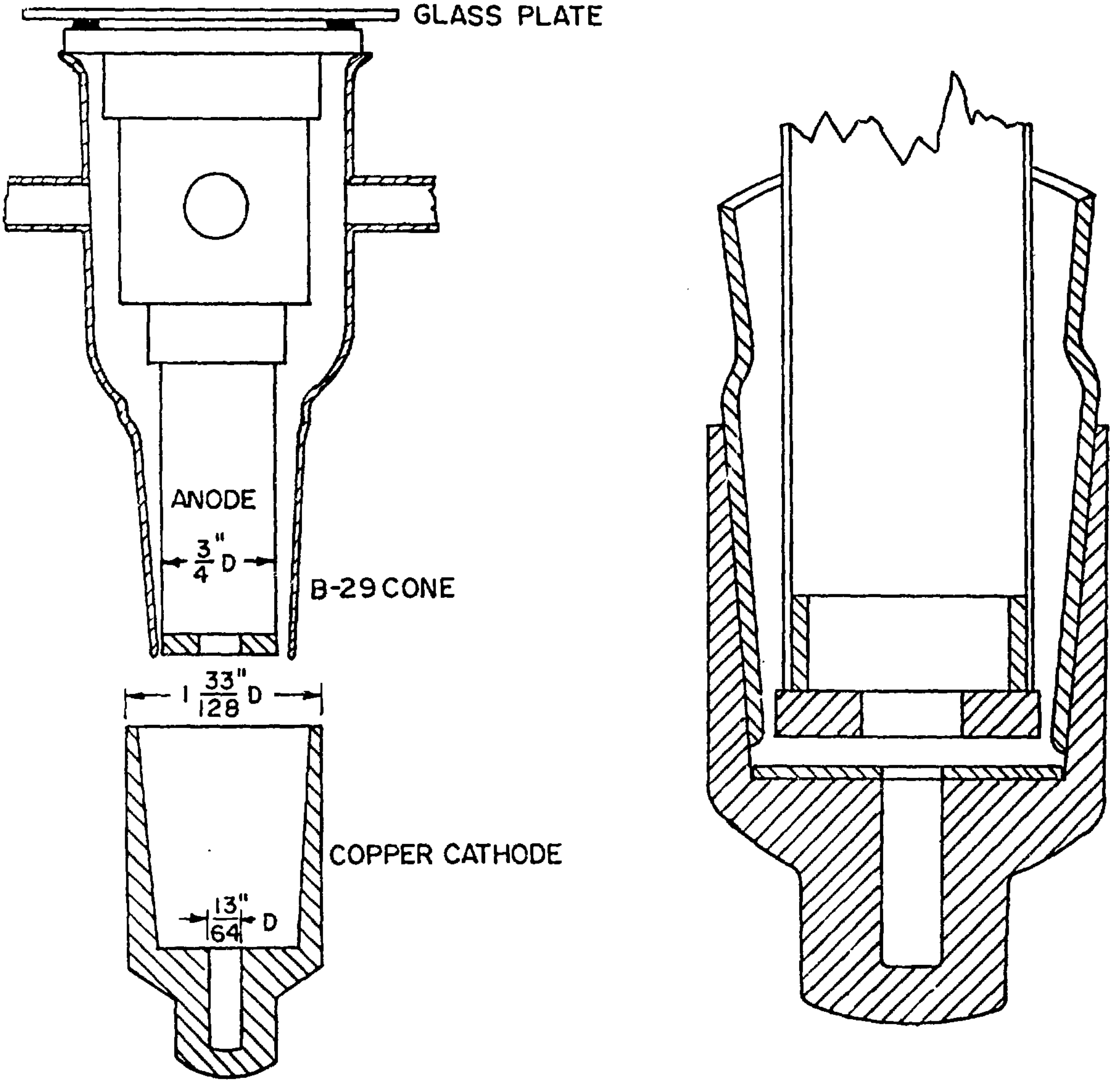

FIG. 1. DEMOUNTABLE HOLLOW CATHODE.

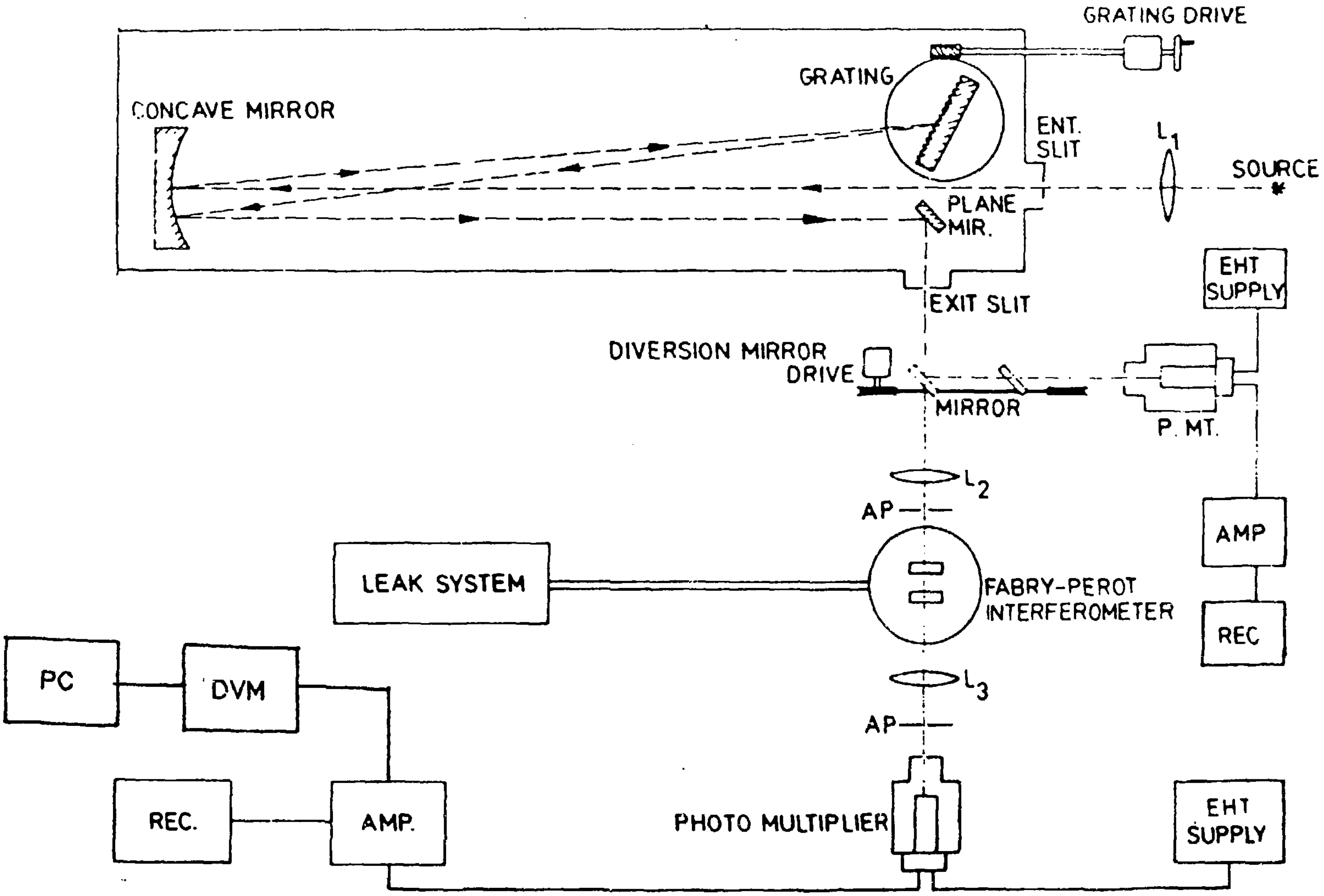

FIG 2. RECORDING FABRY-PEROT SPECTROMETER

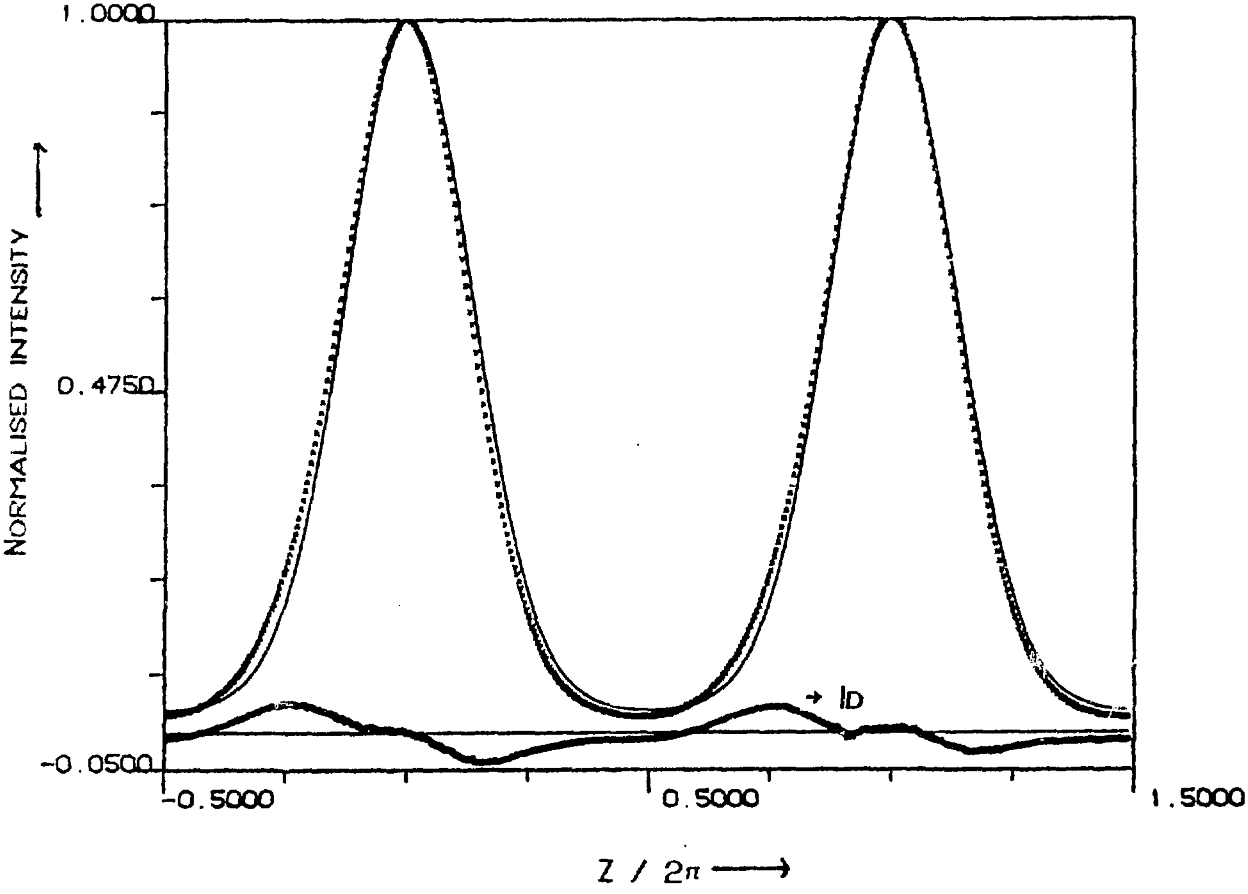

FIG. 3. EXPERIMENTALLY RECORDED DATA POINTS (*) ALONG WITH THEORETICAL FIT (-) FOR 3472.571 Å NEON LINE EXCITED IN A HOLLOW CATHODE DISCHARGE LAMP. THE DATA POINTS WERE OBTAINED WITH FSR = 415.76 mK (THE SEPARATION BETWEEN TWO ORDERS IN THE ABOVE FIGURE). THE DIFFERENCE BETWEEN THE EXPERIMENTAL PROFILE AND THE THEORETICAL FIT IS SHOWN AS A DIFFERENCE CURVE I_D.

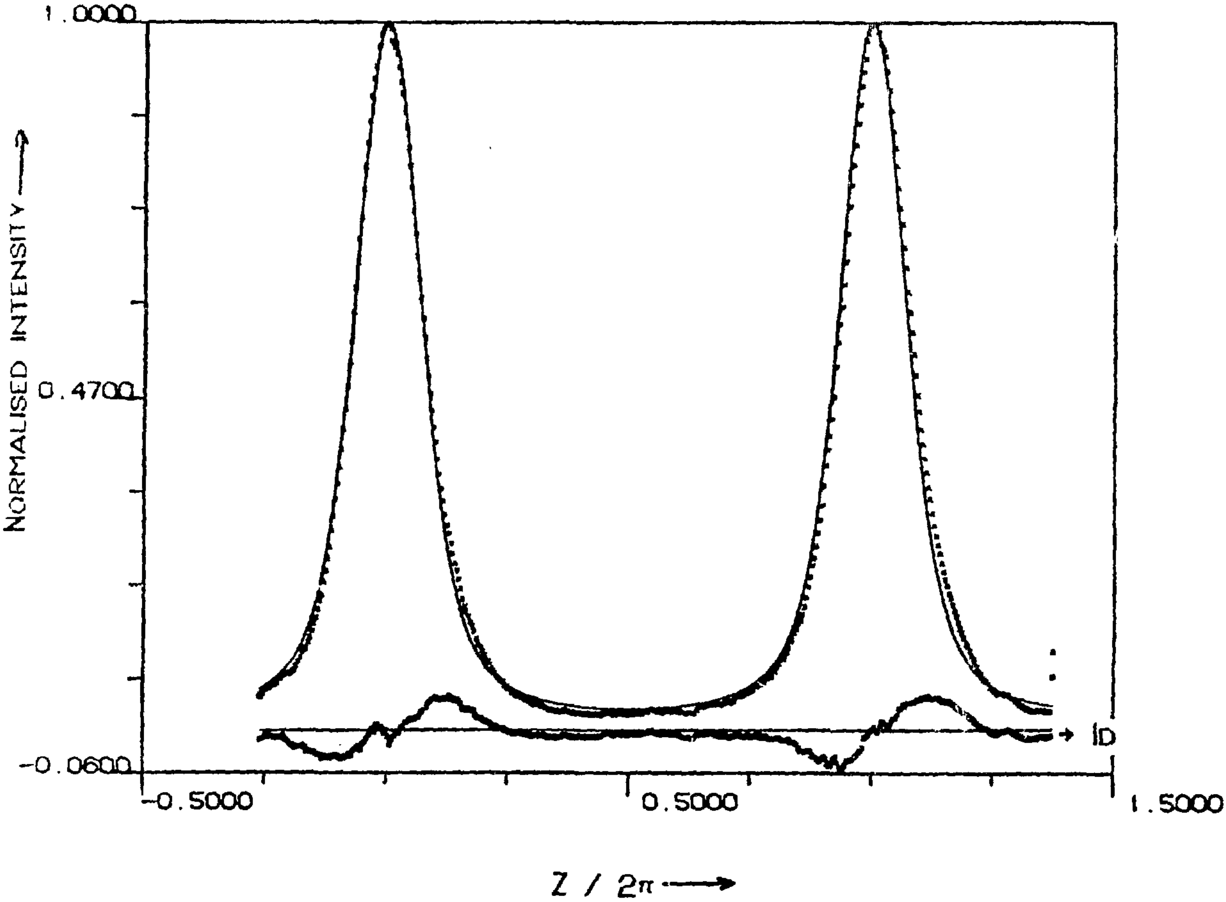

Fig. 4. Experimentally recorded points (*) along with theoretical fit (--) for 3494.40 Å Gadolinium line excited in a hollow cathode discharge lamp. The data points were obtained with FSR = 415.76 mK (the separation between two orders in the above figure). The difference between the experimental profile and the theoretical fit is shown as a difference curve ID.

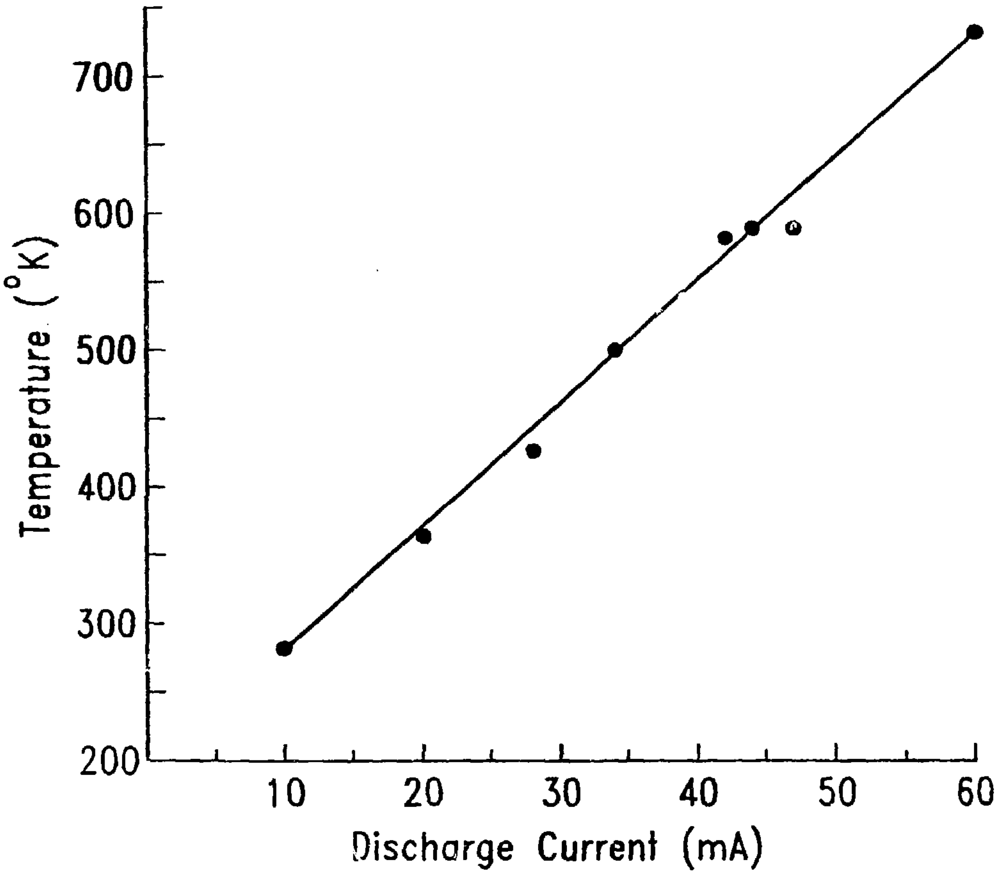

FIG.5. DOPPLER TEMPERATURE OF Ne GAS EVALUATED FOR DIFFERENT HOLLOW CATHODE DISCHARGE CURRENTS.

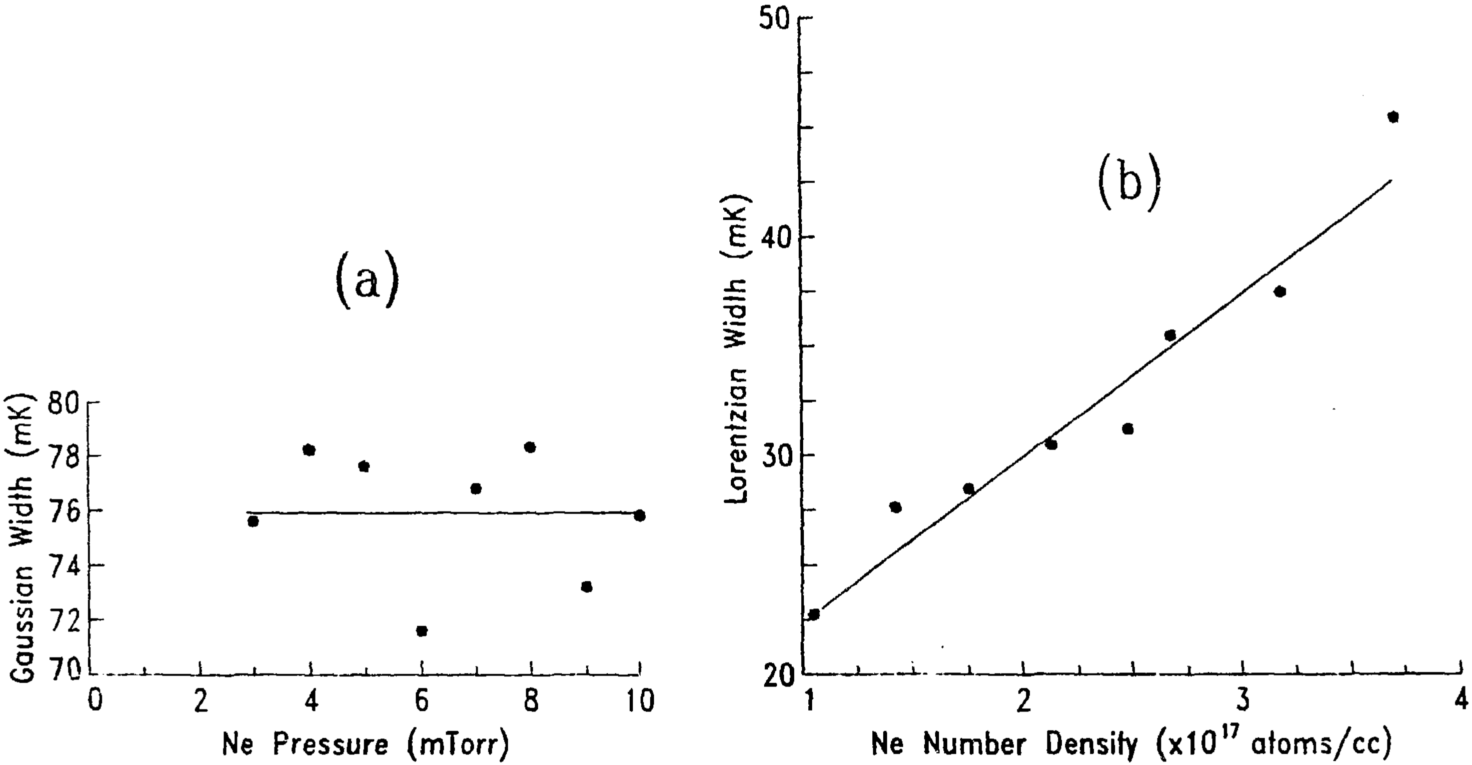

FIG.6. THE GAUSSIAN (a) AND LORENTZIAN (b) WIDTHS OF 3472.571 Å Ne LINE PLOTTED AGAINST NEON GAS PRESSURE AND NUMBER DENSITY RESPECTIVELY. THE EXPERIMENTAL POINTS ARE REPRESENTED WITH '•' AND THE SOLID LINE REPRESENTS LEAST-SQUARE FIT OF THE DATA POINTS.

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