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High resolution optogalvanic study in nitrogen discharge

P R SASI KUMAR, B AMBADI, V P N NAMPOORI and C P G VALLABHAN
Laser Division, Department of Physics, Cochin University of Science and Technology, Cochin 682022, India

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Abstract. Doppler limited high resolution spectrum in the wavelength region 17224 to 17236 cm^{-1} of the first positive system ($B^3\Pi_g$–$A^3\Sigma_u^+$) of the N_2 molecule is recorded by optogalvanic spectroscopic technique using a single mode ring dye laser. It is observed that the intensity and line width of the rotational line increase with the discharge current. Dependence of the collision broadening coefficient on the current was also evaluated.

Keywords. Optogalvanic spectroscopy; nitrogen molecule; rotational analysis.

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1. Introduction

Optogalvanic spectroscopy (OGS) developed by Green *et al* [1] provides an efficient method to study transition between excited states of atoms, molecules etc. The OG effect is essentially the change in the impedance of a gas discharge caused by resonant absorption of radiation and is widely used for spectroscopic studies [2]. High resolution spectroscopic studies can be conveniently carried out with a single mode tunable laser using this technique [3].

Optogalvanic effect in N_2 discharge will give strong signal corresponding to vibrational and rotational transitions. The first positive system of nitrogen molecule is the most prominent band system and has been the subject of many detailed investigations [4]. Feldmann [5] and Ullas *et al* [6] have studied the OG spectrum of N_2 molecule at low resolution. In this paper we report the recording of high resolution spectrum of nitrogen molecule in the region 17224 to 17236 cm^{-1} corresponding to a part of the (11, 7) band of the first positive system ($B^3\Pi_g$–$A^3\Sigma_u^+$). Variation of signal strength and fwhm with discharge current is also studied. Due to the presence of a large number of branches the first positive system shows a very complex structure.

2. Experimental details

The experimental arrangements employed to record high resolution OG spectrum is shown in figure 1. A positive column discharge was obtained in a specially designed discharge cell made of stainless steel electrodes fixed at both ends of a glass tube. The N_2 gas was allowed to flow continuously while the pressure inside the discharge tube was maintained at 0·9 mbar. A steady dc voltage was applied through a ballast resistance (105 KΩ) across the electrodes at a discharge current of 4.5 mA. Radiation from a single mode ring dye laser (spectra physic 380D) operating on Rh6G dye

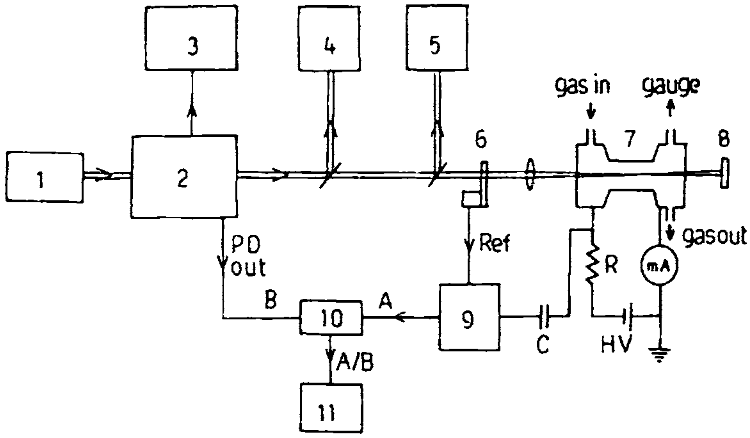

Figure 1. Experimental setup for high resolution OG spectroscopy. 1. Argon laser, 2. ring dye laser, 3. stabilock scanning electronics, 4. spectrum analyzer, 5. wavemeter, 6. chopper, 7. discharge cell, 8. beam stopper, 9. lock-in amp, 10. ratio meter, 11. chart recorder.

pumped by Ar ion laser (spectra physic 171) was passed axially into the discharge. The beam was mechanically chopped at 47 Hz. Stabilock scanning electronics (spectra physic 388, 389 and 481B) were used for frequency locking and the dye laser was scanned over a range of 30 GHz in each sweep. Output power of the laser beam was 300 mW. The OG signal generated was detected using a lock-in amplifier (EG & G 5208), the output of which is given to a chart recorder. The dye laser intensity profile variation was eliminated by normalizing the OG signal with the photo diode output of the dye laser using a ratio meter (EG & G 193). Wavelength of the beam was measured by a wave meter (Burliegh WA-20) and the OG spectrum was recorded by scanning the dye laser from 17224 to 17236 cm^{-1} (in vacuum wave number) in steps of 30 GHz.

3. Results and discussion

3.1 *Low resolution N_2 spectrum*

We have recorded the low resolution OG spectrum (without using frequency locking electronics) in the 570–600 nm region of the first positive system of N_2 at different discharge currents and pressures. Figure 2 shows the spectrum obtained at a pressure of one mbar and current 3·75 mA. The spectrum we obtained had a very good S/N ratio as compared with the earlier studies [5, 6] and all the vibrational bands in this wavelength region ((8, 4), (9, 5) (10, 6) (11, 7) and (12, 8)) are well developed. In general it is observed that the OG signal intensity for different vibrational bands decreases with reducing discharge pressure and current.

3.2 *High resolution spectrum*

Doppler limited high resolution spectrum in the region 17224 to 17236 cm^{-1} of the first positive system of N_2 molecule is shown in figure 3. $B^3\Pi_g$–$A^3\Sigma_u^+$ system of N_2 consists of three subsystems, ${}^3\Pi_0(F_1)$–$A^3\Sigma_u^+$, ${}^3\Pi_1(F_2)$–$A^3\Sigma_u^+$ and ${}^3\Pi_2(F_3)$–$A^3\Sigma_u^+$. These transitions are composed of 27 branches (nine main and eighteen satellites) with three main and six satellite branches contained in each of the subsystem [7].

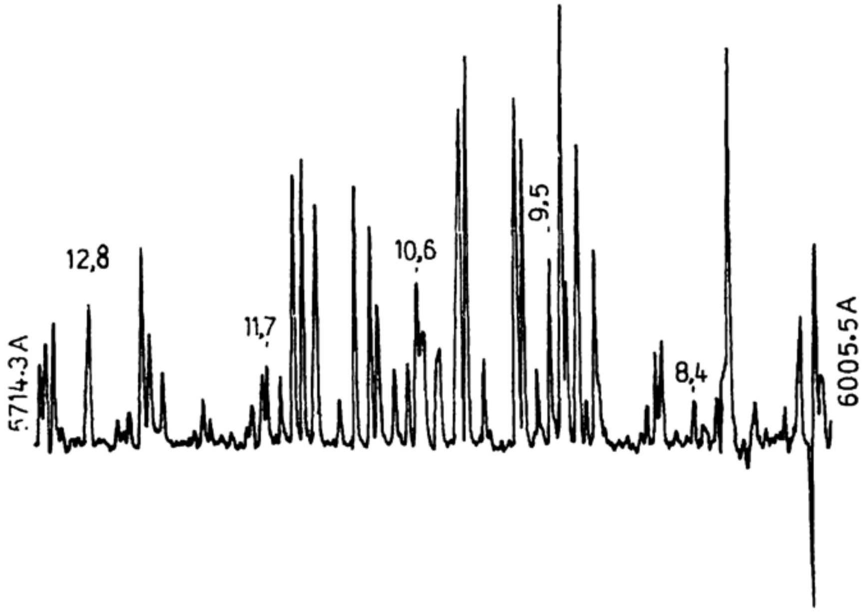

Figure 2. OG spectrum of N_2 at low resolution.

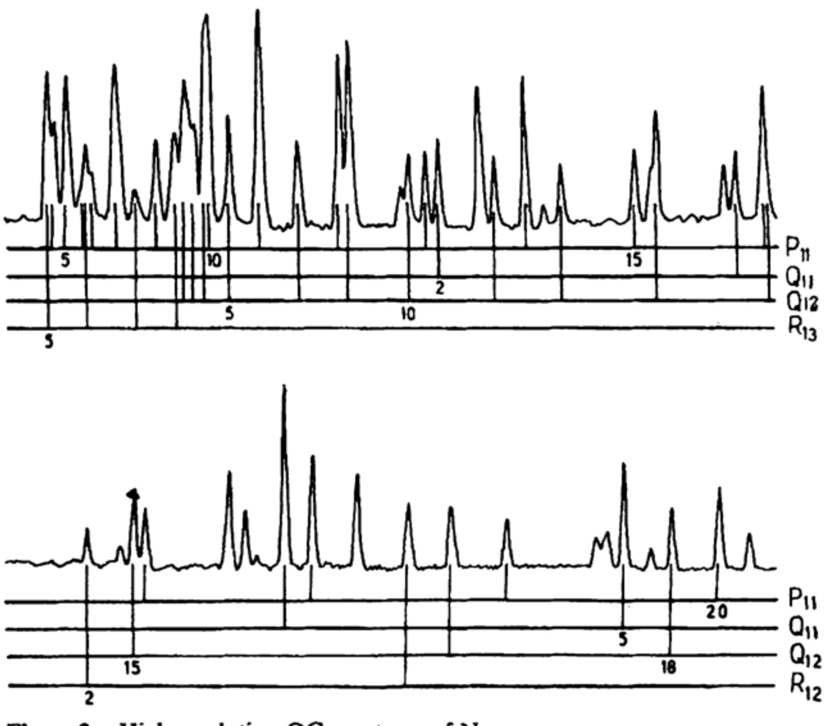

Figure 3. High resolution OG spectrum of N_2.

The observed rotational lines correspond to the $^3\Pi_0(F_1)$–$A^3\Sigma_u^+$ sub-transition and most of them have been assigned to P_{11}, Q_{11}, Q_{12}, R_{12} and R_{13} branches by comparing with the published data [8] and are given in table 1. Other branches will be extended towards higher wave number side and complete rotational analysis can be carried out only after recording and assigning all the branches in the band system.

Table 1. J assignment and vacuum wave numbers (cm^{-1}) of various rotational lines of the (11,7) band of the first positive system of N_2.

J	P_{11}	Q_{11}	Q_{12}	R_{12}	R_{13}
2		17227·38	17225·23	17230·99	
3		17230·01	17225·35	17233·74	
4	17224·26	17232·65	17225·45		
5	17224·37	17235·45	17225·64		17224·21
6	17224·45				17224·51
7	17224·59				17224·98
8	17224·81		17226·24		17225·21
9	17225·11		17226·65		
10	17225·47		17227·10		
11	17225·90		17227·82		
12	17226·57		17228·44		
13	17227·25		17229·26		
14	17228·11		17230·26		
15	17229·03		17231·37		
16	17230·21				
17	17231·48		17234·09		
18	17232·91		17235·80		
19	17234·54				
20	17236·26				

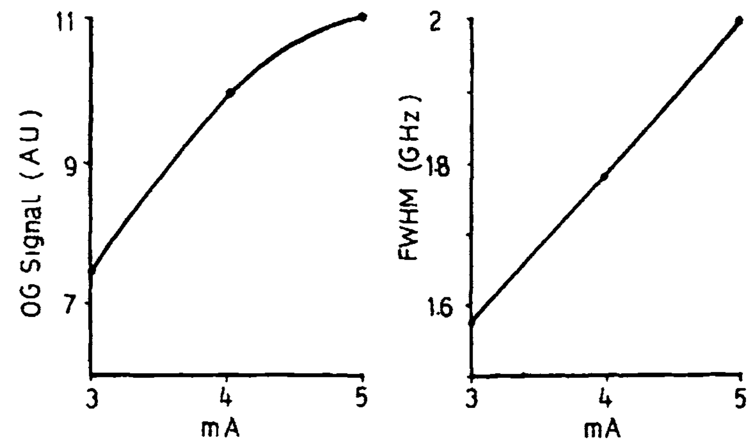

Figure 4. Variation of OG signal and fwhm at 17388·7 cm^{-1} as a function of discharge current.

3.3 *Calculation of line broadening coefficient*

It is found that the OG signal strength increases with current due to the enhanced cross section of molecular excitation caused by electron collision. At the higher current signal shows a saturation effect. Similar behaviour is observed when the pressure was varied. Figure 4 shows the variation of the magnitude of the OG signal and fwhm as a function of the discharge current at a pressure of 3 torr for a typical rotational line at 17388·7 cm^{-1}.

Enhancement in electron collision with the molecules will result in line broadening as observed in figure 4. Collision broadening coefficient of the transition among the

Table 2. Broadening coefficient of N_2 for different discharge current.

Discharge current I(mA)	fwhm γ (cm^{-1})	Temperature (T)K	Broadening coefficient cm^{-1}/molecule/cm^{-3}
3	0·0523	495	$1{\cdot}16 \times 10^{-19}$
4	0·0595	640	$1{\cdot}50 \times 10^{-19}$
5	0·0666	804	$1{\cdot}89 \times 10^{-19}$

excited states can be measured by OG spectroscopy within the Doppler limit [9–11]. It is found that fwhm (γ) varies linearly with the current (I) according to the relation,

$$\gamma = \gamma' + \beta I. \tag{1}$$

The value of β evaluated from the slope of the γ–I plot was $6{\cdot}8 \times 10^{-3}\,cm^{-1}$/mA. Assuming Doppler effect as the dominant line broadening process [12] the average temperature (T) within the discharge column can be calculated from the line width of the measured OG signal for various currents using the formula,

$$\Delta\nu = \frac{2}{c}\nu\left(\frac{2RT}{M}\log 2\right)^{1/2} \tag{2}$$

where $\Delta\nu$ is fwhm in Hz and ν is the frequency at the line centre, M is the molecular weight, and R is the gas constant. Results show increase in temperature as current is increased. Knowing the pressure (P), which is 3 torr in the present case and the temperature (T), one can calculate the number density of the molecules using the relation $n = P/kT$. From this, the broadening coefficient which is defined as β/n can be calculated. The relevant results obtained from the present study are given in table 2.

4. Conclusions

Part of the high resolution spectrum of $B^3\Pi_g$–$A^3\Sigma_u^+$ transition from 17224 to 17236 cm^{-1} region of N_2 molecule is recorded and the assignment is given for the transitions. The magnitude of the OG signal and fwhm are found to increase with discharge current and pressure. The collision broadening coefficient for the transition at 17388·7 cm^{-1} is determined.

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