RÔLE OF ARGON IN THE PRODUCTION OF SWAN BANDS.

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I. *Introduction.*

IN a discharge tube, generally three factors affect the distribution of intensity among the lines of a spectrum. These three factors are :—(i) electrical conditions of excitation, (ii) the pressure of the gas in the discharge tube and (iii) the presence of impurities. Nature of changes in the distribution of intensity due to these factors is the selective transfer of energy from higher term members to lower term members of the series. The presence of the foreign gas in a discharge tube plays an important part, the effects due to this being quite different from those due to the electrical conditions of excitation and the pressure of the gas.

It has been shown in several papers that an admixture of the inactive gas with substances emitting spectra results, in certain cases, in profound modification of these spectra. In some cases, this is in the form of redistribution of spectral intensity, but in others, an isolation of spectra entirely new, is effected. Remarkable results of this kind have been obtained by Merton[1] while working with helium. This includes, a redistribution of intensity in the secondary spectrum of hydrogen and under other conditions, a peculiar facility in isolating higher term members of the Balmer series of hydrogen. In connection with carbon, helium favours the production of certain spectra, appropriate to extremely low pressures and with other spectra, effects an isolation, otherwise difficult or impossible. Various authors have studied the effects of foreign gases, like helium, argon and neon on spectra of many substances, such as hydrogen, carbon, oxygen, nitrogen, sulphur and many others. In view of this work, it occurred to us to study certain aspects of the isolation of spectra under the influence of foreign gases. In particular, there are various spectra associated with carbon. In this connection, we can point out the different band systems attributed to carbon which occur with the change in condition of excitation or change of pressure or in the presence of certain foreign gases. Johnson[2] has shown that in a discharge tube of "H" pattern having a capillary and fitted with carbon electrodes, green light of exceptional brilliance is emitted, if the tube be filled with argon at a pressure of about 30 mm. This stage comes only when the tube is in the right condition. The presence or absence of hydrogen has a marked effect on the

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emission of this light which is due to Swan system. In order to follow this effect of high pressure argon on the isolation of Swan system, we propose to make a systematic study of the intensity changes that take place in the Swan system by the gradual addition of argon to the discharge tube of the type indicated above. The necessity of such an investigation has been pointed out already, by Johnson and Tawde.[4] We shall confine our investigations to only five different pressures (namely 10, 15, 20, 25 and 30 mm.) of argon.

II. *Experimental.*

The Swan bands were excited in discharge tubes filled with argon at different pressures. Such tubes were of the usual "H" pattern having one carbon electrode and one aluminium electrode. The light was examined through the spectroscope by focussing a capillary portion on the slit. The capillary was about 20 cm. in length and 1·5 mm. in diameter. Two small bulbs containing P_2O_5 and KOH were attached to the discharge tube. The palladium regulator was also attached to admit or remove hydrogen from the tube. Spectrally pure argon obtained in small glass cylinder from the British Oxygen Co., Wembley, was used for these experiments. Pressures were measured by a mercury manometer attached to the tube. It was necessary to use liquid-air trap to catch mercury vapours from the manometer diffusing in the discharge tube, as it is known that the argon at high pressure, tremendously enhances the arc spectrum of mercury. The liquid-air trap minimises the trouble to a great extent, though it does not completely remove the mercury vapours. This was evident from the slight traces of intense mercury lines found in the spectrum. The tubes were filled at required pressures (namely 10, 15, 20, 25 and 30 mm.) respectively, sealed and taken for photographing the spectrum, as required. Before doing that, the palladium regulator attached to the tube was heated for a sufficiently long time to remove hydrogen from the tube. This can be done easily when the regulator is heated in the very tip of a Bunsen flame. When hydrogen is completely removed the tube showed only Swan bands and almost negligible traces of intense mercury lines.

In the work of this type where the investigations are concerned with the vibrational intensities, a moderately low dispersion instrument was necessary. So the spectrum was photographed on a Hilger constant deviation spectrograph having dispersion of about 70 Å.U. per mm. at λ 6000 and about 20 Å.U. at λ 3000. The slit of the spectrograph was adjusted to a value which just made the fine structure of bands to disappear leaving the band structure almost a continuous spectrum. The spectrum was photographed by focussing on the slit of the spectrograph, a sharp image of the capillary nearer the carbon electrode by means of a condensing lens.

The discharge was run by a large induction coil giving a high-frequency voltage discharge, no condenser or spark gap being used in the circuit. The primary voltage was 18 volts and with it, the current in the secondary was 50 milliamps maintained fairly constant throughout the work.

Ilford Rapid Process Panchromatic Plates (backed ones) were used. On each plate, two sets of spectra and calibration marks were taken, one set being for a group of strong bands and the other for a slightly weaker group. For the correlation of intensities of bands from one set to another, bands of intermediate intensities were kept common to both the sets. The spectrum was photographed alongside a set of calibration marks due to a standard lamp. For this, a step-slit having six slits of widths, 0·198 ; 0·296 ; 0·471 ; 0·723 ; 1·326 and 2·098 mm. was used. These widths of the slits were so selected that the minimum was such as not to cause any diffraction effects and the maximum was just sufficient to avoid extraneous light filling the spectrograph. This step-slit was illuminated by a standard lamp of known energy distribution. In order to produce uniform illumination over all the slits of the step-slit, the lamp was accurately placed in line with the axis of the collimator tube, at a sufficiently long distance, no lens being used. The lamp used was a special U.V. glass tungsten spiral filament lamp supplied by the General Electric Company. During the time of exposure, the value of the current in the lamp was maintained at a fixed value of 1·9 amperes by means of a rheostat in the circuit, as the lamp was calibrated at that value of the current. The calibration of the lamp used in this work, was carried out by Dr. Johnson and one of us (N.R.T.) at the Government Chemical Laboratories, London, and the data of energy distribution supplied to us through the courtesy of those authors. Times of exposure for the spectrum as well as the calibration marks for the standard lamp were adjusted strictly equal and were such that the spectrum blackening was intermediate between the maximum and minimum of the calibration marks. In case of lamp, intensity being very strong in comparison with that of the spectrum, it was necessary to reduce the lamp intensity. This can be easily done by using neutral filters in front of the lamp and also by varying the distance of the lamp from the slit. Thus it was possible to adjust the exposures strictly equal. The plates were developed and fixed with the usual precaution necessary in such a kind of work. Micro-photometering of plates was carried out on a Zeiss self-recording instrument belonging to the Physics Laboratory of the Science College, Patna. For this purpose, two or three plates were selected for each pressure from the several plates taken as above. The instrument was tested for perfect reproducibility by taking a trial record of a spectrum and then running

the spot of light backwards. The record thus obtained should not show any overlapping by another curve. Records were obtained for the required band heads and the calibration marks at the corresponding points.

Records thus obtained enabled the photographic intensities of each band head to be calculated. This was taken corresponding to the peak of each band immediately near the head. They were then expressed in terms of the energy distribution of the standard lamp by means of the calibration curve $(E_\lambda : \lambda)$ of the lamp. The details of these have been sufficiently treated by Johnson and Tawde and others.[4, 10]

III. Results of Observations.

(1) *Intensities*.—The results of final intensity values for five different pressures of argon gas are recorded in Table I below. They are expressed on a relative scale, with (0, 0) band at λ 6165 as 100.

TABLE I. *Intensities.*

Bands	Pressure of Argon				
	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
0, 2	7·08	7·70	5·91	4·79	5·90
1, 3	8·89	15·89	14·12	8·51	9·58
2, 4	6·50	21·15	15·31	9·24	12·49
3, 5	5·13	14·86	10·80	4·98	8·61
4, 6	2·69	10·65	9·25	3·92	6·17
5, 7	1·06	7·89	4·92	2·6	4·35
0, 1	50·60	35·89	41·98	45·98	47·50
1, 2	26·31	32·95	32·75	24·92	25·77
2, 3	17·95	22·10	27·00	17·33	15·16
3, 4	14·71	19·01	21·00	10·29	11·39
4, 5	9·63	11·79	11·43	7·51	5·02
5, 6	5·80	6·85	8·25	4·25	3·20
0, 0	100·0	100·0	100·0	100·0	100·0
1, 1	44·17	61·39	54·03	56·18	47·39
2, 2	12·8	19·79	13·17	13·69	18·48
1, 0	63·47	72·74	65·99	53·55	69·60
2, 1	31·87	62·66	43·95	29·96	33·27
3, 2	24·69	59·17	40·98	24·30	30·19
4, 3	13·95	49·91	29·85	15·93	18·36
5, 4	10·15	38·05	22·16	11·05	15·26
2, 0	17·21	22·71	10·86	14·76	19·01
3, 1	11·52	23·71	12·26	10·18	16·07
4, 2	12·8	30·30	10·98	11·8	20·85

(2) $I/_\nu{}^4$ *Values and Transition Probabilities.*—The values of $I/_\nu{}^4$ have been calculated in order to arrive at transition probabilities and the statistical weights of initial levels. These values are recorded in Tables II, II (*a*) and III. Most probable transitions have been derived for all the

TABLE II.

$I/_\nu{}^4$ *Values.*

Bands	Argon Pressure				
	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
0, 2	10·38	11·39	8·66	7·02	8·65
1, 3	12·47	22·26	19·79	11·93	13·43
2, 4	8·75	28·46	20·61	12·43	16·81
3, 5	6·65	19·27	14·01	6·46	11·17
4, 6	3·48	13·37	11·61	4·92	7·74
5, 7	1·4	9·63	6·01	3·4	5·4
0, 1	50·91	39·13	42·14	46·25	47·79
1, 2	25·55	31·99	31·79	24·19	25·02
2, 3	16·88	20·78	25·39	16·29	14·23
3, 4	13·45	17·37	19·31	9·41	10·41
4, 5	8·59	10·51	10·19	6·69	4·48
5, 6	5·30	6·5	8·6	4·20	2·30
0, 0	71·00	71·00	71·00	71·00	71·00
1, 1	30·49	42·37	37·29	38·79	32·71
2, 2	8·62	13·32	8·87	9·22	13·44
1, 0	31·85	36·50	33·12	26·87	34·93
2, 1	15·71	30·87	21·65	14·76	16·40
3, 2	11·98	28·71	19·88	11·79	14·65
4, 3	6·68	23·91	14·30	7·63	8·80
5, 4	4·82	18·06	10·52	5·25	7·24
2, 0	6·32	8·34	3·99	5·42	6·98
3, 1	4·19	8·62	4·46	3·70	5·84
4, 2	4·62	10·83	3·96	4·26	7·52

TABLE II (a). $\Sigma\, I/_\nu{}^4$ *Values and Statistical Weights.*

v'' Progression ↓	Argon Pressure				
	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
$v' = 0$	132·30	121·5	121·80	124·27	127·5
$v' = 1$	100·5	133·2	122·00	101·78	106·0
$v' = 2$	56·3	101·8	80·61	58·12	67·9
$v' = 3$	36·4	74·0	57·67	31·46	42·1
$v' = 4$	23·4	58·6	40·76	23·50	28·4
$v' = 5$	11·52	34·2	24·13	12·85	14·9
Total weight for the system →	357·42	523·3	446·97	351·98	386·8

TABLE III. *Transition Probabilities.*

Bands	Argon Pressure				
	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
0, 2	0·079	0·094	0·071	0·056	0·068
1, 3	0·124	0·167	0·162	0·117	0·126
2, 4	0·156	0·287	0·256	0·213	0·247
3, 5	0·184	0·261	0·243	0·207	0·266
4, 6	0·150	0·229	0·289	0·208	0·271
5, 7	0·122	0·281	0·249	0·264	0·362
0, 1	0·385	0·322	0·346	0·373	0·375
1, 2	0·255	0·240	0·267	0·238	0·236
2, 3	0·300	0·204	0·315	0·281	0·211
3, 4	0·371	0·235	0·336	0·299	0·247
4, 5	0·368	0·179	0·254	0·285	0·158
5, 6	0·461	0·190	0·357	0·326	0·354
0, 0	0·537	0·584	0·583	0·591	0·557
1, 1	0·304	0·318	0·306	0·381	0·308
2, 2	0·153	0·137	0·110	0·158	0·197
1, 0	0·318	0·274	0·271	0·264	0·329
2, 1	0·279	0·304	0·269	0·255	0·242
3, 2	0·330	0·384	0·345	0·376	0·347
4, 3	0·286	0·408	0·357	0·323	0·310
5, 4	0·417	0·529	0·436	0·411	0·483
2, 0	0·112	0·082	0·049	0·093	0·103
3, 1	0·115	0·116	0·078	0·118	0·138
4, 2	0·197	0·184	0·100	0·183	0·264

pressures. These values have been tabulated in Table IV, to compare with the theoretical maximum transition probabilities. The latter has been derived from potential energy curves, using Morse and Rydberg functions.[12]

TABLE IV.

Most Probable Transitions in C_2 (Swan) System under Different Pressures of Argon.

Theoretical		Experimental				
Morse	Rydberg	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
		r max. $\longrightarrow$ *r* max.				
1, 0	1, 0	1, 0	1, 0	1, 0	1, 0	1, 0
2, 1	2, 1	2, 1	2, 1	2, 1	2, 1	2, 1
3, 2	3, 2	3, 2	3, 2	3, 2	3, 2	3, 2
4, 3	4, 3	4, 3	4, 3	4, 3	4, 3	4, 3
5, 4	5, 4	5, 4	5, 4	5, 4	5, 4	5, 4
6, 5	6, 5					
7, 6	7, 5 & 7, 6					
		r min. $\longrightarrow$ *r* min.				
0, 2	0, 1	0, 1	0, 1	0, 1	0, 1	0, 1
1, 3	1, 2	1, 2	1, 2 & 1, 3	1, 2	1, 2	1, 2 & 1, 3
2, 4	2, 3 & 2, 4	2, 3	2, 3 & 2, 4	2, 3	2, 3	2, 4
3, 6	3, 4 & 3, 5	3, 4	3, 5	3, 4	3, 4 & 3, 5	3, 5
4, 7	4, 6	4, 5	4, 6	4, 5	4, 5 & 4, 6	4, 6
5, 8	5, 7	5, 6	5, 7	5, 6	5, 6 & 5, 7	5, 7

Intensity Centres.—To compare the gross intensity distribution under all the conditions, it was thought desirable to calculate the intensity centres

as shown by Tawde.[8]   The values of $\bar{\lambda}$ of intensity centres are given in Table V.

TABLE V.

Positions of "Intensity Centres" C_2 (Swan) System.

Sequence	Pressure of Argon				
	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
0, 2	6090·14	6047·38	6050·75	6075·36	6066·26
0, 1	5580·36	5569·52	5569·11	5583·66	5587·09
0, 0	5149·42	5140·01	5148·25	5147·80	5147·36
1, 0	4716·30	4706·68	4710·31	4714·13	4714·45
2, 0	4374·07	4373·50	4372·93	4373·81	4373·63
Intensity Centres of the System ..	5119·50	5064·02	5140·81	5123·63	5091·18

Effective Temperatures.— Though one would expect no appreciable change in temperature from one pressure to the other in the order examined here, the evaluation of these is likely to provide an additional data to study the thermal equilibrium.   On the basis of Boltzmann distribution in the initial states, the temperatures have been calculated from vibrational energy. These are given in Table VI.

TABLE VI.

Effective Temperatures.

Pressure ..	10 mm.	15 mm.	20 mm.	25 mm.	30 mm.
Temperature ..	5500° K	7070° K	6280° K	5630° K	6430° K

IV.   Discussion.

We have found from the intensities that the Condon distribution holds for all the cases studied here.   It is to be noted that this is in agreement with the results of Johnson and Tawde for the case of argon excitation studied by them.   However, in respect of the excitation functions $\Sigma\,I/\nu^4$, the only

case which comes nearer to that of Johnson and Tawde is that of 15 mm. pressure. With this, the statistical weight of the system is 523, the maximum and the only value, nearer to 644 of Johnson and Tawde. One may notice that for this pressure, in comparison to others, the probabilities are unusually strong for the (1, 0) and (0, 2) sequences. But of these the selectivity of (0, 2) sequence has already been noted by Johnson and Tawde.[4]

It can be seen from the following graphs of Figs. 1 and 1 (a), that the intensity distribution in the Swan system as excited in argon gas, has some anomalous features. The graphs show the value of log $I/_\nu{}^4$ of a band in each sequence against the value of $E_{\nu'}$. In every case of argon pressure
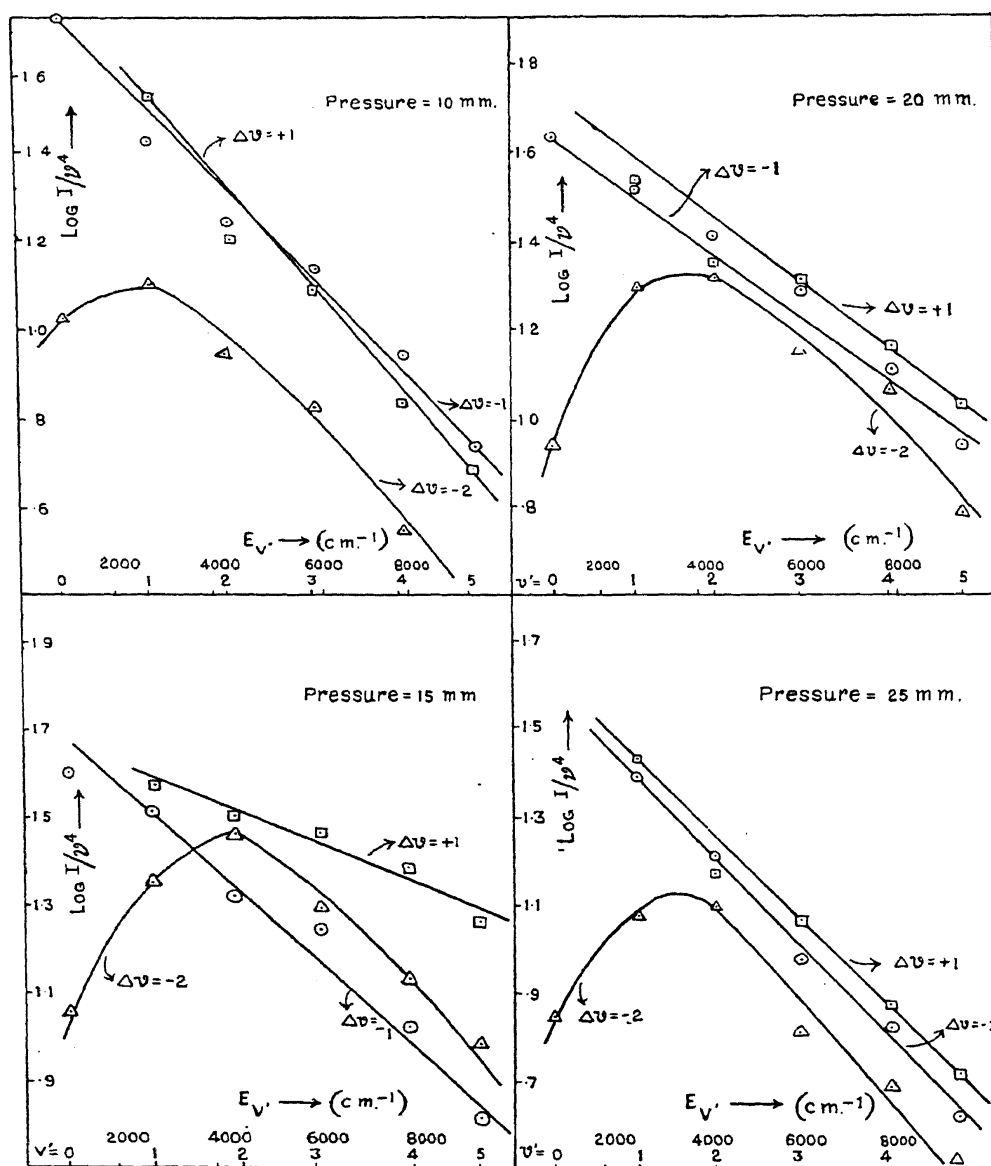
FIG. 1.
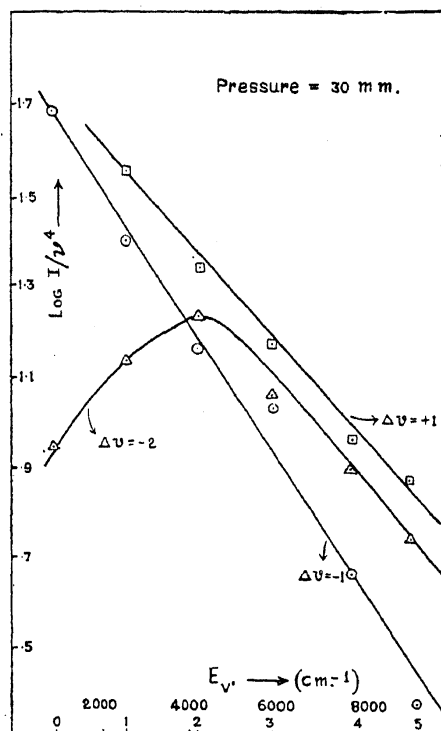
FIG. 1 (a).

that is studied, the sequence with $\Delta v = -2$ shows a relation which is not a straight line; for all other sequences, the graph is more or less a straight line under all conditions. This sequence, as we have noted above, is the one which comes out selectively under the transition probabilities.

It can very well be concluded from the experimental values of Table IV, that the pressure of argon seems to exert practically no effect on the locus of maximum transition probabilities. Comparing, however, these probabilities with the theoretical values, we find that the experimental results are very well agreeable to the theoretical values calculated from the Morse's and Rydberg's potential energy expressions in the region of $r_{max.} \to r_{max.}$. In the region of $r_{min.} \to r_{min.}$, Rydberg's potential energy expression gives much better agreement than Morse's, a result in agreement with the conclusions of Johnson and Dunstan[3] and of Tawde[11] on BeO and N_2 (second positive) systems respectively.

It will be remembered that $\Sigma I/_{v}{}^4$ has been termed as the excitation function which is proportional to the number of molecules in the initial vibrational states. Examination of the same for initial electronic states of the C_2 molecule (Swan system), shows that the change of pressure of the argon gas in the discharge tube does change the excitation function. The change is more marked as we go from lower vibrational states to
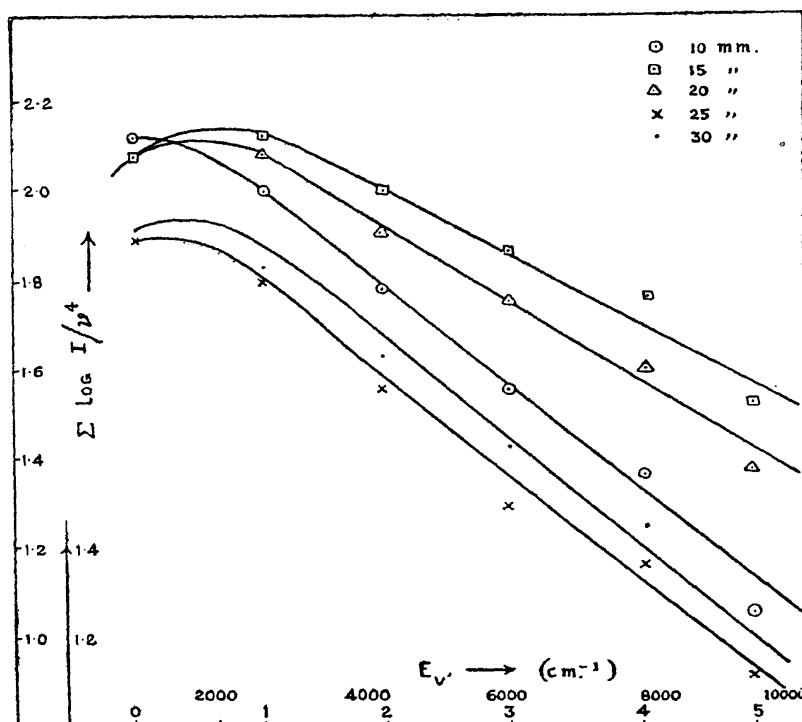
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FIG. 2.

higher ones as can be noticed from the nature of the graphs of Fig. 2. It means that the population of molecules in the higher vibrational levels are more sensitive to changes in the argon pressure. This may be a likely consequence of the greater stability of the first few levels in the excited state, the life period of the molecules within these being higher. To interpret in terms of the collisions of the second kind which we shall discuss in the next paragraph, it seems tempting to assume that a few of the molecules possessing lower vibrational quanta will be able to take energy from the excited argon atoms and turn into molecules of higher vibrational energy and thus cause a relatively large change in the already small populations of the higher states. The relative change thus noticed in the lower states will be comparatively small owing to large populations there. This may be similar to the effect found by Duffendack, Revans and Roy[9] in N_2^+ bands where the energy is spread out towards the higher vibration states when the bands are excited by collisions of the second kind with metastable helium.

Production of C_2 (Swan) System in the Presence of Argon.—As to the manner in which C_2 (Swan) system is excited in the presence of argon it may be mentioned that the explanation given below will be, more or less of a speculative nature, on account of the absence of any accurate knowledge of the energy changes going on in the discharge tube.

In the present case, the discharge tube, as pointed out before, con-tained one carbon electrode and one aluminium electrode and the Swan bands appeared only in a small portion of the capillary nearer the carbon electrode. The minimum pressure of argon necessary for this has been evaluated from the intensity values of the (0, 0) sequence and by drawing a graph of intensity-slopes against argon pressures. From this observa-tion, we have concluded and have reason to believe that for the appearance of (0, 0) band which is the strongest, a minimum pressure of 2 to 3 mm. is necessary and this is sufficiently high for the purposes of explanation that we shall attempt to offer on the basis of Klein and Rosseland's theory of inelastic collisions of the second kind.[13]

Duffendack and Fox[5] have found from experiments that the CO bands can be excited in a discharge tube containing CO gas by direct electron impacts. Duffendack and Smith[6] showed that in the CO—inert gas mix-tures, especially He-CO, the process of direct electron impact is not opera-tive, but they consider the possibility of simultaneous excitation and ionization of CO by excited He and Ne atoms by collisions of the second kind. In view of these contentions we might postulate as follows: If for instance, we assume the energy of the excited argon atoms to be transferred over to CO by collisions of the second kind, we might expect the dissocia-tion of the latter to C and O atoms in their ground-state and the subsequent formation and excitation of the C_2 molecule to emit Swan bands. Carbon being continuously reinforced from carbon electrodes and oxygen being present only as an impurity so as to be a negligible factor in the resultant spectrum, such a process might be said to be highly probable. The only quantitative evidence that we can bring to bear on this view is the energy of the excited argon atoms. These atoms carry various quanta of energy up to a limit of 15·69 electron-volts which is the ionisation potential of argon. This being much in excess of the dissociation energy (about 10 e-volts) of CO, will be able to dissociate it as above, leaving a surplus of less than 5·69 electron-volts, which the argon atoms carry to excite the mole-cular Swan spectrum of carbon (the potential required being 2·4 e-volts). The possibility of this can be demonstrated, if we imagine that before the introduction of the rare gas atoms in the discharge, the entire spectrum is due to Angstrom bands of carbon monoxide, the presence of CO being due to the slight impurity of oxygen left in the discharge tube and invari-ably such is the case in every experiment in which the discharge tube spectra are examined, even though the tube is exhausted to a tolerably low limit of vacuum. In the present case, owing to one electrode being carbon, after the dissociation of CO, it is only the carbon that preponderates and

it is this fact that will bring about the formation of C_2 molecules and their excitation into Swan system as indicated above after the introduction of the high pressure argon. Further support is lent to this by the observation that the Swan bands do not appear all along the capillary but near that end of the capillary where the carbon electrode is fixed. The appearance of the system is strong for a distance of about $1\frac{1}{2}$ inches of the capillary after which it gradually fades. The bands disappeared entirely after a distance of about 4 inches. This may be due to the number of carbon molecules gradually diminishing as we go away from the carbon electrode. In order to test this further, we substituted the aluminium electrode by the carbon so that both the electrodes were similar. Under these circumstances, we found that the original Angstrom bands were replaced in the capillary by Swan bands after the introduction of the argon gas ; but they were strong only at two ends of the capillary. This observation lends further support to the contention that it is the dissociation of carbon monoxide into carbon in an excess, that brings about the appearance of C_2 Swan system and only the collisions of the second kind with rare gas atoms are responsible for bringing this about. After running the discharge tube for a long time and then cutting off the discharge, the tube continues to glow pale white for some time which might resemble the afterglow spectrum of some oxide of carbon.

Examination of temperatures shows that in none of the cases studied, the temperature equilibrium is reached. It is therefore difficult to ascribe a true temperature from the vibrational energy to the Swan system as excited in the present experiments. This non-existence of true temperature can be explained on the basis of general rules given by Oldenberg[7] for determining the true temperatures. According to him a true temperature is indicated in the case of emission spectra excited by electrical discharge at high pressure of added rare gas, if the molecules have a good chance to come to thermal equilibrium during the life-time of the excited state. It is possible in this case that during the life-time of the excited state, the vibrational energy is completely changed due to collisions with excited rare gas atoms. Such is not the case, however, with the rotational energy which is not appreciably changed. In this connection, we can point out the evidence of Duffendack, Revans and Roy[8] who have shown that when N_2^+ bands are excited by impacts of second kind in gas-mixtures, no change in rotational energy occurs. In cases like these, the existence of true rotation temperature as shown by many authors and non-existence of true vibration temperature as found in the present experiments may be a consequence of Oldenberg's rules.

The study of intensity shifts with pressure does not reveal any marked regularity. The wave-length of intensity centres in sequences does not appreciably change as we pass from one pressure to the other. But the intensity centre for the whole system has definite shift towards more refrangible side and occurs at the pressure of 15 mm. of argon. This minimum wave-length coincides with the highest temperature recorded for this pressure within the limits examined, as seen from the following graphs of Figs. 3 and 3 (*a*). In the absence of any knowledge of
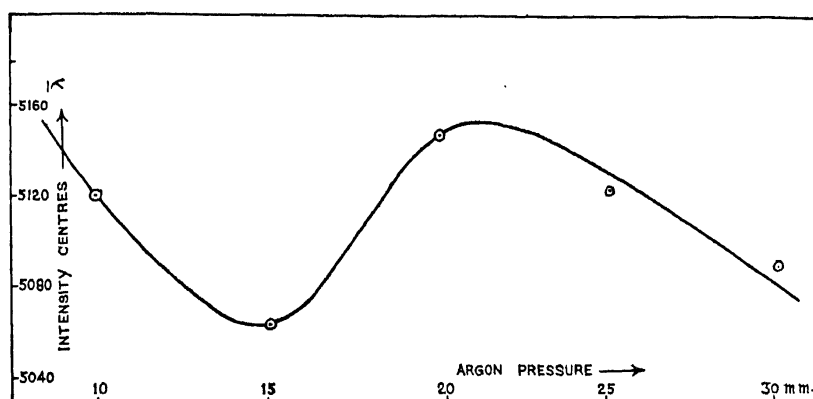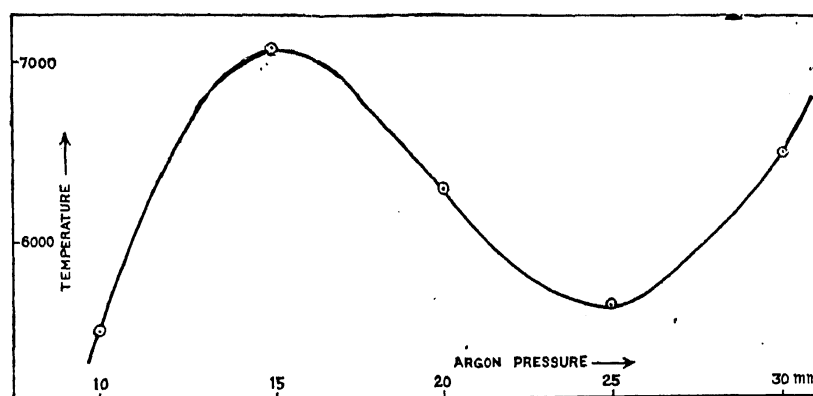

FIG. 3.

FIG. 3 (*a*).

true temperatures existing within the tube, it is impossible to say whether the highest limit of temperature occurs near about this pressure.

It is interesting to add two more relations here, which resemble the nature of graphs, referred to above. They pertain to maxima of the (0, 2) sequence [*cf.* Fig. 1 and 1 (*a*)] and the variation of the statistical weights of the system with pressure [*cf.* Table II (*a*)]. The first relation is the position of maximum of 0, 2 sequence on the E_v', scale and its shift with pressure. This is shown below in Fig. 4. The second relation, namely the variation of $\Sigma I/_v{}^4$ is plotted below, in Fig. 4 (*a*). The graphical relations of Figs. 3 (*a*), 4 and 4 (*a*) resemble each other and the special cases to which they refer require elucidation from theoretical standpoint.
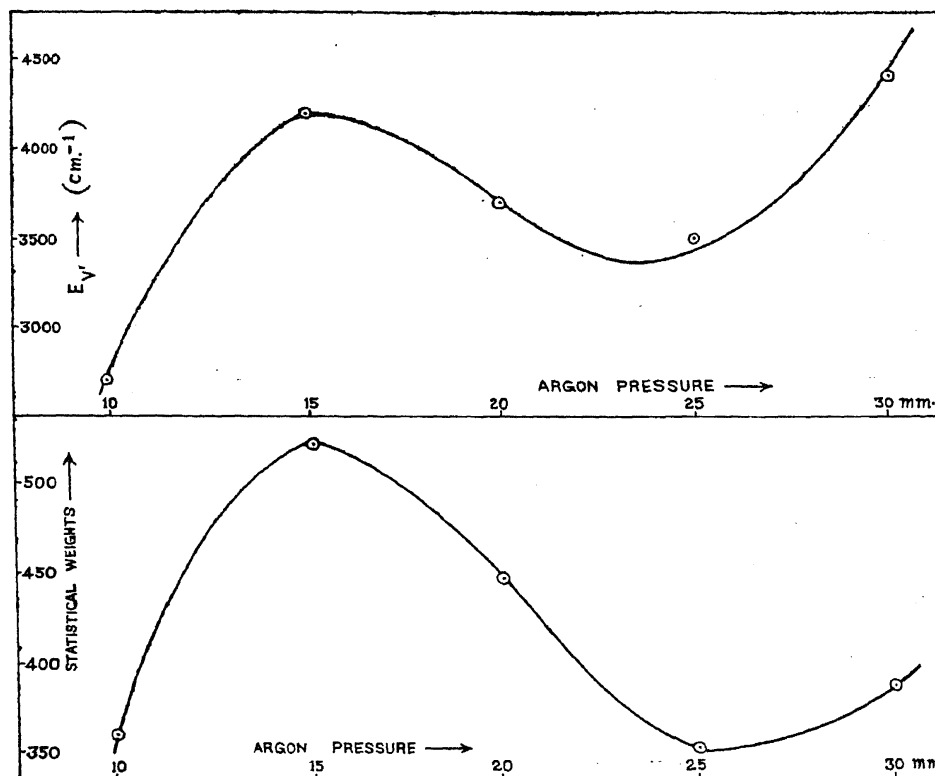
FIG. 4.

FIG. 4 (a).

The authors take great pleasure in expressing their thanks to the authorities of the Science College, Patna, for allowing the use of their microphotometer and to the Principal, Prof. A. T. Mukherjee, for the hospitality extended to one of us (D.D.D.) during the stay at Patna.

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