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Department of Physics
Molecular Excitation Group

Identification Atlas of Molecular Spectra

1: The AlO $A\,^2\Sigma$–$X\,^2\Sigma$ Blue-Green System

D. C. TYTE and R. W. NICHOLLS

March 10, 1964

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ABSTRACT

This report contains a set of vibrationally identified spectrograms of the Blue-Green ($A\ ^2\Sigma$–$X\ ^2\Sigma$) system of AlO. A compilation of all the available molecular data on the states involved, a brief description of the appearance, occurrence and history of the system and a selected bibliography are also included.

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Research grants from The Ontario Research Foundation for the past few years have maintained the continuing "Identification Atlas of Molecular Spectra" project and made possible the production of this report.

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During the past decade, spectroscopic methods and techniques have been increasingly employed for a variety of research purposes in many branches of physics, chemistry, aeronomy and astronomy, and a need for a working knowledge of spectroscopic methods has been felt by many whose main field of research is not spectroscopy *per se*.

Positive identification of spectroscopic features is the first and most common requirement in any application of spectroscopy to research, whatever its orientation. Wavelength lists of many atomic lines and some molecular bands are of course available and well known. Wavelength coincidence alone, however, is not always sufficient for positive identifications and photographic atlases for direct comparison with experimental spectra are always extremely useful.

Such atlases are neither common nor comprehensive. The purpose of the present series of reports is thus to provide photographs (at two or three commonly used dispersions) of important band systems, to each set of which vibrational numbering and a wavelength scale have been added for direct comparisons with experimental spectra for which identifications are sought. The reciprocal dispersions most commonly used in the reports are: high ($\sim$150 Å/mm), medium ($\sim$15 Å/mm) and low ($\sim$5 Å/mm). In addition to the identified photographs each report includes brief comments on the character of the spectrum, its common conditions of occurrence and excitation, some historical background of the relevant research, a bibliography of papers which have been found most valuable in the compilation of the atlas, and a tabulation of essential molecular data. Emphasis is placed on vibrational rather than on rotational structure as it is with vibrational structure of band systems that the first problems of identification usually arise in practice.

The most valuable single aid to identification of molecular spectra is of course, Pearse and Gaydon's indispensable work "The Identification of Molecular Spectra" (1941, 1950, 1963). The tables in this are arranged with the practical spectroscopist in mind, and the photographic reproductions of a number of common and important molecular spectra are particularly useful. The comparable work "Données Spectroscopiques" edited by Rosen (1951) presents the wavelengths of many band systems in Deslandres tables, essential molecular constants and a very complete bibliography (up to 1950). Two significant compilations of molecular spectra which are important in astronomical and aeronomical applications have recently been published by Wallace (1962*a*, *b*). He has the user very much in mind and gives very useful bibliographies.

Phillips and his colleagues at Berkeley (1963) are making distinguished contributions to the detailed compilation of rotational structure (to high quantum numbers) of band systems important in astronomy. Bass and Broida (1953, 1961) have produced valuable spectrophotometric atlases of OH and CH in which the rotational structure is clearly identified. Dieke and his colleagues (1959, 1962) have done a similar service to N_2 and OH band systems. Herman and Hornbeck (1951) published an article some years ago in which vibrational identifications were made of many of the spectra commonly met in combustion spectroscopy.

While the present series of reports is not as ambitious as some of the above works, it is hoped that it will fill a gap which many have felt existed in the reference literature of molecular spectroscopy.

R. W. NICHOLLS, *Editor*.

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1. INTRODUCTION

The blue-green system of aluminium oxide lies in the wavelength range 5700–4200 Å. 100 red degraded bands in the sequences $-4 < \Delta v < 6$ are known. Their wavenumbers are displayed in a Deslandres array in Table II. The most commonly observed bands occur in the sequences for which $-2 < \Delta v < 3$ and for the vibrational quanta $0 < v' < 10$ and $0 < v'' < 9$. However in the (0, 0) sequence no bands are normally observed beyond (3, 3). The most prominent band heads are:

Band	Wavelength (Å)	Approximate Relative Intensity
1, 0	4648.1	2
0, 0	4842.2	10
0, 1	5079.4	1

The system arises from the transition $A\ ^2\Sigma$–$X\ ^2\Sigma$ (ground state) as shown in Figure 1 which relates this transition to the other known

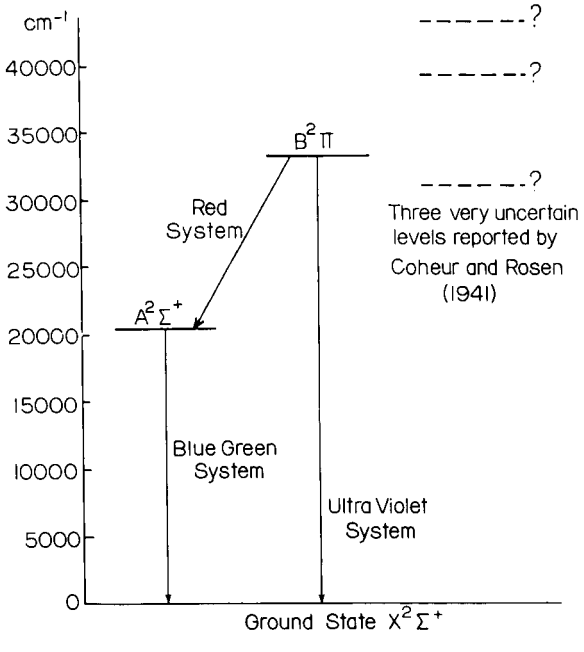

Fig.1 Energy levels and band systems of AlO

AlO transitions. Under moderate resolution each band appears to consist of one P branch and one R branch. The origin of each band is never completely clear. Under higher resolution each "line" of the P and R branches is found to consist of two strong components; the doublet nature of the transition is thus clearly indicated. Weak lines belonging to the satellite ^RQ and ^PQ branches have not yet been observed (a full discussion of the rotational structure of a $^2\Sigma$–$^2\Sigma$ transition is given in Herzberg 1950, page 247).

Klein-Dunham, or realistic, potentials for the $A\ ^2\Sigma$ and $X\ ^2\Sigma$ energy states are shown in Figure 2.

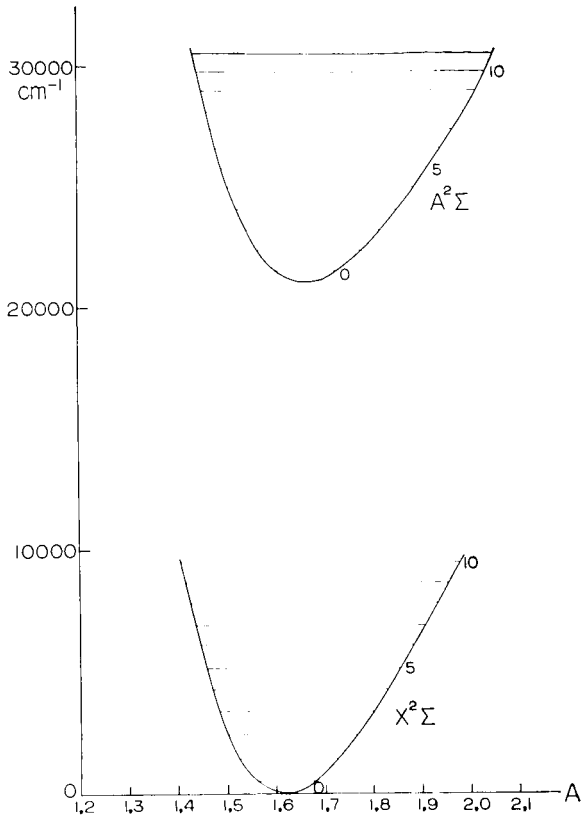

Fig.2 Klein-Dunham Potentials for AlO

2. OCCURRENCE

The blue-green system of AlO is very readily observed both in emission and absorption. Many different types of spectroscopic source excite the system in the emission. Examples of commonly used sources are (1) an envelope of an arc running between aluminium electrodes in air or oxygen (King 1925, Lagerqvist, Nilsson and Barrow 1956, 1957, Shimauchi, 1958), (2) exploding aluminium wires in an oxidizing atmosphere (Rosen, 1944, 1945, 1946, Loginov, 1959), (3)

shock excited Al_2O_3 or $Al + O_2$. Thermal excitation in a shock tube provides a spectrum particularly free of atomic lines (Nicholls, Parkinson and Reeves 1963).

Other laboratory sources which have been used to excite this system include a carbon arc containing aluminium compounds (Tawde and Trivedi 1939) and a high current hollow cathode containing pellets of an Al–Al_2O_3 mixture (Goodlett and Innes 1959).

The system is also of astrophysical importance as a contribution to the spectra of some stellar atmospheres (Davis 1947). It also plays a role in contemporary aeronomical research as a contributor to the emission spectrum of the luminous trails resulting from seeding the high atmosphere with aluminium containing compounds in rocket experiments (Armstrong 1963, Authier *et al.* 1963).

3. HISTORICAL SURVEY

The intense blue-green system of AlO has been known for many years. It was first observed by Thalen in 1866. Many lines of the system were measured by Hasselberg in 1891 and further work was done in 1903 by Lauwartz. The vibrational analysis of the system was achieved independently by Birge and by Mecke in 1925 and in the same year Mulliken (1925) proved conclusively that AlO was the emitter responsible for the system. Mecke based his analysis on the measurements of Mörköfer (1925) and this reference is also said to contain a good review of the work on the system prior to 1925. It is however rather difficult to obtain. Bodson and Dehalu (1937) reported observing tail bands in the $\Delta v = 0$ and 2 sequences and in 1939 Roy made precise measurements of a large number of band heads including many new ones. The most complete vibrational analysis published is that of Shimauchi (1958) who has extended the observed levels to $v = 12$ for the $X\ ^2\Sigma$ state and $v = 16$ for the $A\ ^2\Sigma$ state. She considered that the $v = 9$ level of the $X\ ^2\Sigma$ state is raised about 10 cm^{-1} by a perturbation. More recently Bécart and Mahieu (1963) have reported some 12 new bands.

The correct rotational analysis of the system was accomplished by Pomeroy who, in 1927, analysed the (1, 0), (0, 0), (0, 1) bands. He also corrected the earlier analysis of Eriksson and Hulthén (1925). Sen (1937) and Dehalu (1937) extended the rotational analysis further.

Dehalu (1937), Coheur-Dehalu (1941), and Rosen (1944, 1945, 1946) have reported various perturbations and predissociations in both states basing their observations on intensity anomalies in microphotometer traces of the rotational structure of some bands. However Lagerqvist, Nilsson and Barrow (1956, 1957) have shown by reanalysing some bands and extending the analysis to the (1, 3), (2, 4) and (3, 5) bands that these anomalies are an instrumental effect and do not represent either perturbations or predissociations. Some values for the spin doubling constant are given by Lagerqvist, Nilsson and Barrow (1957).

4. MOLECULAR DATA

The fundamental constants of the $X\ ^2\Sigma$ and $A\ ^2\Sigma$ states of AlO are displayed in Table I.

TABLE I

Constants of the $X\ ^2\Sigma$ and $A\ ^2\Sigma$ states of AlO

μ_A	= 10.0452	
$X\ ^2\Sigma$	T_e	= 0 cm^{-1}
	ω_e	= 979.23 cm^{-1}
	$\omega_e x_e$	= 6.97 cm^{-1}
	B_v	= $0.6413_6 - 0.0058_0\ (v + \frac{1}{2})$ cm^{-1}
	D_v	= $[1.08 + 0.02\ (v + \frac{1}{2})] \times 10^{-6}$ cm^{-1}
	r_e	= 1.6176 Å
$A\ ^2\Sigma$	T_e	= 20688.95 cm^{-1}
	ω_e	= 870.05 cm^{-1}
	$\omega_e x_e$	= 3.52 cm^{-1}
	B_v	= $0.6040_8 - 0.0044_7\ (v + \frac{1}{2})$ cm^{-1}
	D_v	= 1.16×10^{-6} cm^{-1}
	r_e	= 1.6668 Å

(1) Nomenclature as used by Herzberg (1950).
(2) These constants, taken from the work of Lagerqvist, Nilsson and Barrow (1957) are based on band origin measurements.
(3) The dissociation energy D_e of the X state is 5.0 ± 0.2 ev (Drowart *et al.* 1960) from mass spectroscopic measurements. No reliable data appears to be available from spectroscopic methods.
(4) The ionization potential is 9.5 ± 0.5 ev (also based on mass spectroscopic data, Drowart *et al.* 1960).

The band head wavelengths (in air) and the band head wave numbers (*in vacuo*) are displayed in a Deslandres array in Table II. The wave numbers of the band heads are best represented by the formula

$$\nu = 20699.25 + 870.0\ (v' + \tfrac{1}{2}) - 3.80\ (v' + \tfrac{1}{2})^2 - 978.2\ (v'' + \tfrac{1}{2}) + 7.12\ (v'' + \tfrac{1}{2})^2$$

Wavenumbers (in vacuum) of band origins observed by Lagerqvist, Nilsson and Barrow (1957) are displayed in Table III.

TABLE II

Deslandres Table of Band Head Wavelengths (in air) (Å) and Band Head Wavenumbers (*in vacuo*) (cm^{-1})

v' \ v''	0	1	2	3	4	5	6	7	8	9	10	11	12
0	4842.18 20646.1	5079.37 19682.0	5337.03 18731.8	5615.88 17801.7									
1	4648.08 21508.3	4866.23 20544.0	5102.01 19594.7	5357.75 18659.4	5635.60 17739.4								
2	4470.38 22363.2	4671.94 21398.4	4888.83 20449.1	5123.08 19514.0	5376.93 18592.8	5652.21 17687.3							
3	4307.21 23210.4	4493.82 22246.6	4694.40 21296.0	4910.02 20360.8	5142.60 19440.0	5394.27 18533.0	5666.62 17642.3						
4	4156.80 24050.2	4330.39 23086.1	4516.24 22136.1	4715.50 21200.7	4929.1* 20282	5160.62 19372.1	5409.73 18480.1	5679.45 17602.5					
5		4179.78 23918.0	4352.64 22968.1	4537.57 22032.1	4735.92 21109.3	4946.0* 20213	5176.75 19311.8	5422.70 18435.9	5690.20 17569.2				
6		4041.2 24738.2	4201.81 23792.6	4373.78 22857.1	4557.44 21936.0	4754.04 21028.9	4961.7* 20149	5191.44 19257.1	5434.64 18395.4	5701.8 17533.4			
7			4062.4 24609.1	4222.85 23674.0	4393.81 22752.8	4576.26 21845.8	4771.26 20953.0	4975.5* 20093	5204.25 19209.7	5147.0 18353.6	5704.41 17525.4		
8				4082.7 24486.7	4242.94 23561.9	4412.58 22656.1	4593.85 21762.2	4787.10 20883.6	4994.2 20017.7	5218.0 19159.3	5453.52 18331.7	5708.80 17512.0	
9					4102.6 24367.9	4262.03 23456.4	4430.31 22565.5	4610.07 21685.6	4800.92 20823.5	5009.0 19958.5	5228.4* 19121	5458.94 18313.5	
10						4121.6 24255.6	4280.05 23357.7	4447.08 22480.4	4625.20 21614.6	4817.5* 20752	5016.9 19927.1	5235.2* 19096	5461.0 18296.5
11							4139.6 24150.1	4296.44 23268.6	4462.33 22403.5	4640.7 21542.5	4829.8* 20699	5024.1 19897.3	5242.7* 19069
12									4311.5 23187.3	4478.2 22324.2	4650.30 21498.0		5032.6 19864.9
13									4172.1 23962.0		4490.38 22263.6	4662.8 21440.3	
14										4190.8 23855.1	4341.0 23029.7	4502.34 22201.5	
15													4512.3 22155.5
16												4216.5 23709.6	4361.5 22905.7

The band heads in this table are taken from Shimauchi (1958) except those marked * which are from Bécart and Mahieu (1963).

TABLE III

Deslandres Table of Frequencies of Band Origins (cm^{-1})

v' \ v''	0	1	2	3	4	5
0	20635.18	19669.95	18718.50			
1		20533.04	19581.49	18644.13		
2					18576.75	
3						18516.17

The band origins can best be represented by the formula

$$\nu = 20688.95 + 870.05\,(v' + \tfrac{1}{2}) - 3.52\,(v' + \tfrac{1}{2})^2 - 979.23\,(v'' + \tfrac{1}{2}) + 6.97\,(v'' + \tfrac{1}{2})^2$$

The position of the vibrational levels (G_v' and G_v'') and separation between levels $\Delta G_{(v+\frac{1}{2})}$ are displayed in Table IV. (Computed from the band head data of Shimauchi, 1958).

Franck-Condon factor and r-centroid arrays appropriate to Morse potentials are displayed in Tables V and VI.

Relative intensities of AlO bands characteristically excited in an A.C. AlO arc are displayed in Table VII and smoothed relative band strengths derived from the data of Tables V, VI, and VII are displayed in Table VIII.

Relative band intensities at an infinite temperature ($\nu^4\, p_{v'v''}$) derived from Table VIII are displayed in Table IX.

TABLE IV

Vibrational Levels and Vibrational Energy Intervals

v	G_v'	$\Delta G_{(v+\frac{1}{2})}'$	G_v''	$\Delta G_{(v+\frac{1}{2})}''$
0	21123.1 cm^{-1}	861.2 (863.54) cm^{-1}	487.9 cm^{-1}	964.2 (965.23) cm^{-1}
1	21984.3	854.3	1452.1	949.3 (951.50)
2	22838.6	847.0	2401.4	934.7 (937.36)
3	23685.6	839.4	3336.1	921.7
4	24525.0	832.1	4257.8	907.3
5	25357.1	824.0	5165.1	893.0
6	26181.1	817.0	6058.1	878.5
7	26998.1	809.1	6936.6	864.5
8	27807.2	802.8	7801.1	859.3
9	28610.0	794.3	8660.4	826.9
10	29404.3	790.0	9487.3	822.7
11	30194.3	783.1	10310.0	803.9
12	30977.4	774.7	11113.9	
13	31752.1	765.3		
14	32517.4			
15		750.2		

The figures in brackets represent energy intervals calculated from the band origin data of Lagerqvist, Nilsson and Barrow (1957).

TABLE V

Franck-Condon Factors

v' \ v''	0	1	2	3	4	5	6	7
0	7.2982−1	2.3789−1	3.0705−2	1.5771−3	1.9682−5	8.9921−8	1.8183−8	7.1036−12
1	2.2444−1	3.5651−1	3.4285−1	7.1471−2	4.6854−3	5.5900−5	9.3316−7	9.2120−8
2	4.0241−2	3.0060−1	1.6040−1	3.7759−1	1.1242−1	8.6846−3	8.6806−5	4.4297−6
3	5.0370−3	8.7939−2	3.0381−1	6.3203−2	3.7775−1	1.4936−1	1.2817−2	8.9526−5
4	4.3343−4	1.5266−2	1.2897−1	2.7522−1	1.9453−2	3.6320−1	1.8100−1	1.6406−2
5	2.0003−5	1.6954−3	2.8999−2	1.5887−1	2.3629−1	3.2152−3	3.4466−1	2.0728−1
6	3.8435−8	9.9463−5	3.9698−3	4.4225−2	1.7773−1	1.9746−1	9.6592−6	3.2774−1
7	2.3041−7	4.4677−7	2.8478−4	7.2186−3	5.9265−2	1.8748−1	1.6320−1	1.9966−3

Data from Nicholls (1962). The negative number in each entry is the power of 10 by which it is multiplied.

TABLE VI

r-centroids (Å)

v' \ v''	0	1	2	3	4	5	6	7
0	1.646	1.727	1.813	1.937	2.215			
1	1.574	1.657	1.739	1.826	1.957	2.295		
2	1.505	1.580	1.669	1.752	1.839	1.979	2.411	
3	1.429	1.511	1.586	1.685	1.765	1.852	2.003	2.606
4	1.326	1.436	1.517	1.591	1.713	1.779	1.866	2.030
5		1.336	1.443	1.522	1.596		1.794	1.881
6			1.346	1.449	1.527	1.600		1.809
7				1.354	1.454	1.531		

Data from Nicholls (unpublished work).

TABLE VII

Relative Intensities

v' \ v''	0	1	2	3	4	5	6	7
0	100	9.4	0.5					
1	17.6	14.3	13.8	1.1				
2	0.7	25.1	3.9	12.3	1.9			
3		4.2	12.6		6.7	1.0		
4			5.3	7.1		2.7	0.2	
5				4.3	3.4_5		1.7	
6					3.5			1.1
7						2.2		
8							1.0	
9								0.4_5

Data from Hébert and Tyte (1964).

TABLE VIII

Smoothed Relative Band Strengths

v' \ v''	0	1	2	3	4	5	6
0	1.00	0.24	0.02				
1	0.40	0.46	0.33	0.04			
2	0.09	0.51	0.20	0.34	0.06		
3		0.19	0.51	0.07_5	0.31	0.07_5	
4			0.28	0.46	0.02_5	0.28	0.09
5				0.33	0.39		0.25
6					0.36	0.31	
7						0.38	

Data from Hébert and Tyte (1964).

TABLE IX

Relative Band Intensities at Infinite Temperature (I_∞)

v' \ v''	0	1	2	3	4	5	6
0	1.00	0.20	0.01				
1	0.47	0.45	0.27	0.03			
2	0.12	0.59	0.19	0.27	0.04		
3		0.26	0.58	0.07	0.24	0.05	
4			0.37	0.51	0.02	0.22	0.06
5				0.43	0.43		0.19
6					0.46	0.33	
7						0.48	

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While not claiming to be exhaustive, it is hoped that this list contains all the major papers relevant to the analysis and understanding of the system and some of the more recent papers which illustrate the varying conditions under which the system is encountered.

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6. SPECTRA

Description of the Plates

Plate I shows the appearance of the system under low dispersion. The spectrogram was taken on a Hilger *f*4 small glass prism spectrograph with a reciprocal dispersion of approximately 80 Å/mm at 5000 Å. The source was a Mach 5.5 shock wave driven with hydrogen into argon containing 5% oxygen and a finely divided sample of aluminium. Band sequences and a wavelength scale are indicated.

Plate II shows the appearance of the system under moderate dispersion. The spectrogram was taken on a Bausch and Lomb 1.5-metre grating spectrograph with a reciprocal dispersion of approximately 15 Å/mm. The source was the same as that used for Plate I. The vibrational quantum numbers of the bands and a wavelength scale are indicated.

Plate III shows the appearance of the system under high dispersion. The spectrogram was taken on a 3-metre (Eagle Mounted) grating spectrograph with a reciprocal dispersion of approximately 5 Å/mm. The source was a high voltage A.C. arc run between aluminium electrodes in an atmosphere of oxygen at a pressure of $\frac{1}{2}$ an atmosphere. The vibrational quantum numbers and band head wavelengths are indicated.

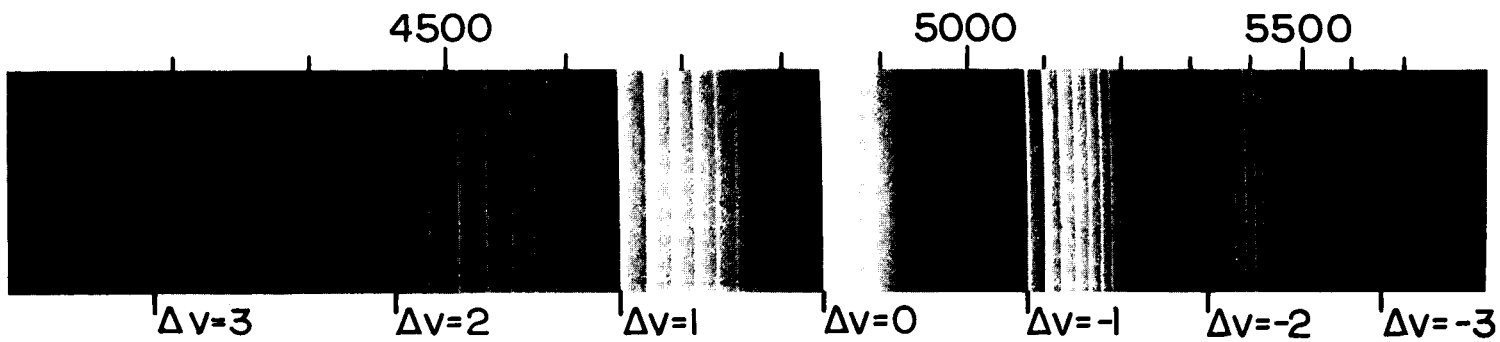
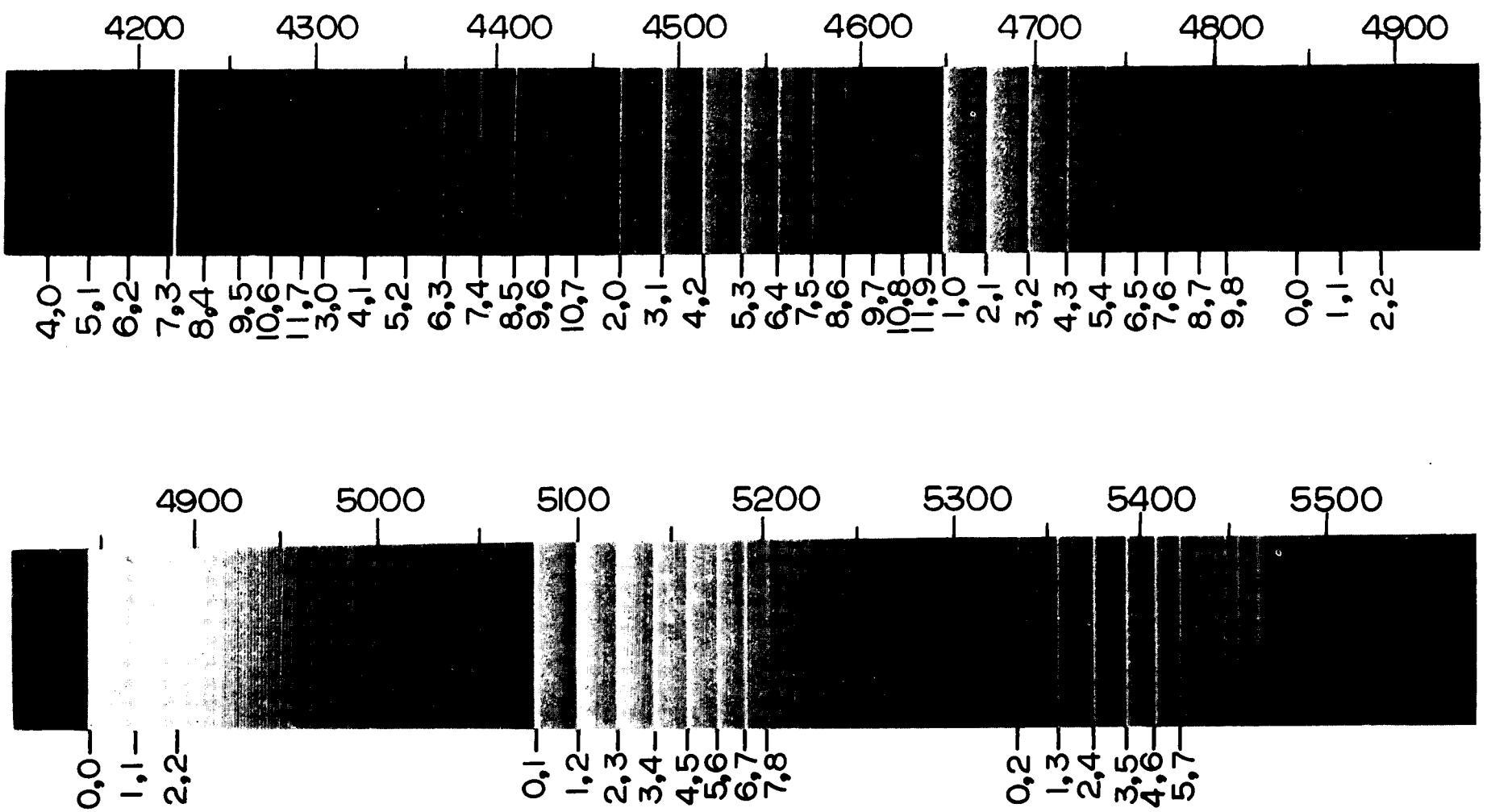


PLATE I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11,7
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5400
5500
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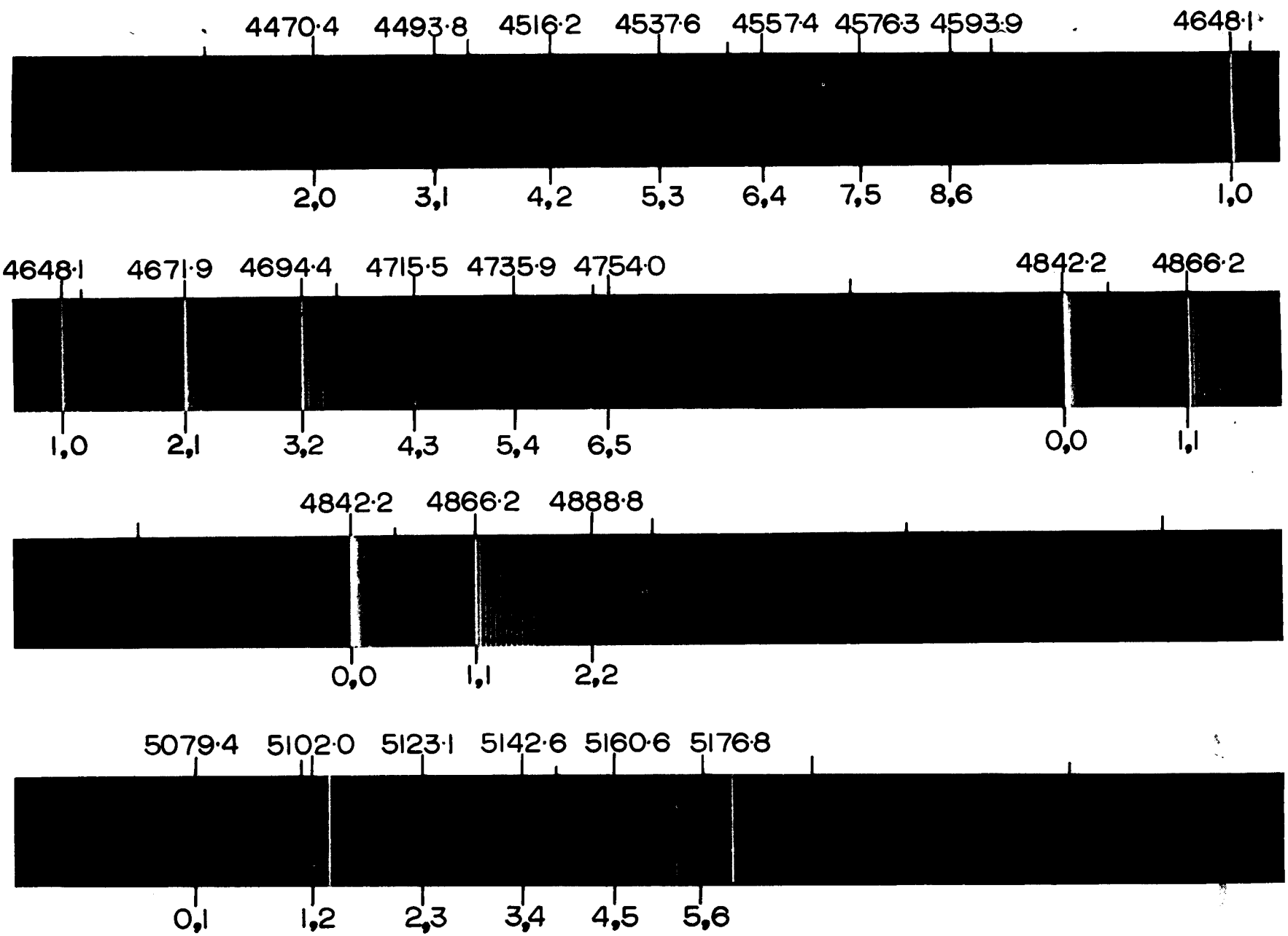
3,5
4,6
5,7

PLATE 2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5160·6
5176·8
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4,5
5,6

PLATE 3