ELECTRONIC TRANSITION MOMENT VARIATION IN ($B\,^1\Sigma \rightarrow X\,^1\Sigma$) BANDS OF BeO

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1. Introduction

The object of the present investigation is to undertake an experimental study on vibrational intensity distribution in electronic bands, and put the data to rigorous test in the light of the recent theoretical developments, particularly with reference to the behaviour of the electronic transition moment variation with internuclear separation r. The band system chosen for study is the BeO ($B\,^1\Sigma \rightarrow X\,^1\Sigma$) system, particularly because of its astrophysical interest. The related oxides too of alkaline earths are of similar interest in stellar sources (Davis, 1947). Much work so far done on this system (Johnson and Dunstan, 1933, Tawde and Hussain, 1949) pertains only to estimation of peak intensity values of the band heads. This is not of much consequence for such finer theoretical aspects as the rôle of electronic transition moment R_e in vibrational transition probabiltiies. Shuler (1950, 1952) has noted that knowledge of variation of R_e with r is a very important factor in any theory of vibrational transition probabilities. This relation is not yet available in the case of BeO ($B \rightarrow X$) system. It is therefore proposed to investigate it here and this involves both experimental and theoretical intensities. Experimental study is proposed to be done by accurate quantitative measures of intensities under intensity contours of unresolved bands. This technique and the relevant theoretical aspects of electronic transition moments are briefly described in the following sections.

2. Experimental

The BeO ($B \rightarrow X$) band system has been excited in a steady carbon arc at atmospheric pressure with pure beryllium (E. Merck, Dermstadt) packed in lower positive carbon electrode. The electrical parameters of the arc are maintained constant throughout the observations. The band system has been photographed on a Hilger glass prism spectrograph with a dispersion

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ranging from 23·3 Å/mm. at λ 4425 Å to 57·4 Å/mm. at λ 5485 Å. The technique of photographic photometry is followed for the quantitative estimation of integrated intensities (Elliott, 1933; Elliott and Cameron, 1934; Tawde and Patankar, 1943). A stepslit and a standard lamp with known energy distribution have been employed for impressing the calibration marks. The spectra are micro-photometered as usual and overlapping, if any, eliminated by careful extrapolation. Each trace of blackening contour of a band has been converted to intensity contour point by point over the whole band and a new intensity wavelength curve prepared with the help of characteristic curves. The area under the contour is computed by means of a planimeter and the necessary corrections applied. The intensities of the bands are ultimately expressed in terms of the energy units of the standard lamp and put on a relative scale with (0, 0) band as 100 units. A large number of plates have been taken to test the reproducibility of results.

The observed intensities I have been used to compute the I/ν^4 values which are taken as the experimental transition probabilities. These have been compared with theory. The theoretical methods of deriving the transition probability are indicated briefly in the following sections.

3. THEORETICAL

Methods Involving Constancy of Electronic Transition Moment

The intensity, I_{em} in emission is given by

$$I_{em}^{v',v''} = A\nu^4 N_{v'} \bar{R}_e^2 \left[\int \psi_{v'}(r)\ \psi_{v''}(r)\ dr\right]^2.$$

Here, $\int \psi_{v'}(r)\ \psi_{v''}(r)\ dr$ is called the overlap integral and its square gives the transition probability or Franck-Condon factor. $\bar{R}_e$ is assumed to vary slowly with r and thus $\bar{R}_e$ is written as the average value of it.

(*a*) *Analytical methods using simple harmonic wave function*

(*i*) *Hutchisson's method.*—The overlap integral for symmetric molecules, of Hutchisson (1930) with applicability extended by Dunham (1930) to asymmetric molecules involves among others the constant C_3, which has been recently corrected by Tawde, Patil and Savadatti (1959). It has been used here for computations of transition probabilities.

(*ii*) *Manneback's method.*—The calculations by the above method of Hutchisson have been confirmed from the recurrence relations provided by Manneback (1951). The equivalence between the two treatments has been recently shown by Tawde, Patil, Sreedhara Murthy and Katti (1957). The

results by the two treatments are expected to be identical according to them. Although both the Hutchisson's and Manneback's methods are analytical, the latter offers a less laborious, simpler and hence a quicker process of theoretical calculations.

(*b*) *Methods using anharmonic wave functions*

(*i*) *Distortion of harmonic to Morse wave functions.*—In this method, the harmonic wave functions are distorted so as to approximate to anharmonic conditions in the manner shown first by Gaydon and Pearse (1939) and later modified and refined by Pillow (1951). This latter has been followed here for the evaluation of the overlap integral graphically.

(*ii*) *Direct use of Morse potential in the wave functions.*—The details of the treatment and its application to the calculation of overlap integrals of the bands of this system are given in an independent paper by Tawde and Sreedhara Murthy (1959). These calculations have been extended to a few more bands in the meantime and have been used for the purposes of the present problem.

The molecular constants needed for the theoretical calculations of vibrational transition probabilities are taken from the compilation of Herzberg (1950).

4. Results and Discussions

(*i*) *Experimental results*

The final corrected values of relative integrated intensities I of bands of BeO ($B \rightarrow X$) system have been entered in column 2 of Table I. Utilizing

TABLE I

BeO ($B \rightarrow X$) *system*

Band v', v''	Integrated intensity I	I/ν^4	Band v', v''	Integrated intensity I	I/ν^4
0, 0	100	49·21	2, 1	3·66	1·44
0, 1	17·7	11·57	2, 2	33·2	17·01
0, 2	0·51	0·45	2, 3	17·3	11·66
1, 0	3·95	1·52	2, 4	0·71	0·64
1, 1	62·8	31·51	3, 2	2·76	1·11
1, 2	23·9	15·89	3, 3	16·6	8·64
1, 3	0·84	0·75	3, 4	12·3	8·38
			3, 5	0·97	0·88

the wavelength measurements of Lagerqvist (1948), I/ν^4 values have been obtained in each case. These have been entered in column 3 of the same table.

(*ii*) *Theoretical results*

The results of Franck-Condon factors derived from Hutchisson's and Manneback's methods are entered in Table II.

TABLE II

Franck-Condon factors
BeO ($B \rightarrow X$) *system*

Band v', v''	Hutchisson	Manneback	Band v', v''	Hutchisson	Manneback
0, 0	0·886	0·886	2, 1	0·204	0·204
0, 1	0·102	0·102	2, 2	0·518	0·518
0, 2	0·011	0·011	2, 3	0·216	0·216
1, 0	0·111	0·111	2, 4	0·050	0·050
1, 1	0·685	0·685	3, 2	0·280	0·280
1, 2	0·172	0·172	3, 3	0·383	0·383
1, 3	0·029	0·029	3, 4	0·239	0·239
			3, 5	0·073	0·073

As expected the values under both the treatments are identical and they should be, so on considerations advanced by Tawde, Patil and Savadatti (1959).

For comparing the theoretical predictions with experimental values, the bands of each v''-progression are considered separately, since $N_{v'}$ populations are said to be constant for the group, and hence we should expect the two to vary relatively in the same proportion. The lowest band of each v''-progression is made to take the value unity for experimental I/ν^4 and similarly $P_{v'v''} = 1$ for theoretical transition probability. The results of other bands of the various v''-progressions are expressed on this basis. These values along with the ratio

$$\beta = \frac{\text{experimental } \frac{I}{\nu^4}}{P_{v'v''}}$$ are given in Table III.

If the theoretical predictions were to approximate to experimental data we would have expected the ratio β to be either unity or in the close neighbourhood of unity. But, that is not so. The ratio on the whole tends to depart widely with increasing v'' (*vide* Table III). The departure seems to be a

TABLE III

v''	Expt. I/ν^4	$P_{v'v''}$ (Manneback or Hutchisson)	$P_{v'v''}$ Pillow	$P_{v'v''}$ Bates	β_{SH} Manneback or Hutchisson	β Pillow	β Bates
			$v' = 0$				
0	1·000	1·000	1·000	1·000	1·00	1·00	1·00
1	0·235	0·115	0·066	0·115	2·04	3·56	2·04
2	0·009	0·012	0·005	0·004	0·75	1·80	2·25
			$v' = 1$				
0	1·000	1·000	1·000	1·000	1·00	1·00	1·00
1	20·730	6·171	6·229	7·120	3·36	3·33	2·91
2	10·454	1·550	0·898	1·780	6·74	11·64	5·87
3	0·493	0·261	0·068	0·100	1·89	7·25	4·93
			$v' = 2$				
1	1·000	1·000	1·000	1·000	1·00	1·00	1·00
2	11·813	2·539	2·823	3·476	4·65	4·18	3·40
3	8·097	1·059	0·790	1·404	7·65	10·25	5·77
4	0·444	0·245	0·006	0·102	1·81	74·00	4·35
			$v' = 3$				
2	1·000	1·000	1·000	1·000	1·00	1·00	1·00
3	7·784	1·368	1·346	2·309	5·69	5·78	3·37
4	7·550	0·854	0·710	1·319	8·84	10·63	5·72
5	0·793	0·261	0·035	0·111	3·04	22·66	7·14

consequence of the approximation taken, *viz.*, the simple harmonic character of the wave function. Actually, such an assumption is far from realisation in electronic bands where departure from parabolic curve towards anharmonic condition sets in after the first few vibrational levels. In considering, therefore, the verification of the theoretical predictions with experimental data, mechanical anharmonicity of the wave function would have to be taken into account. Further, the assumption, made in the two theories that the electronic transition moment is constant over the entire extent of internuclear separation, may not really hold. In order to have a systematic check on the results by inclusion of both of these factors, we shall first attempt to see if introduction of mechanical anharmonicity alone brings about improvement. For this we consider results from the method of Pillow (1951) and of Bates (1949).

(*a*) *Pillow's method.*—The values of transition probabilities obtained by this method are entered in Table IV. They are converted to the form of ratio β as in Manneback's method for comparison with experimental data and entered in last two columns of Table III.

TABLE IV

Transition probabilities (Pillow's New Distortion Process)
BeO ($B \rightarrow X$) system

v' \ v''	0	1	2	3	4	5
0	0·888	0·059	0·004	..	..	..
1	0·118	0·735	0·106	0·008	..	..
2	..	0·181	0·511	0·143	0·001	..
3	..	..	0·286	0·385	0·203	0·010

We find from the values of β (Pillow) that there is still a large gap between theory and experiment although it appears relatively reduced in comparison to results from simple harmonic considerations. It is evident that the departure of theory from experiment increases with higher quanta. This aspect has been the subject of some discussion by Ta-You-Wu (1952). According to him, the wave functions obtained by distortion process have the same amplitude near both the turning points and consequently the error introduced on this account increases with vibrational quantum numbers. He has also

pointed out that there is a lack of orthogonality in the wave functions obtained by distortion process for the various states of anharmonic oscillator. This is also likely to introduce some error in relative transition probability. In our comparative results, the departure noticed may be a consequence of the above considerations, acting alone or in conjunction with the assumed constancy of R_e with r.

(*b*) *Bates' method.*—The resulting values of transition probabilities derived by this method with the use of Morse wave function (Bates, 1949) are entered in Table V. A check on these calculations was provided by the

TABLE V

Transition probabilities—Bates' method

BeO ($B \rightarrow X$) *system*

v' \ v''	0	1	2	3	4	5
0	0·893 (0·897)	0·103 (0·103)	0·004	..	..	..
1	0·100 (0·099)	0·712 (0·718)	0·178 ..	0·010	..	..
2	..	0·166 (0·164)	0·577	0·233	0·017	..
3	..	..	0·207	0·478	0·273	0·023

Note.— First row: Numerical integration (Morse).
Second row: Graphical integration (Lagerqvist, 1948).

values of a few bands available from the work on the verification of the absence of (3, 0) band in the system by Lagerqvist (1948). These values are given in parenthesis in Table V. It is obvious from the magnitude of β (Bates) (Table III) that there is an improvement of results over Pillow's method. However, on absolute considerations there is still an appreciable gap between the theoretical and experimental sets of results, as the latter values continue to be many times the theoretical ones, in some bands. This may mean that the factor of mechanical anharmonicity alone of the wave functions may not be enough in the theoretical concepts. The exact variation

of electronic transition moment R_e, which has been neglected in the theory by considering it as a constant, may have to be taken into account along with anharmonicity. Shuler (1950, 1952) has shown it to be effective in the case of bands of OH ($A\,^2\Sigma \rightarrow X\,^2\Sigma$) system. Hence we shall see how our results on BeO system shape under the contribution of R_e as a function of r, in addition to anharmonicity of the wave functions.

Methods Involving Variation of Electronic Transition Moment

(*a*) *Shuler's procedure.*—The method of Shuler as applied by him to OH ($A \rightarrow X$) system has been extended to the BeO system. For the evaluation of the integral

$$| M_{v'v''} | = \int \psi_{v'}(r)\, \psi_{v''}(r)\, R_e\, dr. \qquad \text{(i)}$$

Shuler has assumed a linear variation of R_e as follows:

$$R_e = c\,(1 + \rho r) \qquad \text{(ii)}$$

where ρ is the expansion coefficient. For a pair of bands, arising from the same initial level, the ratio of quantities I/ν^4 associated with them, will be the ratio (P_{jk}/P_{ji}) of the transition probabilities of these bands. Using this ratio and equations (i) and (ii), ρ can be evaluated with the knowledge of the integrals $\int \psi_{v'}(r)\, \psi_{v''}(r)\, dr$ and $\int \psi_{v'}(r)\, \psi_{v''}(r)\, r dr$.

From the observed integrated intensities of bands in BeO ($B \rightarrow X$) system, the following ratios are obtained:

$$\frac{|M_{00}|^2}{|M_{01}|^2} = 4{\cdot}25_3; \quad \frac{|M_{11}|^2}{|M_{12}|^2} = 1{\cdot}98_3; \quad \text{and} \quad \frac{|M_{23}|^2}{|M_{21}|^2} = 8{\cdot}09_7.$$

The values of integrals $\int \psi_{v'}(r)\, \psi_{v''}(r)\, dr$ and $\int \psi_{v'}(r)\, \psi_{v''}(r)\, r dr$ are available from the results of Bates' method. With the help of these integrals and the above ratios for three pairs, ρ has been evaluated. Since ρ appears as a quadratic in the equation, it has double roots. Out of the two roots, $\rho_{\text{average}} = -0{\cdot}982_2$ and $\rho'_{\text{average}} = -0{\cdot}724_2$, the physically meaningful root $\rho_{av} = -0{\cdot}982_2$ is chosen. Hence the electronic transition moment variation is given by

$$R_e = c\,(-1 + 0{\cdot}982_2\, r)$$

where c is a constant. This expression therefore governs the variation of R_e with r for BeO ($B \rightarrow X$) system. The values of $P_{v'v''}/P_{00}$ obtained from Bates' method have been corrected in terms of the above variation of electronic transition moment and are recorded in Table VI.

TABLE VI

Corrected $P_{v'v''}/P_{00}$ for BeO system

v' \ v''	0	1	2	3	4	5
0	1·000	0·234	0·016	..	..	..
1	0·042	0·853	0·431	0·042	..	..
2	..	0·073	0·741	0·601	0·075	..
3	..	..	0·096	0·658	0·753	0·111

(*b*) *Method of r-centroids.*—From the concepts of *r*-centroids first developed by Nicholls and Jarmain (1956), the following expression involving $R_e(r)$ may be evolved:

$$\left(\frac{I_{v'v''}}{q_{v'v''}\,\nu^4_{v'v''}}\right)^{\frac{1}{2}} = (KN_{v'})^{\frac{1}{2}}\, R_e(\bar{r}_{v'v''})$$

where

$I_{v'v''}$ = measured integrated intensity,

$q_{v'v''}$ = Franck-Condon factor

and

$\bar{r}_{v'v''}$ = *r*-centroid.

Nicholls (1956) has used this to determine variation of $R_e(r)$ with r for each v''-progression. For this purpose, the values of $q_{v'v''}$ and $\bar{r}_{v'v''}$ need to be known. The former of these are available from Table V above under Bates' method. For the values of the latter, *viz.*, $\bar{r}_{v'v''}$ computations are made by procedures which are given in a separate paper under publication elsewhere. These two quantities for BeO bands as well as $I_{v'v''}/\nu^4_{v'v''}$ data are entered in Table VII. A graphical plot of the quantity $(I_{v'v''}/\nu^4_{v'v''}\, q_{v'v''})^{\frac{1}{2}}$ is made against $\bar{r}_{v'v''}$. By adopting the rescaling procedure of Turner and Nicholls (1954) segments are placed on the same ordinate scale. The plot of $R_e(\bar{r}_{v'v''})$ against $\bar{r}_{v'v''}$ is made. By applying the least square technique, the best fit for the straight line is found to be satisfied by the relation of the standard form:

$$R_e(r) = c(-1 + 0{\cdot}990_5\, r),$$

TABLE VII

Band v', v''	$r_{v'v''}$ (Numerical integration, Morse)	$q_{v'v''}$ (Numerical integration, Morse)	$\frac{I_{v'v''}}{\nu^4_{v'v''}}$	$\frac{P_{v'v''}}{P_{00}}$
0, 0	1·352	0·893	49·21	1·000
0, 1	1·492	0·103	11·57	0·229
0. 2	1·642	0·004	0·45	0·015
1, 0	1·222	0·100	1·52	0·043
1, 1	1·363	0·712	31·51	0·849
1, 2	1·508	0·178	15·89	0·422
1, 3	1·662	0·010	0·75	0·041
2, 1	1·227	0·166	1·44	0·075
2, 2	1·375	0·577	17·01	0·736
2, 3	1·524	0·233	11·66	0·589
2, 4	1·685	0·017	0·64	0·074
3, 2	1·232	0·207	1·11	0·098
3, 3	1·388	0·478	8·64	0·654
3, 4	1·541	0·273	8·38	0·736
3, 5	1·708	0·023	0·88	0·107

This equation represents therefore the behaviour of the variation of electronic transition moment $R_e(r)$ with r, the internuclear distance for the bands of BeO ($B\,^1\Sigma \rightarrow X\,^1\Sigma$) system over the range $1{\cdot}222\,\text{Å} > r > 1{\cdot}708\,\text{Å}$. A set of smoothed $R_e(\bar{r}_{v'v''})$ has been obtained from the above equation. These values are multiplied by appropriate Franck-Condon factors, $q_{v'v''}$, to correct the transition probabilities obtained with Bates' method. Thus the final corrected or smoothed set of $P_{v'v''}/P_{00}$ values are evolved for the system which are entered in column 5 of Table VII.

For comparing the transition probabilities obtained independently from Shuler's and r-centroid methods with the experimental data, Table VIII has been compiled.

Discussion

The expression found for $R_e(r)$ by the two independent approaches are:

$$\text{Shuler's method:} \quad R_e(r) = c\,(-1 + 0{\cdot}982_2\, r)$$

$$r\text{-centroid method:} \quad R_e(r) = c\,(-1 + 0{\cdot}990_5\, r)$$

Both the expressions are uniform giving nearly the same expansion coefficient, ρ for r. Of the two methods, the r-centroid method uses larger number of measured intensities for the derivation of the relation, while that of Shuler makes use of only a few measured intensities. The former, *viz.*, the r-centroid method is therefore considered to be a better representative and hence preferred to the latter, *i.e.*, Shuler's method. Both the methods depend on the experimental intensity data and are thus quasi-theoretical. The comparison between the experimental I/ν^4 and corrected or smoothed $P_{v'v''}$ has been made here, only to ascertain whether the linear trend of the variation of $R_e(r)$ is justified. In the last two columns of Table VIII, the ratio β is almost unity for bands in all the sequences except $\Delta v = -2$. This indicates that the corrected or sooothed transition probabilities by both the methods are in general agreement with experimental measures. The large deviations in the bands of $\Delta v = -2$ sequence are attributed to want of precision (1) in experimental intensities due to comparative feebleness of these bands and (2) in theoretical measures due to low values of overlap integrals arising from the large cancellation of positive and negative contributions of the products of the wave functions of comparable magnitude, the partial neglect of vibration rotation interaction and, to some extent, the limited applicability of Morse potential. For these reasons, the evaluation of relation of $R_e(r)$ with r is made independent of these bands by excluding them in the graphical plot.

The relatively much closer agreement between these results of quasi-theoretical concepts and experimental observations on the bands of BeO ($B \rightarrow X$) system confirm the conclusion of Shuler (1950, 1952) that the variation of $R_e(r)$ with r is a great contributing factor, besides the anharmonicity of the wave functions in theoretical considerations of transition probabilities. The results indicate that the assumption generally made that the electronic transition moment is constant along r is therefore not justified.

TABLE VIII

v''	Experimental I/ν^4	$P_{v'v''}$ (Shuler)	$P_{v'v''}$ (*r*-centroid)	$\beta = \frac{\text{Expt. } (I/\nu^4)}{P_{v'v''}}$ (Shuler)	β (*r*-centroid)
			$v' = 0$		
0	1·000	1·000	1·000	1·00	1·00
1	0·235	0·234	0·229	1·00	1·03
2	0·009	0·016	0·015	0·56	0·60
			$v' = 1$		
0	1·000	1·000	1·000	1·00	1·00
1	20·730	20·310	19·744	1·02	1·05
2	10·454	10·262	9·814	1·02	1·07
3	0·493	1·000	0·953	0·49	0·52
			$v' = 2$		
1	1·000	1·000	1·000	1·00	1·00
2	11·813	10·151	9·813	1·16	1·20
3	8·097	8·233	7·853	0·98	1·03
4	0·444	1·027	0·987	0·43	0·45
			$v' = 3$		
2	1·000	1·000	1·000	1·00	1·00
3	7·784	6·854	6·673	1·14	1·17
4	7·550	7·844	7·510	0·96	1·01
5	0·793	1·156	1·092	0·69	0·73

5. SUMMARY

Accurate integrated intensity data of the bands of BeO ($B\,^1\Sigma \rightarrow X\,^1\Sigma$) system has been obtained experimentally by the technique of photographic photometry. With the use of Franck-Condon factors and *r*-centroids for

the bands available from the work of the authors reported in papers elsewhere, the relation of electronic transition moment R_e with internuclear separation r, has been evolved. The vibrational transition probabilities have been corrected for the resulting variation of R_e with r. These corrected values have been examined in relation to those under assumptions of constancy of R_e in conjunction with (i) mechanical anharmonicity and also (ii) mechanical harmonicity.

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7. References

Bates, D. R. .. *Proc. Roy. Soc.*, 1949, **196 A**, 217.

Davis, D. N. .. *Ap. J.*, 1947, **106**, 28.

Dunham, J. L. .. *Phys. Rev.*, 1930, **36**, 1553.

Elliott, A. .. *Proc. Phys. Soc.* (*Lond.*), 1933, **45**, 627.

——— and Cameron, W. H. B. *Ibid.*, 1934, **46**, 801.

Gaydon, A. G. and Pearse, R. W. B. *Ibid.*, 1939, **173 A**, 37.

Herzberg, G. .. *Spectra of Diatomic Molecules*, 2nd Edition, D. Van. Nostrand Co., New York, 1950, p. 509.

Hutchisson, E. .. *Phys. Rev.*, 1930, **36**, 410.

Johnson, R. C. and Dunstan, E. G. *Phil. Mag.*, 1933, **16**, 472.

Lagerqvist, A. .. *Dissertation*, Stockholm, 1948, p. 27.

Manneback, C. .. *Physica*, 1951, **17**, 1001.

Nicholls, R. W. .. *Proc. Phys. Soc.* (*Lond.*), 1956, **69 A**, 741.

——— and Jarmain, W. R. .. *Ibid.*, 1956, **69 A**, 253.

Pillow, M. E. .. *Ibid.*, 1951, **64 A**, 772.

Shuler, K. E. .. *J. Chem. Phys.*, 1950, **18**, 1221.

——— .. *Proc. Phys. Soc.* (*Lond.*), 1952, **65 A**, 12.

Tawde, N. R. and Patankar, V. S. *Ibid.*, 1943, **55**, 396.

——— and Hussain, A. G. .. *Bom. Uni. Jour.*, 1949, **17**, 12.

———, Patil, B. S. Sreedhara Murthy, N. and Katti, M. R. *Physica*, 1957, **23**, 154.

———, and Patil, B. S. *Bull. Soc. Royale, Sciences Liege Belgique*, 1959, **5–6**, 150.

———, Savadatti, M. I. and Sreedhara Murthy, N. *Physica*, 1959, **25**, 610.

Turner, R. G. and Nicholls, R. W. *Canad. J. Phys.*, 1954, **32**, 475.

Wu-Ta-You .. *Proc. Phys. Soc.* (*Lond.*), 1952, **65 A**, 965.