LINE BREADTHS AND ABSORPTION PROBABILITIES IN SODIUM VAPOR

By George R. Harrison[1] and J. C. Slater

Abstract

Molecular broadening of the absorption lines of the principal series of sodium.—From recently published experimental data the theoretical half-breadths of the lines 2 to 8 are calculated, together with the half-breadths in frequency units $(P/2\pi)$, which are proportional to the probabilities. The broadening of the first line is at least 100 times as great as that to be expected from Stark effect and collision broadening and the breadths of succeeding lines are found to decrease rapidly with increasing term number at the saturated vapor pressures used (450 to 600°C), a result contrary to that commonly found at low vapor pressures. It is suggested that this high pressure broadening is due to the presence of a large proportion of diatomic sodium molecules in varying states of stability which disturb the absorbing atoms far more than an inert gas would, the perturbation being greater the lower the quantum state. As evidence for this the *absolute values of the probabilities* A_{ij} *and* B_{ij} come out only about one fiftieth as large as would be expected if all the gas was in the atomic condition; moreover, the bands associated with line 3303 have about 50 times the total intensity of the line. Incidentally, the half-breadth is found to vary approximately as the square root of the number of absorbing atoms.

Molecular aggregation in saturated sodium vapor at high temperatures.—The above results indicate that the proportion of diatomic molecules increases with the temperature, possibly greatly exceeding the proportion of atoms at 600°C. This suggests that the *energy of dissociation of* Na_2 at absolute zero may be negative.

Introduction

In a paper recently published by one of the writers[2] the results of an experimental investigation of the shapes and intensities of absorption lines in the principal series of sodium were given, and from these were calculated the relative probabilities of the various transitions involved, by means of formulas derived by the other writer[3] on the basis of his theory of virtual oscillators. The relative probabilities B_{ij} were obtained for the second to sixteenth lines, and from these were calculated the relative probabilities A_{ij} by means of Einstein's relation, $A_{ij}=(8\pi h\nu_0^3/C^3)B_{ij}$.

In the present paper are given the half-breadths of the second to eighth lines at various vapor densities, and from these are computed the corresponding values of the probability $P/2\pi$, which controls line breadths.

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[1] National Research Fellow.

[2] Harrison, Phys. Rev. **25**, 768 (June 1925).

[3] Slater, Phys. Rev. **25**, 783 (June 1925).

Approximate determinations of the absolute values of A_{ij} and B_{ij} are also given, and the apparent necessity of assuming the presence of large numbers of diatomic molecules of varying grades of stability in saturated sodium vapor at temperatures over 400° C is discussed. The notation in this paper is the same as in the preceding papers.[2,3]

Determination of Line Breadths

It has become the custom to specify the amount of broadening of an absorption line in terms of its half-breadth, or breadth at the point where the amount of absorption is one-half that at the maximum. Theoretically, the full breadth of a single line is infinite, and the absorption is still very appreciable at a point where the breadth is three times the half-breadth, when the broadening is produced by collisions, Stark effect, or other effect giving the broadening formula used in the present work. The shape of a line changes with the distance it has traveled through the absorbing medium, and although each point on the curve obeys the exponential law, its area, and consequently the amount of absorption, does not. This is illustrated in Fig. 1, which shows the changing shape of a line with length of absorbing column x as computed from the formula $\log_{10}(I/I_0) = -C_1/[(\lambda-\lambda_0)^2+C_1C_2]$, where

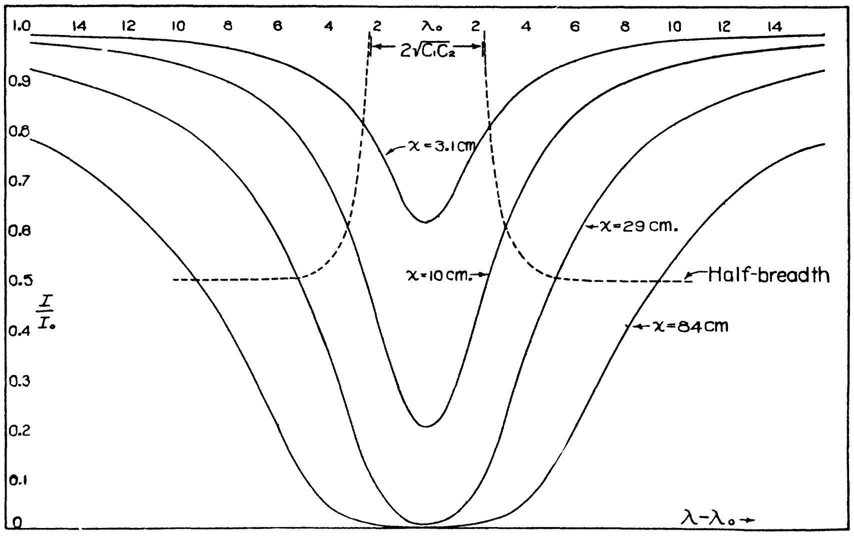

Fig. 1. Curves showing the change in shape of an absorption line with length of the absorbing column. The dotted line shows the changing value of apparent half-breadth with length of column, although the true half-breadth, given by the fundamental value $2\sqrt{C_1C_2}$, remains constant.

$$C_1=(NB_{ij}hP\lambda_0^3\cdot 10^{-8}/2\pi^2c^2 \log_e 10)x \text{ and } C_1C_2=[P\lambda_0^2/2\pi c10^8]^2.$$

The half-breadth of a line with which we are concerned in finding $P/2\pi$ is the fundamental value given by $2\sqrt{C_1C_2}$ and is that for an infinitesimal layer of absorbing medium; the half-breadth measured directly on the photographic plate, on the other hand, is given by a more complex formula and is of small importance, varying tremendously, as the figure shows, with the value of x used. The values of C_1 and C_2 were taken from experimental data for the value $x=10$ cm, and the product C_1C_2 was kept constant while C_1 was varied in the other curves to show the effect, on a given line, of using vapor columns of different length but the same vapor density. From these curves it appears that the best condition for measuring C_1 and C_2 accurately is a short column of vapor for the lower members of the series, and a long column for the higher members, such as was used by Wood and Fortrat[4] in extending the sodium principal series to 58 members.

Values of C_1 and C_2 were found for each line, as in the previous paper,[2] and from these were calculated the true half-breadths in angstroms from the formula $\Delta=2\sqrt{C_1C_2}$, and also the probabilities $P/2\pi$, where $P/2\pi$ is the half-breadth in frequency units divided by two. In Table I is given a typical set of values of C_1 and C_2 and the corresponding values of half-breadth and probability.

TABLE I

Line breadths

Plate 25B4 Vapor pressure 18 mm

Line	Wave-length	C_1	C_2	$\sqrt{C_1C_2}$	Half-breadth	$P/2\pi$
2	3303A	17.90	1.1	4.43	8.86A	12.1×10^{11}
3	2853	2.45	1.2	1.71	3.42	6.3
4	2680	.95	1.3	1.11	2.22	4.65
5	2594	.42	1.5	.794	1.58	3.54
6	2543	.22	1.8	.629	1.25	2.91
7	2512	.153	2.1	.567	1.13	2.70
8	2491	.129	2.4	.557	1.11	2.69
9	2475	.107	2.8	.548	1.09	2.70

In the calculation of A_{ij} and B_{ij} from the measured constants it was possible to average the relative values for all vapor densities and thus get very accurate values by taking the mean of eighteen determinations, once it had been found that the relative probabilities were not a function of vapor density. In this way N, the relative number of absorbing atoms in each case, was found with apparently considerable accuracy. In the case of $P/2\pi$, which depends on N, this was not possible, since the product of C_1 and C_2 is involved, whereas in the preceding case their

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[4] Wood and Fortrat, Astrophys. Jour. **43**, 73 (1916).

ratios were averaged and the curve smoothed out. In Fig. 2 the half-breadths of three lines are plotted against the relative numbers of atoms absorbing them, the latter being determined, as in the previous paper, from the calculations of B_{ij}. Although the points do not lie very exactly on smooth curves, the general nature of the relation between Δ and N is fairly evident, since it seems logical to assume that the curves all pass through the origin. For line 3303 ($n=2$) the curve has the form $\Delta = kN^{.58}$; for line 2853 ($n=3$) the equation is $\Delta = k'N^{.54}$; and for line 2680 ($n=4$) $\Delta = k''N^{.51}$. Whether or not the apparent variation of the exponent with wave-length is real, the similar values of the exponents obtained for the three lines would indicate that Δ may be expected to vary, for a given line, with a power of N close to one-half, if not this exactly. While it is true that both coordinates in these curves were calculated from C_1 and C_2 as determined in the present experiments, one depends on their ratio and the other on their product. Thus one would not expect experimental errors to be masked.

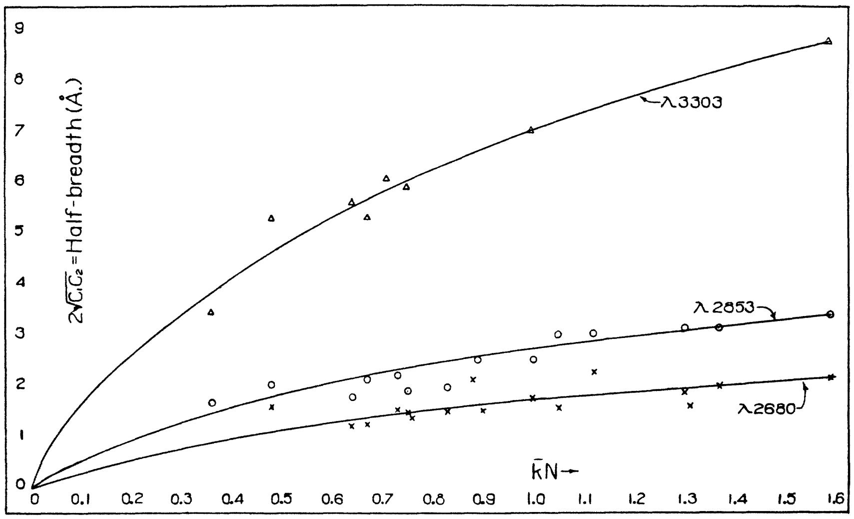

Fig. 2. Curves showing the relation of fundamental half-breadth of lines 2, 3 and 4 to number of absorbing atoms. K is a proportionality constant. The curves are approximately simple parabolas in the region studied, and although they undoubtedly pass through the origin, should not be as drawn at low vapor pressures because of the relatively increased importance of Stark effect or broadening due to collisions of the second kind.

In Table II are given the values of $P/2\pi$ determined for lines 2 to 7 at selected vapor densities. Where several values were obtained at almost the same density they were averaged by means of curves similar to those given in Fig. 2. The half-breadths of the lines in angstroms can be found from the values obtained by means of the formulas given above. Although values of $P/2\pi$ for lines up to the sixteenth were obtained, no great confidence is felt in the accuracy of those above the eighth, because of the difficulty of determining exactly the resolving power of the spectrograph used. If this was greater than the value assumed, it would change the half-breadths measured for the higher members considerably, but would not greatly affect those before the eighth line, beyond making the $P/2\pi$ vs n curve slightly steeper. An error of this sort would not alter greatly the A_{ij} and B_{ij} values given in the previous paper.

TABLE II

Line breadth and vapor density

kN	$P/2\pi \times 10^{-11}$ for lines 2 to 7 2	3	4	5	6	7
.48	6.4	3.9	3.3	3.1	2.8	2.4
.67	7.3	4.0	2.9	2.9	2.8	2.6
.73	8.2	4.2	3.3	2.9	2.6	2.3
.88	9.1	4.8	3.5	3.6	3.2	2.8
1.00	9.6	5.1	3.8	3.3	2.8	2.6
1.37	11.3	5.9	4.2	3.8	3.5	3.1
1.59	12.1	6.3	4.6	3.5	2.9	2.7

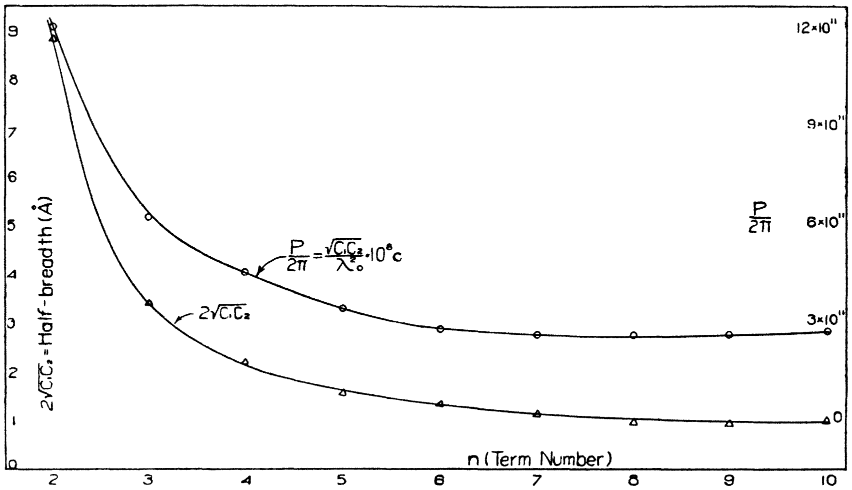

Fig. 3. Curves showing the variation of half-breadth, both in wave-length and frequency units, with term number, for the second to tenth lines of the sodium principal series absorbed by saturated vapor at 590°C.

In Fig. 3 are shown values of the half-breadth in angstroms, and also of $P/2\pi$, plotted against term number for a given vapor density. Curves similar to these were plotted at a number of vapor densities; some were found to exhibit a more pronounced rise in $P/2\pi$ after the sixth term, increasing steadily out to the sixteenth. This may be a spurious effect resulting from incorrect estimate of resolving power, or a real phenomenon due to Stark effect or collision broadening.

Discussion of the Cause of Line Broadening

A most important result is the fact that the half-breadths of lines in the principal series of sodium, and presumably in all the alkali metals, decrease as the term number increases, for the first few members, at least, at the vapor pressures used. This is at variance with experimental results at low pressures and with theoretical attempts to explain the broadening on the basis of collisions or Stark effect. The magnitude of the breadth for the first few lines is extraordinarily great in comparison with the breadth at low pressures. Füchtbauer and Schell,[5] for example, give a value for $P/2\pi$ of only 1.73×10^{11} for the D-lines at a temperature of 174° C, with a pressure of 1733 mm of nitrogen. They found that the broadening varied directly with the pressure of foreign gas, so that at 30 mm this would give a value of $P/2\pi=3\times10^{9}$. We find for the second line of the series $P/2\pi=1.2\times10^{12}$ at a pressure of not over 30 mm and a temperature of 590° C, or 400 times the computed broadening. This is a minimum value, since in our experiments the D-lines were undoubtedly broader than line 3303. The explanation of this difference would seem to be that the present large broadening is bound up with the fact that the pressure here is largely the vapor pressure of sodium itself at high temperature, while in the other experiments the pressure was produced by a foreign gas. This would imply that neighboring sodium atoms or molecules possess a capacity of disturbing other sodium atoms very much greater than that possessed by other kinds of molecules, except possibly those of other alkali metals. This is discussed later.

It should be pointed out that the very great broadening of the lower series lines obtained in this work is not unique, but is apparent in all published photographs of alkali vapor absorption where high vapor pressures were used and where much band absorption was present. Wood's photographs[6] of sodium absorption show a rapid decrease in breadth with increasing term number, as do those of Datta[7] for potassium vapor. While certain photographic phenomena might combine to create a false appearance of decreasing breadth in particular cases, careful

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[5] Füchtbauer and Schell, Phys. Zeits. **14,** 1164 (1913). See also Füchtbauer and Hoffmann, Ann. der Phys. **43,** 96 (1914).

[6] Wood, Phil. Mag. **18,** 530 (1909).

[7] Datta, Proc. Roy. Soc. **101,** 539 (1922).

study shows that this cannot explain away the effect in all, and in the present work, of course, such possibilities are definitely ruled out. At low vapor pressures, on the other hand, it is frequently observed that the breadth of series lines increases rapidly with term number.

The band spectrum of sodium consists of a number of groups or systems, one apparently associated with each of the first few lines of the principal series, rapidly decreasing both in intensity and extent as we go up the series. This strongly suggests that corresponding to each principal series level of the atom there is a system of molecular energy levels, the band about a line arising when the electron in the molecule makes the same transition that the electron in the atom would make for the line. In a situation where dissociation is taking place the molecules are not all in one definite state, but there are a variety of possible states for them, varying from strongly bound molecules to half-formed ones which persist only a short time, and finally to mere pairs of mutually disturbing atoms. The last would form band energy levels only slightly displaced from the atomic energy levels, and a great many such would coalesce to produce the effect of a broadened atomic level. The absorption spectrum would then consist of bands about the lines, with broadened lines about the frequencies of pure atomic transitions. Assuming this to be the cause of the observed broadening we can connect the empirical fact that the bands become much less conspicuous about higher lines of the series with the decrease in broadening in passing up the series. The interpretation of the decrease in intensity in bands is not obvious, but if the bands are due to stable molecules, the broadening to unstable ones, it seems reasonable to suppose that the same factors that affect the magnitude of the bands should also affect the amount of broadening.

An interesting observation in connection with this possible explanation of the present broadening is furnished by the analogous cases of potassium, rubidium, and caesium. There the lines show distinct satellites[8] which seem to be of molecular origin. It may well be that these satellites represent very loosely bound molecules; and that the situation in sodium is similar, but that the satellites, instead of being separate lines, are distributed in such a way as to produce simply a broadened line of regular form.

The necessary condition, both for the large broadening and its decrease with increasing term number, thus becomes the presence of a sufficient number of molecules to produce the broadening, which will be indicated

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[8] See Datta loc. cit.,[7] and rubidium and caesium absorption photographs by Bevan, Proc. Roy. Soc. **83,** 421 (1910) and **85,** 54 (1912).

by the associated band absorption. There may then be three causes of broadening: that connected with bands, that due to Stark effect, and that due to collisions of the second kind, the two latter causes predominating at low vapor pressures, and the first at high vapor pressures.

Holtsmark[9] has treated the observed broadening at low vapor pressures on the basis of the Stark effect from the electric field of neighboring atoms, and one of the authors[10] has suggested a broadening on account of the collisions of the second kind. It seems likely that in a more complete development of quantum theory than we have at present, these two effects will appear as related. Neither is at all capable of explaining the present phenomena observed at high vapor pressures. The Stark effect seems to indicate broadening of the order of that observed by others when the pressure arises from a foreign gas, but there is no apparent reason why a greatly increased action should arise when the external gas is sodium. The effect of collisions of the second kind seems small in the present case; it would be important only if atoms could produce such collisions when they approached within distances of the order of 10^{-6} cm from atoms in the second or third quantum states.

The decrease of breadth with term number is also difficult to interpret on the basis of Stark effect or collision broadening. For the excited states connected with the principal series transitions are states where the valence electron is in a long, eccentric orbit, the length increasing rapidly with term number. The Stark effect would increase with term number, for it depends on the amount of time the electron is out of the intense field of the inner part of the atom and is comparatively free to be influenced by the external field, and this increases rapidly with the longer orbits. Collisions of the second kind might naturally be supposed to increase with term number, the larger orbits being easier to hit than the small ones. These explanations then suggest increase of breadth with term number, which is observed only at low vapor pressures. If we suppose that both these effects and the molecular broadening are operating, however, all the experimental facts are explained.[11]

It should be pointed out that in the previous paper[2] the constants C_1 and C_2 were determined on the basis of a formula for line shapes which

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[9] Holtsmark, Ann. der Phys. **58,** 577 (1919).

[10] Slater, Phys. Rev. **25,** 396 (1925).

[11] Since the above was written, a paper by Born and Franck, Zeits. f. Phys. **31,** 411 (1925) has appeared, in which the authors suggest the existance of unstable or quasi-molecules in a dissociating gas. Their idea is that the broadened bands around absorption lines may be connected with such molecules. Their point of view is similar to that suggested in the present paper.

was found to hold experimentally. Such shapes would be given by Stark effect or collisions of the second kind, but no assumption need be made as to the exact cause of broadening.

Absolute Absorption Probabilities

In the formula for the probabilities of transition from the *i* state to the *j* state with the absorption of radiation, $B_{ij}=(3.31\times10^{37}/Nx\lambda_0)\sqrt{C_1/C_2}$, we can assign absolute experimental values to every factor except N, the number of absorbing atoms, from data already given. To determine N, however, involves the determination of the vapor densities in each case, and, since saturated vapor was always used, of the temperature. In previous similar investigations of line absorption coefficients the experiments have been made at low vapor pressures, only the first few members of the series being studied, and it was possible to enclose the absorbing vapor in a glass or quartz vessel which furnished a definite boundary to the absorbing column. Also the temperature could be determined accurately, the vapor being kept under uniform conditions for long periods of time. In the work here discussed this was not possible, since sodium vapor at the temperatures desired attacks all known substances transparent to ultraviolet light, and it was necessary to use the dynamic equilibrium method common in qualitative studies of alkali vapor absorption, the hot vapor being enclosed in a steel tube provided with water jackets which cooled the ends and condensed the vapor before it reached the windows. The molten pool of sodium in the center of the tube was thus continually providing vapor for distillation into the ends, the rate of distillation being made slow by small-apertured stops at the ends of the hot portion of the tube, and by the presence of a few centimeters pressure of hydrogen. In order to get even an approximate value of the pressure it is necessary to have recourse to indirect methods.

The *relative* number of absorbing atoms in each of the eighteen vapor densities studied was obtained with a fair degree of accuracy in the manner indicated in the previous paper.[2] A fair absolute approximation for N in any given case could thus be made by assigning upper and lower limits to the temperature, determining the corresponding vapor pressures, and balancing the 4.42-fold range of densities between them. It should be emphasized that the relative values of N can be determined with an accuracy entirely independent of the error in the absolute values.

Ladenburg and Minkowski[12] have summarized the available data on the vapor pressure-temperature curve of sodium, and from their curve

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[12] Ladenburg and Minkowski, Zeits. f. Phys. **6**, 153 (1921).

and the relative values of N obtained from absorption measurements in this work the approximate vapor densities were computed. The highest pressures used were not over 30 mm, as measured on a mercury manometer, the cold hydrogen in the manometer and the hot hydrogen and hot sodium in the furnace being practically in mechanical equilibrium. Ladenburg and Minkowski[12] state that in their experiments on the D-lines, the red and yellow absorption bands appeared in a short vapor column at temperatures near 400°C, which checks with previous measurements made on Na vapor by one of the writers. At the lowest density used in this work for which reliable relative values of N were obtained, the bands around the D-lines were very strong, completely wiping out the lines, while the second set of bands around line 3303 was beginning to appear. It was possible, however, to get approximate measurements for a few lines under conditions close to those used by Ladenburg and Minkowski, and by using their data on the vapor pressure-temperature curve, and their observations on the temperature at which the first bands appeared, the probable range in our experiments was set at 4 mm to 18 mm of sodium vapor pressure. We will proceed to calculate N on this basis, and from it B_{ij} and A_{ij}. Using the gas law, we have for the number of molecules per cm^3, $N=2.71\times10^{19}(273/T^0)p/760$. We will assume the vapor to be monatomic, as is usually done, although forced to amend this view later, and taking the case of line 3303 at the highest vapor density, we have $T=863$°K., and $p=18$ mm, so $N=2.02\times10^{17}$. Then since $C_1=17.9$ and $C_2=1.1$, we have $B_{ij}=3.31\times10^{37}\times4.04/2.02\times10^{17}\times10\times3303=2\times10^{16}$. Also $A_{ij}=(5.45\times10^{36}/Nx\lambda_0^4)\sqrt{C_1/C_2}=0.9\times10^5$. This value is for the second line, and the curve connecting A_{ij} and term number was rising so steeply at this point that it was impossible to extrapolate to the D-lines with any accuracy. We may take 30, the value of $(A_{ij})_{5893}/(A_{ij})_{3303}$ computed by Ladenburg from Bevan's data, as a maximum value; we then obtain roughly $(A_{ij})_{5893}=3\times10^6$. If we assume that the a priori probability is proportional to the number of energy levels into which the term is split up in a magnetic field, this would make A_{ij} one-third as great. In general, lines of this sort are found to have an A_{ij} of the order of 10^8. Line 2537 of Hg vapor, on which much work has been done, has a probability of about one-tenth this value, but this is explained on the basis that it is a combination line. We may suppose, therefore, that the value of A_{ij} computed above is perhaps one hundredth of the value to be expected.

On going over the details of the computation and the assumptions made, we find that A_{ij} can be increased by making N or x smaller, or by making C_2 relatively smaller to C_1. The main purpose of the previous investigation[2] was to determine the last two factors, and our arguments rest on the assumption that the values obtained for them are substantially correct. The fact that the relative values of B_{ij} were found to be independent of N is taken to indicate that no large error was made. The length of the absorbing column x was uniformly 10 cm, which was the distance between the stops in the furnace. The column might have been slightly greater in length at higher vapor densities, but it could not have been less, as required to increase A_{ij}.

N could be made less if we assume the temperature to have been taken too high, but a number of things make this highly improbable, and at best it could only account for a factor of about two. The chief cause of the discrepancy may well be the presence of molecular aggregates, which were neglected in the above computation and which would give a new value of N, only a fraction of the molecules being monatomic and contributing to the line absorption. In order to make A_{ij} attain the theoretical value, we should have to assume the presence of from ten to 100 times as many molecules as atoms, with the most probable figure around fifty. That this is not an extravagant assumption we will endeavor to show below.

The Evidence for Molecular Aggregates

Vapor density determinations in the alkali metals are extremely difficult, and have generally been carried out at much lower temperatures than those used here. There is considerable disagreement between the results of different observers, especially at high temperatures, when different methods were used. The presence of band absorption can be taken as quite certain indication of aggregates of atoms into molecules, and Smith[13] has recently analyzed some of the visible bands in Na and finds moments of inertia which seem to indicate the presence of Na_2. We accordingly have here a further justification in assuming the presence of a certain number of Na_2 molecules in the present case, although at temperatures below 400°C, where the vapor pressure is about 0.3 mm, the number of molecules is undoubtedly smaller than the number of Na atoms. The effect of rising temperature is to increase dissociation, but where the vapor is saturated, as in this work, the pressure rises faster than the temperature, tending to reverse the effect.

If we were to apply conventional thermodynamics, we should have the equilibrium constant, or the ratio of the square of atomic concentra-

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[13] Smith. Proc. Roy. Soc. **106**, 400 (1924).

tion to molecular concentration, equal to $aT\ /pe^{-q_0/RT}$, where a is a constant, β equals 3/2 if the specific heat of the molecules is $(5/2)R$, and q_0 is the energy required to dissociate a molecule at absolute zero. With increasing T and p, the factor T^β increases, as does also $e^{-q_0/RT}$, if q_0 is positive, but p, the vapor pressure, increases so rapidly that the whole quantity would decrease, for reasonable choice of constants, meaning an association into molecules. It sometimes happens, however, that q_0 is negative, in which case we find that the formula permits even so large a change as from practically all atoms at 400°C to mostly molecules at 600°C. Also, if there are many partly formed molecules, these would have energy in their internal degrees of freedom, and hence the specific heat of the molecules would be greater than $(5/2)R$, and β would be reduced. While some exception might be taken to the use of the above formula in the present case, it seems to be true that such formulas work better than would be expected from the assumptions made in their derivation. It is interesting to note that the chemical constants of sodium and potassium, as determined by Ladenburg and Minkowski,[14] show discrepancies from the theoretical values larger than for any other monatomic vapor, giving the impression that they may depart appreciably from being monatomic even in the region where measurements have been made on them with presumably accurate results.

Approximate photometric measurements on the bands associated with line 3303 indicate about fifty times as much total intensity of band absorption as of line absorption at 600°C. Then, even if we assume all the absorption in the broadened line to have been due to atoms, if the molecular and atomic absorption coefficients are of the same order of magnitude we have definite indication of many more fairly stable molecules than of atoms. There seems few data available to test the justification of such an assumption.

Finally, the intensity of band absorption was measured at various vapor pressures, and a function of this which should be proportional to the number of absorbing molecules was plotted against the previously determined relative numbers of atoms absorbing at the corresponding vapor pressures, the latter being plotted as abscissas. These curves showed a gradually increasing slope as the vapor pressure increased, indicating that the number of molecules was increasing faster than the number of atoms.

We may conclude, then, that the presence of bands indicates with certainty that the alkali metal vapors are not entirely monatomic when

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[14] Ladenburg and Minkowski, Zeits. f. Phys. **8**, 137 (1921).

saturated above 400°C, and that there is good evidence from the relative changes in band and line absorption that the number of molecules increases faster than the number of atoms with increasing temperature. If we assume the molecular absorption coefficient to be of the same order of magnitude as that of the atom, there must be more molecules than atoms at 600°C. In case there are many more molecules than atoms at high temperatures, the low values for A_{ij} found in the present work are explained; that the spectroscopic data on which these values are based are trustworthy is indicated by the fact that the ratios of the A_{ij}'s are found independent of N. Although it seems a rather forced assumption, and we make it only tentatively, it seems worth considering.

We wish to acknowledge the kindness of Professor E. C. Kemble in giving this paper the benefit of his criticism.

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Harvard University,
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