

Excitation of Molecular Rotation by Slow Electrons. II*

S. GELTMAN

Joint Institute for Laboratory Astrophysics,† Boulder, Colorado

AND

K. TAKAYANAGI

Department of Physics, Saitama University, Urawa, Japan

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A previous study is here extended to consider the effects on rotational excitation of the nonvanishing electronic spin of O_2 and the short-range static fields of the H_2 , N_2 , and O_2 molecules. The spin effect is found to be negligible, while the effect of short-range interactions can be quite large in certain energy regions. This permits a qualitative explanation of the heretofore unexplained ratio of the observed electron relaxation rates in oxygen and nitrogen.

I. INTRODUCTION

ROTATIONAL excitation of homonuclear diatomic molecules by slow electrons has been extensively studied in the past. The present work is a continuation of our previous work¹ which will be designated as I. References to previous theoretical work can be found in I, and more recent work is referred to below.²⁻⁴

In I a distorted-wave calculation was done for the rotational excitation of H_2 and N_2 . There the distorted waves were based upon the spherical part of the polarization potential and the transition-inducing interaction was taken to be the sum of the quadrupole and nonspherical part of the polarization potentials. To make these potentials more realistic at small distances, they were assumed to be constant for $r < R$, where R was chosen to give the best result for the elastic-scattering cross sections.

A Born calculation⁴ for O_2 , in which only the long-range parts of the nonspherical interaction are retained, yields a result which is smaller than is obtained in a corresponding calculation for N_2 by more than a factor of 10 in the electron energy range $k^2 < 10^{-2}$ which roughly corresponds to electron temperatures $T < 10^3$ °K. These relative results are not changed appreciably in the distorted-wave approximation (paper I and additional calculations to be discussed later). However, a recent experimental study by Mentzoni and Rao⁵ indicates that the rotational-excitation cross section of O_2 is actually considerably large than that for N_2 for 300 °K $< T < 1000$ °K. This is contrary to the results of the above-described calculations.

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† Of the National Bureau of Standards and the University of Colorado.

¹ K. Takayanagi and S. Geltman, *Phys. Rev.* **138**, A1003 (1965).

² A. Dalgarno and R. J. W. Henry, *Proc. Phys. Soc. (London)* **85**, 679 (1965).

³ D. H. Sampson and R. C. Mjolsness, *Phys. Rev.* **140**, A1466 (1965).

⁴ K. Takayanagi, *Rept. Ionos. Space Res. Japan* **19**, 1 (1965).

⁵ M. H. Mentzoni and K. V. Narasinga Rao, *Phys. Rev. Letters* **14**, 779 (1965).

The purpose of the present work is to study the following two effects, which have been neglected in previous calculations on N_2 and O_2 :

(a) the coupling of the electronic spin of O_2 in the $^3\Sigma_g^-$ state with its rotational motion (this does not arise in H_2 and N_2 , since they are in $^1\Sigma$ states), and

(b) the contribution to the nonspherical part of the total potential made by the electron's short-range interaction with the static fields of the atomic cores. The effect of this short-range interaction disappears near threshold but becomes important at some higher energy whose value depends upon the magnitudes of the long-range interaction.

It will be seen that the spin-coupling (a) has a minor effect, while the short-range interaction (b) can have a great effect on the rotational excitation cross sections in the energy range of interest.

II. EFFECT OF NONVANISHING SPIN

The electronic ground state of O_2 is a triplet state. The total angular momentum \mathbf{J} of the molecule is given by the sum of two operators

$$\mathbf{J} = \mathbf{K} + \mathbf{S}, \quad (1)$$

where \mathbf{K} is the angular momentum of rotation of the molecular framework and \mathbf{S} the spin angular momentum. For oxygen in the $^3\Sigma_g^-$ state, there are good quantum numbers K and S , such that the operators \mathbf{K}^2 and \mathbf{S}^2 have the eigenvalues

$$K(K+1) \quad \text{and} \quad S(S+1), \quad (2)$$

with

$$K = 1, 3, 5, \dots; \quad S = 1. \quad (3)$$

The quantum number K is odd in order that the Pauli principle be satisfied for nuclei of zero spin. The coupling between \mathbf{K} and \mathbf{S} results in the splitting of each rotational energy level into three sublevels⁶ corresponding to the total-angular-momentum quantum numbers $J = K, K+1$, and $K-1$.

⁶ G. Herzberg, *Spectra of Diatomic Molecules* (D. Van Nostrand, Inc., Princeton, New Jersey 1950), pp. 223-224.

The coupled wave function is given by

$$\phi_{J\mu}^{K} = \sum_m Y(K \mu - m | \hat{s}) S(m) \times (K 1 \mu - m m | K 1 J \mu), \quad (4)$$

where $(j j' m m' | j j' J m + m')$ is the Clebsch-Gordan coefficient,⁷ $S(m)$ the normalized spin function corresponding to the state in which the z component of spin is $m\hbar$, μ the z component of J , $Y(j m | \hat{s})$ the normalized spherical harmonic function, and \hat{s} the unit vector along the molecular axis.

We now apply the first order Born approximation to see the effect of this nonvanishing spin on the rotational transitions. As long as we neglect the short-range part, the electron-molecule interaction depends negligibly on electron spin, so that the calculation can be done fairly easily. The effective cross section for the rotational transition $K, J \rightarrow K', J'$ is given by

$$\sigma(K, J \rightarrow K', J') = \frac{1}{2J+1} \frac{k'}{k} \int d\hat{k} \sum_{\mu} \sum_{\mu'} \left| \frac{1}{4\pi} \int \exp(-i\mathbf{q} \cdot \mathbf{r}) \times (K', J', \mu' | 2V_{ns} | K, J, \mu) d\mathbf{r} \right|^2, \quad (5)$$

where \mathbf{k} and \mathbf{k}' are the initial and the final wave vectors of the scattered electron, $\mathbf{q} = \mathbf{k}' - \mathbf{k}$, \mathbf{r} is the position of the electron relative to the center of the molecule, the V_{ns} is the nonspherical interaction between the molecule and the electron. Here and in the following, atomic units (a.u.) are used unless otherwise stated. In this section, we shall take the following form of the nonspherical interaction which has been used by Dalgarno and Moffett⁸ for electron- N_2 collisions:

$$V_{ns} = -(\alpha'/2r^4 + Q/r^3)P_2(\hat{s} \cdot \hat{r}). \quad (6)$$

For the rotational excitation, $J, K \rightarrow J', K+2$, (5) becomes

$$8\pi \frac{1}{2J+1} \frac{k'}{k} \left[\frac{2}{9} Q^2 + \frac{Q\alpha'\pi}{72} \left(4k - \frac{\Delta k^2}{k} \right) + \frac{\alpha'^2\pi^2}{512} (2k^2 - \Delta k^2) \right] \times \sum_{\mu} \left\{ \sum_m c^2(K \mu - m; K' \mu - m) (K' 1 \mu - m m | K' 1 J' \mu) \times (K 1 \mu - m m | K 1 J \mu) \right\}^2, \quad (7)$$

where

$$c^L(jm; j'm') = \left(\frac{4\pi}{2L+1} \right)^{1/2} \int Y^*(jm | \hat{r}) \times Y(1m - m' | \hat{r}) Y(j'm' | \hat{r}) d\hat{r}, \quad (8)$$

and $\Delta k^2 = k^2 - k'^2$. From this cross section, the reduced cross section is obtained by

$$\sigma(K \rightarrow K') = \sum_J \sum_{J'} w_{JK} \sigma(K, J \rightarrow K', J'), \quad (9)$$

⁷ Notation of E. U. Condon and G. H. Shortley, *Theory of Atomic Spectra* (Cambridge University Press, New York, 1935).

⁸ A. Dalgarno and R. J. Moffett, Proc. Nat. Acad. Sci., India A33, 511 (1963).

with

$$w_{JK} = (2J+1)/3(2K+1).$$

If the level splitting due to $\mathbf{S}-\mathbf{K}$ coupling is neglected, the energy difference $\Delta k^2/2$ does not depend on J and J' . Then it is easily shown that $\sigma(K \rightarrow K')$ becomes the Dalgarno-Moffett formula, which we shall call $\sigma_0(K \rightarrow K')$.

Using $Q = -0.30$ a.u. (Ref. 9) and $\alpha' = 5.00$ a.u. (Ref. 10), the effective cross section has been calculated for $K=1, K'=3$. The result is shown in Table I. The reduced cross section $\sigma(1 \rightarrow 3)$ calculated from Table I

TABLE I. Effective cross section $\sigma(K, J \rightarrow K', J')$ in the Born approximation with $K=1, K'=3$ [in units of $10^{-2}a_0^2$].

J	J'	$k^2=0.0006$	0.002	0.006	0.02	0.04
0	2	3.792	3.008	1.655	0.676	1.857
1	2	1.307	1.009	0.552	0.226	0.620
1	3	2.569	2.011	1.104	0.451	1.239
2	2	0.110	0.086	0.047	0.019	0.053
2	3	0.758	0.602	0.331	0.135	0.372
2	4	2.974	2.328	1.278	0.522	1.434

with the use of the observed spin splittings¹¹ proves to be very close to $\sigma_0(1 \rightarrow 3)$, the difference being less than 0.2% for $k^2 \geq 0.0006$. To estimate the rate of energy loss of electrons in oxygen gas, which is the measured quantity in the experiment in Ref. 5, we have to calculate the quantity

$$\sum_J \sum_{J'} w_{JK} \Delta E(K, J \rightarrow K', J') \sigma(K, J \rightarrow K', J'), \quad (10)$$

where $\Delta E(K, J \rightarrow K', J')$ is the rotational-energy change in the transition $K, J \rightarrow K', J'$.¹² For $K=1, K'=3$, it has been found that the quantity (10) is almost the same as the corresponding quantity

$$\Delta E(K=1 \rightarrow K'=3) \sigma_0(K=1 \rightarrow K'=3),$$

which one obtains by neglecting spin. For higher K and K' values, the effect will be even smaller.

It is true that we now have a new process of the type $K=K', J \neq J'$. In the energy region of interest, however, we can assume that $\Delta k^2 \ll k^2$ for this type of inelastic process. Thus the cross section (7) becomes symmetric with respect to J and J' except for the factor $1/(2J+1)$. Therefore, $w_{JK} \sigma(K, J \rightarrow K, J')$ is symmetric, while $\Delta E(K, J \rightarrow K, J')$ is antisymmetric. Thus the quantity (10) vanishes to a fairly good approximation.

We have not made the distorted wave calculation with spin coupling, but it is unlikely that such a calculation would lead us to a very different conclusion. On

⁹ A. D. Buckingham (private communication).

¹⁰ N. J. Bridge and A. D. Buckingham, J. Chem. Phys. 40, 2733 (1964).

¹¹ H. D. Babcock and L. Herzberg, Astrophys. J. 108, 167 (1948).

¹² We have neglected the Boltzmann factor and thus are assuming that the level splitting is negligible compared with kT .

the basis of the above discussion we neglect the presence of spin in O₂ and treat its rotational excitation on the same basis as the singlet-state molecules.

III. EFFECT OF THE SHORT-RANGE INTERACTION

From symmetry considerations, it is easy to see that the nonspherical part of the interaction should vanish at the midpoint of the molecule ($r=0$). Thus we slightly modify the nonspherical long-range interaction used in I, so that it becomes

$$V^{LR}(r)P_2(\xi \cdot \hat{r}) = - (r/R)^2(Q/R^3 + \alpha'/2R^4)P_2(\xi \cdot \hat{r}), \quad r \leq R \\ = - (Q/r^3 + \alpha'/2r^4)P_2(\xi \cdot \hat{r}), \quad r > R. \quad (11)$$

For the spherical part of the interaction we adopt, as in I,

$$V_s(r) = -\alpha/2R^4, \quad r \leq R \quad (12) \\ = -\alpha/2r^4, \quad r > R.$$

Here α is the spherical part of the polarizability of the molecule. The distorted waves are solutions of the radial Schrödinger equation containing this spherical potential. The cutoff distance R was chosen by comparing the elastic scattering cross section calculated from (12) with the existing experimental data.^{12a} These curves for oxygen are shown in Fig. 1. Experimental data are those of Ramsauer and Kollath,¹³ of Brüche,¹⁴ and of Aberth *et al.*¹⁵

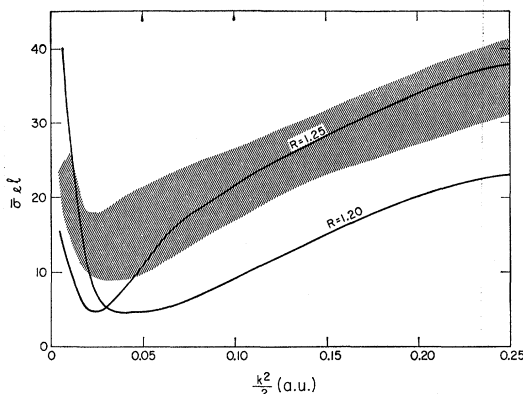


FIG. 1. Average elastic-scattering cross-section curves for O₂ for the two cutoff radii which give best agreement with experiment. The cross hatching corresponds to the region of the experimental data contained in Refs. 13, 14, and 15. (a.u. ≡ atomic units.)

^{12a} Note added in proof. We are assuming this is a reasonable procedure for determining the elastic scattering, even in the presence of the strong short-range nonspherical interaction that is introduced in this paper. The values of R which we obtain for H₂, N₂, and O₂ are reasonable in relation to the internuclear separations of these molecules.

¹³ C. Ramsauer and R. Kollath, *Ann. Physik* 4, 91 (1930).

¹⁴ E. Brüche, *Ann. Physik* 83, 1065 (1927).

¹⁵ W. Aberth, G. Sunshine, and B. Bederson, *Atomic Collision Processes*, edited by M. R. C. McDowell (North-Holland Publishing Company, Amsterdam 1964), p. 53.

We found that $R=1.25$ a.u. provides about the best fit with the observations we can obtain. The distorted-wave calculation was made with this cutoff distance, and $\alpha=10.8$ a.u. The result is shown in Fig. 2. The corresponding result for nitrogen is also shown for comparison. Note that effect of the modification in V_{ns} [Eq. (11) compared with nonspherical part of (5) in I] is negligible. For this molecule, $R=1.75$, $\alpha=12.0$, $\alpha'=4.2$, and $Q=-1.10$ were adopted (as in I). As is seen in the figure, the oxygen cross section is much less than the nitrogen cross section for $k^2 < 10^{-2}$ in accordance with the discussion in the Introduction.

Since the long-range interaction (at least the quadrupole part) is relatively weak in oxygen, the short-range part of the interaction may play an important role. For the short-range potential, we adopt simply the sum of the electrostatic potentials of isolated oxygen atoms,

$$V^{SR}(\mathbf{r}) = v(\mathbf{r}_1) + v(\mathbf{r}_2), \quad (13)$$

\mathbf{r}_1 and \mathbf{r}_2 being the radius vectors of the incident electron from the nuclei. The electronic charge distribution calculated by Herman and Skillman¹⁶ for an oxygen atom is approximately represented by

$$4\pi r^2 \rho \cong 30.75 \exp(-1.71r) \quad (14)$$

for $1 < r < 6$. From this, the electrostatic field was calculated by

$$v(r) = -\frac{1}{r} \int_r^\infty 4\pi r'^2 \rho dr' + \int_r^\infty \frac{-4\pi r'^2 \rho dr'}{r} \\ \cong - (2.52/r) \exp(-1.71r) \\ - (17.98/r) \exp(-3.42r), \quad (15)$$

which is probably a good approximation for $r > 1$, but is a considerable overestimate for smaller r . The Born approximation has been applied first using (13) (without including the long range interactions). The calculation is similar to that of Carson,¹⁷ so we shall not repeat the details. The resulting cross section for $K=1 \rightarrow K'=3$ is very small for low-energy collisions, as we expect from the short-range nature of the interaction and also from the empirical fact that the main contribution to the cross section comes from the incident p wave.¹ For $k^2 \gtrsim 10^{-2}$, however, the cross section becomes much larger than the corresponding Born cross section with the long-range interactions only.

Thus we proceeded to do the distorted wave calculation with both short-range as well as long-range interactions. For this, we expanded the short-range interaction (13) in terms of Legendre polynomials as

$$V^{SR}(\mathbf{r}) = \sum_{n(\text{even})} V_n^{SR}(r) P_n(\xi \cdot \hat{r}). \quad (16)$$

¹⁶ F. Herman and S. Skillman, *Atomic Structure Calculations* (Prentice-Hall, Inc., Englewood Cliffs, New Jersey, 1963).

¹⁷ T. R. Carson, *Proc. Phys. Soc. (London)* A67, 909 (1954).

The P_0 term is implicitly included in (12), since we determine the cutoff distance R by fitting the elastic cross section to the observed data. The spherical part (P_0 term) and the nonspherical part (P_2 term) of the total assumed interaction are shown in Fig. 3. It is seen that the short range interaction is dominant for $r < 2.5$. The contribution from the higher order terms of (16) has been neglected, as it has been shown by Dalgarno and Henry² that $K \rightarrow K+4$ cross sections are negligible compared with those for $K \rightarrow K+2$. For

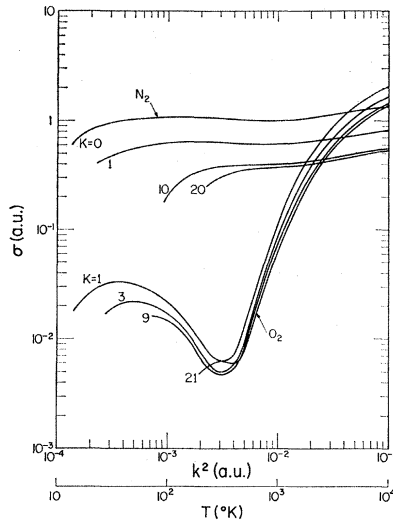


FIG. 2. Distorted-wave cross sections for the rotational excitation $K \rightarrow K+2$ when only the long-range part [Eq. (11)] of the nonspherical interaction is included. This is a plot of the cross section versus electron energy ($k^2=1$ corresponds to 13.6 eV). The lower scale is for convenience in estimating the temperature, where $\frac{3}{2}\kappa T \cong$ electron energy (κ =Boltzmann's constant).

$V_2^{SR}(r)$, we have used the approximate analytic function,

$$V_2^{SR}(r) = -A \exp[-B|r-s/2|]. \quad (17)$$

For oxygen, $A=20$, $B=3.8$, and $s/2=1.13$ are adopted. The distorted wave calculation is straightforwardly done as in I. The resulting cross sections are shown in Figs. 4 and 5. In Fig. 4, the great difference between the cross section obtained by neglecting the short-range force ($A=0$) and between those obtained by including the short-range force ($A=10$ and 20) should be noticed. Analysis of the calculated cross section again shows that the main contribution comes from the combination of incident and final p waves. Thus the cross section should not exceed $\pi(2l+1)/k^2$ with $l=1$. For higher incident energy, however, the calculated cross section with the short-range force becomes so large that it considerably violates this probability conservation theorem. Therefore, the actual cross section should be lower than the calculated values. The fact that the experimental total cross section of oxygen is less than 40 a.u. for $k^2=0.1$ to 1, while it is less than

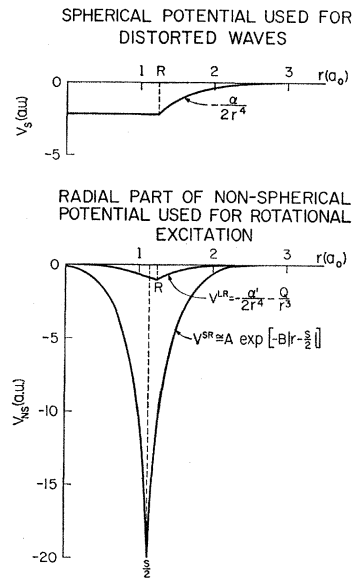


FIG. 3. Radial parts of the spherical and non-spherical (coefficient of $P_2(\hat{s} \cdot \hat{r})$ only) potentials used for the present calculations. The numerical values above apply to O_2 .

20 a.u. around $k^2=0.01$, also provides an upper bound to the rotational transition cross section. In addition to these, the short-range force we have adopted is too large in the vicinity of nuclei. Also, in the actual oxygen molecule, the short-range force might be somewhat more spherical than the simple sum of the two atomic fields

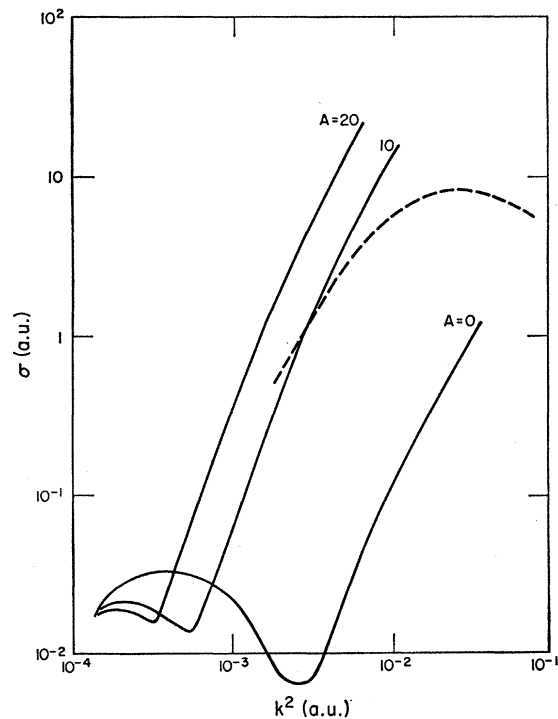
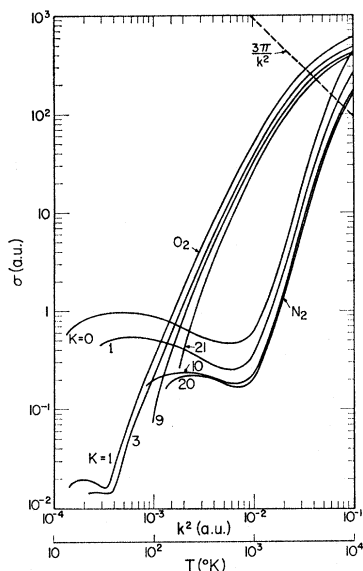


FIG. 4. Distorted-wave cross sections for the $K=1 \rightarrow 3$ rotational excitation of O_2 for different values of the short-range field parameter, A . The dashed curve is a guess of where the true cross section may lie, such that it goes into the $A=20$ curve at low energies and is well below the conservation limit at higher energies.

FIG. 5. Distorted-wave cross sections for the rotational excitation $K \rightarrow K+2$ when both the long-range and short-range parts of the nonspherical interaction are included. The k^2 and T scales are as explained in Fig. 2.



because of the chemical binding. Taking the above factors into account we suggest, rather arbitrarily, that the correct cross section would be near the dashed curve in Fig. 4. This curve is comparable with the cross section¹⁸ required to explain the experimental data by Mentzoni and Rao.⁵ A close-coupling treatment of the problem (or some other method in which the unitarity of the S matrix is preserved) is necessary to confirm this suggested cross section. In Fig. 5, the corresponding cross section for nitrogen is also shown. Here, we have assumed (17) for the short-range force with $A=17.5$,

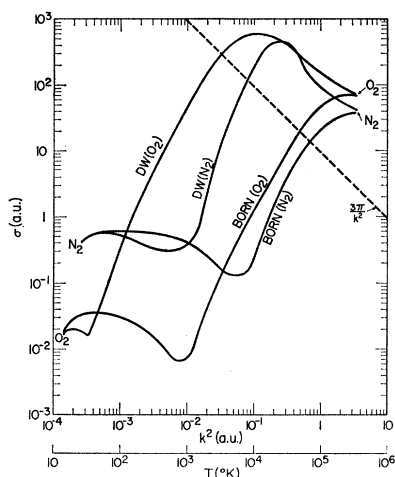


FIG. 6. Born and distorted-wave (DW) cross sections for the rotational excitation $K=1 \rightarrow 3$ when both the long-range and short-range parts of the nonspherical interaction are included. The k^2 and T scales are as explained in Fig. 2.

¹⁸ As long as the incident electron energy is far above the threshold, the stopping cross section $\Delta E(K \rightarrow K+2)\sigma(K \rightarrow K+2) - \Delta E(K \rightarrow K-2)\sigma(K \rightarrow K-2)$ is almost independent of K . Thus $\Delta E(1 \rightarrow 3)\sigma(1 \rightarrow 3)$ gives us a reasonably accurate measure of the energy loss rate.

which simply comes from the value adopted for oxygen multiplied by the ratio of the nuclear charges, and we have also taken $s/2=1.03$ for half the internuclear distance. It is seen that the nitrogen cross sections do not change very much from their values in Fig. 2 when the short-range force is included, while the oxygen cross sections change drastically and are now much larger than those of nitrogen in the energy region of interest, i.e., corresponding to electron temperatures of 300 to 1000°K. This is consistent with the findings of Mentzoni and Rao. One might think that the main difference between the oxygen and the nitrogen cross sections in Fig. 5 comes from the different cutoff distances R adopted in the model potentials for these two molecules. So, we have repeated the calculation for nitrogen with $R=1.25$. The resulting cross section is still less than the oxygen cross section in the energy region of interest.

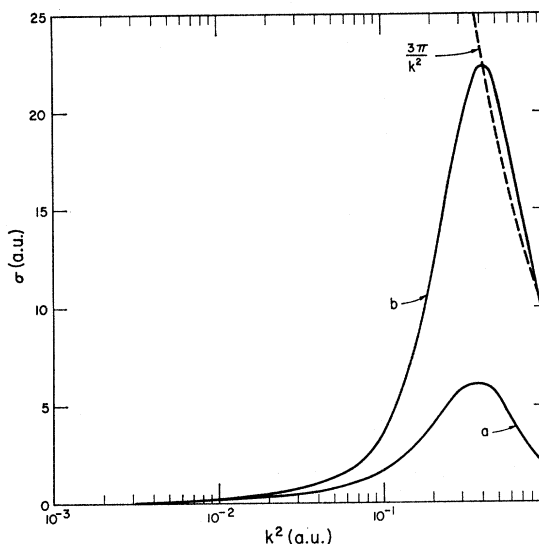


FIG. 7. Distorted-wave cross sections for the rotational excitation $K=0 \rightarrow 2$ in H_2 when (a) only long-range, and (b) both long-range and short-range parts of the nonspherical interaction are included.

In Fig. 6, a comparison of the distorted-wave and the Born calculations for oxygen and nitrogen is made. The importance of the distortion effect is clearly seen. The effect of the distorted waves is to lower the energy at which the sharp rise (due to the short-range force) takes place.

The effect of the short-range potential on the rotational excitation of hydrogen is shown in Fig. 7. The increase in cross section here is very small and occurs at higher energies than in the case of O_2 and N_2 . Also the violation of the flux-conservation theorem is relatively mild for the case of H_2 . These are consequences of the low nuclear charge and of smaller internuclear separation in H_2 than in the heavier molecules. The short-range static field used for H_2 was

derived from

$$v(r) = -e^{-2r}(1+1/r) \quad (18)$$

which led to the approximate analytic form (17) with $A=2.6$, $B=3.5$ and $s/2=0.70$. Form (18) again ignores the effect of chemical binding, which is consistent with our use of atomic fields for oxygen and nitrogen.

IV. DISCUSSION

To explain the results obtained in the last section, it is instructive to look carefully at the interaction potential as a function of r . Both nitrogen and oxygen have negative quadrupole moments Q . For large r , therefore, the radial part of $V_{ns}(r)$ is positive. As r decreases, the nonspherical part of the polarization interaction becomes relatively more important. Since this has the opposite sign to the quadrupole interaction, there is a cancellation. At a certain distance the potential has a zero and becomes negative at smaller r . Finally, as r becomes less than about 2.5 a.u., the rapid increase of the short-range force makes the potential well very deep. The p waves provide the main contribution to the cross section under consideration. (From the nature of the transition, an incident s wave is always associated with the outgoing d wave, or vice versa. The d wave cannot penetrate deeply into the inner region for a low-energy collision, so that the incident p wave becomes more important than the s wave.) When the incident energy is very low, the classical turning point r_c for the p wave is given by

$$kr_c \cong \sqrt{2},$$

and r_c is very large. Thus only the quadrupole interaction contributes to the rotational excitation.

As k increases, the p wave penetrates into the region where the mutual cancellation of the long-range interactions takes place. The effect of this cancellation is to yield a minimum in the calculated cross sections. The nonspherical part of the short-range potential has the same sign as the nonspherical polarization interaction, so that the introduction of the short-range force will shift the zero of the potential $V_{ns}(r)$ outwards. The

cross-section minimum is thus shifted to a lower energy as the strength of the short-range interaction is increased. This behavior can be seen in Fig. 4.

When the incident energy is further increased, the p wave will more strongly overlap the main part of the short-range force. The distortion of the electron wave function by the polarization force makes the penetration of the p wave into the inner region easier. As we have already noticed in I, we obtain a peak in the cross section when the incident energy is such that there is maximum overlap between the first loop of the distorted p wave and the inner part of the nonspherical interaction.

Examination of the numerical values of the total $V_{ns}(r)$ (long- and short-range contribution) shows that the zeros occur at $r \cong 2.5a_0$ for nitrogen and $r \cong 8.5a_0$ for oxygen. This large difference is due primarily to the factor of four difference in the Q values for the two molecules. From the above discussion it is quite understandable why the positions of minimum and maximum for oxygen occur for lower incident energies than for which they occur in nitrogen (Fig. 6). In Born approximation the energies at the minima differ by a factor of 10, which is consistent with the ratio $(8.5/2.5)^2$. It is difficult to estimate *a priori* the positions of minimum cross section in the distorted wave approximation, but the results of Fig. 6 show that they are even more widely separated than in Born approximation.

Thus we may summarize by saying that the distorted-wave curves for oxygen and nitrogen in Fig. 5 are in qualitative agreement with the swarm experiments of Mentzoni and Rao in their temperature range of 300 to 1000°K. It is interesting to note that for $T < 100^\circ\text{K}$ the oxygen cross sections become much lower than those for nitrogen. Experiments in that temperature range would be extremely useful in checking the present theory.

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