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Angular Distribution of O⁻ from Dissociative Electron Attachment to O₂[†]

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The angular distribution of O⁻ produced by electron bombardment of O₂ has been measured in the electron energy range 5.75–8.40 eV. The results show a strong energy dependence and are consistent with the theory of O'Malley and Taylor if the final O₂⁻ repulsive resonance state is assumed to have the symmetry ²Π_u, and if only the first two allowed partial waves of the incident electron corresponding to L=1 and L=3 contribute. The results indicate that the L=3 term becomes more important as energy increases and thereby demonstrate that the single-term approximation for the angular distribution does not apply for this process.

I. INTRODUCTION

Measurements of the cross section as a function of electron energy for the process



have yielded a simple structureless bell-shaped curve indicative of dissociation from a single resonant repulsive molecular negative-ion state.¹⁻⁵ Figure 1 shows the results of cross-section measurements made at room temperature (300 °K) by Schulz³ and by Rapp and Briglia.⁵ Recent observations^{6,7} of the temperature dependence of this process agree quite well with a semiempirical calculation⁸ which assumes that it proceeds via a single repulsive resonance state. A slight discrepancy between the calculated and observed results at the

high-energy tail of the cross-section curve at all temperatures, however, suggests that more than one O₂⁻ repulsive state might contribute to dissociative attachment. From his calculation, O'Malley suggested that the O₂⁻ state responsible for dissociative attachment is probably the ²Π_u state proposed by Gilmore.⁹ Recent calculations of the O₂⁻ states provide additional support for this assignment.¹⁰ It has been suggested, however, that the symmetry of the O₂⁻ state might also be ²Σ_g⁺, ⁴Σ_w⁻, ²Σ_w⁻, ⁴Π_g, ⁴Π_w, or ²Δ_u.⁹⁻¹²

Using simple-symmetry arguments, Dunn¹³ showed that the differential cross section for production of negative ions by dissociative attachment should have an angular dependence determined by the symmetries of the initial and final molecular states. The selection rules derived by Dunn can be used to

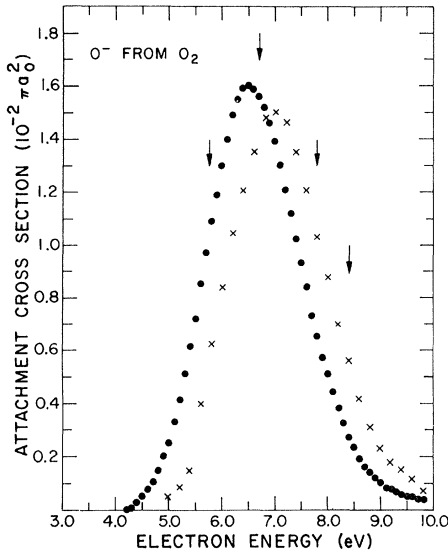


FIG. 1. Measured cross section for dissociative attachment of oxygen. The dots represent data from Ref. 5, and the x's represent data from Ref. 3. The arrows indicate the electron energies at which the present angular distribution measurements were made.

predict whether or not the cross section will vanish when the molecular axis is oriented in a direction either parallel or perpendicular to the incident electron beam. From these arguments it can be shown that a transition from the ground $^3\Sigma_g^-$ state of O_2 to a $^2\Pi_u$ state of O_2^- gives a differential cross section for dissociative attachment which vanishes along the direction of the incident electron beam. For a final Σ_u^- state, the cross section should vanish along the direction perpendicular to the beam, whereas for final Σ_g^+ , Π_g , and Δ_u states, the cross section should vanish in both directions, i. e., along and perpendicular to the beam. Since there are no outgoing electrons, the symmetry axis which defines the angular distribution of the dissociation products for dissociative attachment lies along the direction of the incident electron. Dunn's predictions apply if the effects of molecular thermal and rotational motion can be neglected. These effects result in a blurring of the angular distribution but they are important only if the kinetic energy of the dissociation products is comparable to the mean thermal and rotational energy.¹³⁻¹⁶ For oxygen at room temperature the mean thermal and rotational energy is on the order of 0.02 eV, whereas the observed kinetic energy of O^- corresponding to the data of Fig. 1 is greater than 1.0 eV for all electron energies.^{3,17} Assuming a $^2\Pi_u$ state, O'Malley and Taylor¹⁸ predicted from a single-term approximation that O^- formation should have a $\sin^2\theta$ dependence with respect to the incident electron's direction.

In the present work, a measurement of the angular distribution of O^- was performed in order to obtain information about the number and symmetries of the repulsive electronic resonance states of O_2^- involved in dissociative attachment. Prior to this work, there have been no reported attempts to experimentally identify these states.

II. THEORY

According to the theory of O'Malley and Taylor,¹⁸ the angular dependence for dissociative attachment has the general form

$$I(k, \theta, \phi) \sim \left| \sum_{L=\mu}^{\infty} a_{L,|\mu|}(k) Y_{L\mu}(\theta, \phi) \right|^2, \quad (2)$$

where the $Y_{L\mu}(\theta, \phi)$'s are spherical harmonics, and the $a_{L,|\mu|}(k)$'s are expansion coefficients which depend on the incident electron energy $E_e = k^2\hbar^2/2m_e$. Equation (2) is a valid expression provided either the conditions $\frac{1}{2}\Gamma \ll 2\mu R_0^2/\hbar^2 J(J+1)$ or $E_0 \gg \hbar^2 J \times (J+1)/2\mu R_0^2$ is satisfied, where J is the molecular rotational quantum number, μ is the reduced mass of the molecule, R_0 is the internuclear separation, Γ is the width of the resonance, and E_0 is the kinetic energy of the fragments which, for a homonuclear diatomic molecule, is given by

$$E_0 = \frac{1}{2} [E_e - (D - A)] . \quad (3)$$

Here D is the dissociation energy of the molecule, and A is the electron affinity of the resulting negative ion. These conditions essentially require that either the lifetime of the resonance be short compared to the molecular rotation period or the kinetic energy of the fragments be large compared to the mean rotational energy.

The partial-wave expansion of the incident electron plane wave can be written as

$$e^{i\vec{k}\cdot\vec{r}} = 4\pi \sum_{L=0}^{\infty} \sum_{\mu=-L}^L (i)^L Y_{L\mu}(\theta, \phi) Y_{L-\mu}(\theta_e, \phi_e) j_L(kr_e), \quad (4)$$

where θ, ϕ give the orientation of the molecular axis with respect to \vec{k} and θ_e, ϕ_e, r_e give the electron's position relative to the molecular axis \vec{R} . When Eq. (4) is inserted in the electronic matrix element which determines the scattering amplitude [Eq. (2) of O'Malley and Taylor], then certain restrictions occur on the summation indices μ and L appearing in Eq. (2). From conservation of axial orbital angular momentum, the lowest allowed value of μ is given by

$$\mu = \Lambda_f - |\Lambda_i|, \quad (5)$$

where Λ_i and Λ_f are the projections of orbital angular momentum along the internuclear axis for the initial and final molecular states. In Eq. (2) the summation is restricted to even or odd values of L , depending on whether the initial and final states have the same or opposite parity with respect to

reflection through the midpoint of the internuclear axis. Since the attached electron carries one-half unit of spin, there is also the obvious selection rule

$$S_f = S_i \pm \frac{1}{2}, \quad (6)$$

where S_f and S_i are the total spins of the initial and final states. Another rule applying to $\Sigma(\Lambda=0)$ states requires that symmetry with respect to reflection in a plane passing through the molecular axis be preserved; i. e., only $\Sigma^+ \rightarrow \Sigma^+$ and $\Sigma^- \rightarrow \Sigma^-$ transitions can occur. This is necessary because an electron with $\mu=0$ has a plus symmetry. The last two conditions do not influence the angular dependence given by Eq. (2), but are important in choosing the proper electronic states for the process.

O'Malley and Taylor argue that at sufficiently low electron energies the first term should dominate in Eq. (2); i. e., the angular distribution of the dissociation products is primarily determined by the lowest-order partial wave absorbed by the system. A transition from the ground $^3\Sigma_g^-$ state of O₂ to a $^2\Pi_u$ state requires $|\mu|=1$ and a summation over odd values of L ; thus from the first term in Eq. (2) the angular distribution of O⁻ is given by $|Y_{1,1}(\theta, \phi)|^2 \propto \sin^2\theta$. For a Π_g final state, one obtains a $\sin^2 2\theta$; for a Δ_u state, a $\sin^2 2\theta \sin^2\theta$; and for a Σ_u^+ , a $\cos^2\theta$ distribution. A Σ_g^+ state is removed from consideration since it involves a forbidden - to + transition. These predictions are consistent with the selection rules derived by Dunn.¹³

III. MEASUREMENTS

The apparatus used to measure the angular distribution of O⁻ from O₂ is the same as that used to study dissociative ionization of hydrogen and deuterium.¹⁹ A detailed description of the instrument is given in Ref. 19.

The negative ions, produced in a well-collimated low-energy electron beam which passed through a field-free region, drifted with their initial velocity through a pair of apertures which defined the angular resolution and then entered a 60°-sector magnet. The magnet filtered out the scattered electrons and selected ions of a particular momentum which were then accelerated and detected with an electron multiplier. The electron gun rotated with respect to the ion detection system, so that measurements of ion intensity could be made in the laboratory angular range $157^\circ \geq \Theta \geq 23^\circ$, where Θ is the angle between the electron beam axis and the axis of the detector apertures such that the low angles ($\Theta < 90^\circ$) correspond to "forward" directions. The angular resolution was estimated to be $\pm 1.2^\circ$.

It should be emphasized that the apparatus was completely shielded from outside electric and magnetic fields. The electron gun, collision chamber, magnet, ion-drift region, and multiplier were de-

signed to be self-shielding so that fields produced by one did not influence the operation of the other. Magnetic fields in the interaction region were always less than 0.05 G, and it was estimated that a uniform field of this strength would produce a 1.5° deflection of a 6.0-eV electron beam, which is slightly less than the angular resolution of the instrument.

The method of taking and analyzing data was similar to that used for hydrogen and deuterium.¹⁹ At each electron energy a plot of magnetic field versus ion count was obtained to determine the kinetic-energy distribution of the O⁻ ions. During each angular distribution measurement, the magnet was set at the peak of the observed ion-energy distribution. Since the process of O⁻ formation by dissociative attachment is resonant, the O⁻ kinetic-energy distribution should essentially reflect the electron-energy distribution, if the effect of thermal motion is neglected; i. e., it should be sharply peaked about a value E_0 given by Eq. (3). The effect of molecular thermal motion is to broaden the kinetic-energy distribution, but it should still be peaked about a value of E_0 corresponding to the peak of the electron-energy distribution.^{17,20} The measured values of E_0 , from the peak value of the observed energy distribution using the magnet calibration, were always systematically higher than the calculated values by about 0.25 eV. This small discrepancy is easily rationalized in terms of the contact potentials and errors in determining the electron-energy scale. Both the ion and electron energies could only be determined to within ± 0.2 eV,¹⁹ and no attempt was made here to correct for the effect of contact potentials. To calculate E_0 from Eq. (3) for a particular E_0 , the measured values $D=5.1$ eV²¹ and $A=1.5$ eV^{17,22} were used. It should be noted from Eq. (3) that when the electron energy is changed by an amount ΔE_e , the O⁻ kinetic energy should increase by $\frac{1}{2} \Delta E_e$. The differences in the observed, most probable kinetic energies for O⁻, calculated from the magnet calibration, always agreed with those predicted from the difference in measured electron energy.

Scans of magnetic field versus ion count for electron energies in the range 5–10 eV showed that background due to negative ions from impurities was negligible. The most abundant impurity was H₂O; however, for electron energies corresponding to dissociative attachment of O₂, the negative ions from H₂O are mostly of low momentum (H⁻ with a kinetic energy of 2.0 eV), and are formed with a cross section comparable to that for O⁻ from O₂.^{1,3,23} The cross sections for negative-ion formation from the possible contaminants CO₂ and CO are known to be an order of magnitude smaller than for O⁻ from O₂.³ Moreover, these should also produce ions with lower momentum. From magnet scans

there was never any evidence of negative ions with momentum lower or higher than that of the O^- from O_2 for all electron energies used.

A background of about 8 counts/min was always measured when the system was tuned to observe negative ions. This background was found to be independent of pressure and the operation of the apparatus components (excluding the multiplier) such as the ion gauges, ion pump, and electron gun. It was also independent of magnet-current setting and angle of observation and was typically less than 3% of the lowest ion-count rate. During each data run the background count was determined at several different angles by tuning the magnet to a point sufficiently far removed from the O^- peak. The observed background was, on the average, the same at all angles and magnetic field settings. The same average background was subtracted from the ion count at all angles before the data were corrected for variations in the interaction volume. The volume correction was made by multiplying the ion count by the sine of the angle.^{15,19}

The O^- measurements were typically made at a collision chamber pressure of 8×10^{-5} Torr and with an electron beam current of about 2×10^{-7} A. The gas sample was of ultrahigh purity and consisted of a 1-liter flask of 99.99% O_2 at atmospheric pressure. The dependence of the O^- count rate on pressure was found to be linear below 1.3×10^{-4} Torr with a zero intercept. The data were corrected for variations in electron current and pressure by the technique described in Refs. 16 and 19.

A 6.5-V 0.2- μ A electron beam was estimated to create a 0.01-V well in the collision chamber. Calculations of the effect of a field of this size on the trajectory of a 1.0-eV ion showed that it produces an angular deviation in the ion path which is much smaller than the angular resolution. Therefore, because of the small beam currents used in this experiment, the effect of electron-beam space charge is negligible.

The data shown in Fig. 2 have been arbitrarily normalized to 90° , and except for the data at 8.40 eV, all of the points shown in this figure represent an average of three data runs taken on three separate days. The data at 8.40 eV are from a single data run.

The data runs were performed in exactly the same way as described for hydrogen and deuterium.¹⁹ The duration of a typical data run was about two hours. The calculated voltage corrections needed to compensate for molecular recoil were applied to the spectrometer, although in this case the corrections were quite small, i.e., always less than 20 mV.

IV. RESULTS

The results shown in Figs. 2(a) and 2(b) indicate

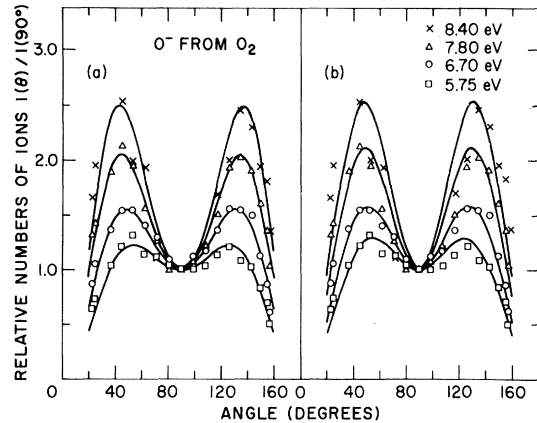


FIG. 2. Angular distribution of O^- from O_2 for $E_e = 8.40, 7.80, 6.70,$ and 5.75 eV. (a) Solid lines correspond to a single-state theory assuming a Π_u final state; and (b) curves represent a double-state theory assuming Π_u and Π_g final states and including contributions from only the lowest-order allowed partial waves.

that the angular distribution of O^- has a fairly strong energy dependence. The cross section appears to rapidly approach zero at 0° and 180° for all electron energies; therefore, on the basis of this observation the Σ_u^- states of O_2^- are immediately ruled out since those require a nonvanishing cross section in the forward and backward directions.¹³ The measurements were made at a temperature of about 330° K; therefore the data at $E_e = 6.7$ eV roughly correspond to the peak of the cross section. The arrows in Fig. 1 indicate where the angular distribution measurements were made. The observed relative ion count as a function of electron energy in this experiment was more consistent with the data of Schulz.³

An attempt has been made to explain the results in terms of one-parameter single- and double-state theories. The solid lines in Fig. 2(a) are of the form

$$|\alpha Y_{1,1}(\theta, \phi) + \beta Y_{1,3}(\theta, \phi)|^2 = |\sin\theta + 5\beta' \sin\theta \cos^2\theta|^2,$$

where

$$\beta/\alpha = (\frac{2}{7})^{1/2} \beta' / (1 + \beta'). \quad (7)$$

This corresponds to the first two terms in Eq. (2) for a ${}^2\Pi_u$ final O_2^- state with appropriate normalization at $\theta = 90^\circ$.²⁴ The parameter β' has been adjusted to give the "best fit" to the data at each energy. The lines in Fig. 2(b) are given by the function $\sin^2\theta + b \sin^2 2\theta$, corresponding to a sum of the lowest-order allowed terms in Eq. (2) for Π_u and Π_g final states. The relative contribution from these two states is determined by the adjustable parameter b . A sum of the first terms in Eq. (2) for a Π_u state plus a Δ_u state also qualitatively gives the same shape for the observed angular distribu-

TABLE I. Dependence of the fitting parameters on electron energy.

E_e (eV)	$\sigma_t(10^{-2} \pi a_0^2)$	$\sigma(\Pi_g)$	$\sigma(\Pi_u)$	b	β/α
8.40	0.27	0.08	0.19	2.00	0.353
7.80	0.65	0.16	0.49	1.57	0.309
6.70	1.56	0.26	1.30	1.02	0.247
5.75	1.03	0.13	0.90	0.69	0.187

tion; however, the fit to the data is poor and therefore not included.

The O₂⁻ states with configurations $(\Pi_u 2p)^4 (\Pi_g 2p)^3 (\sigma_u 2p)^2 \Pi_g$ and $(\Pi_u 2p)^3 (\Pi_g 2p)^4 (\sigma_u 2p)^2 \Pi_u$ should lie far above the O(³P)+O⁻(²P) energy limit¹² and could be responsible for the observed dissociation, although recent calculations indicate that they are probably too far above this limit to be important.¹⁰ The curves in Fig. 2(b) do not fit the data points very well at the higher energies, which means that if dissociative attachment is a two-state process involving either a Π_g plus a Π_u or a Π_u plus a Δ_u state, higher-order term in Eq. (2) must contribute significantly to the angular dependence. No attempt was made to fit the data to a multiparameter sum of two expansions, since the fit would not be unique. The relative contributions of the Π_u and Π_g states to the dissociative-attachment cross section at a given electron energy can be estimated from the parameter b , and the known total cross section σ_t , using the relationships $\sigma(\Pi_u) = 5\sigma_t/(5+b)$ and $\sigma(\Pi_g) = b\sigma_t/(5+b)$. Table I shows the dependence of $\sigma(\Pi_u)$, $\sigma(\Pi_g)$, and the parameters b and β/α on electron energy, where the values for σ_t were taken from Rapp and Briglia.⁵ As seen from this table, the estimated contribution to the cross section from a Π_g state is less than 20% of that from a Π_u state.

The curves in Fig. 2(a) corresponding to the single-state interpretation appear to fit the data quite well. From Table I, we have $\beta/\alpha = 0.353$ at the highest energy, implying that the largest observed contribution from the $L = 3$ term in Eq. (2) is 25%. Since the energy dependence for dissociative attachment is contained in the partial-wave expansion [Eq. (3)], a consideration of the behavior of $j_L(kr)$ shows that contributions from higher values of L become more important as k increases. This trend is consistent with the data of Fig. 2(a), which show an increasing contribution from the $L = 3$ wave as electron energy increases. For $E_e = 8.0$ eV, we have $k = 0.76(1/a_0)$, where a_0 is one Bohr radius; thus if the "range" of the molecular potential is taken to be $3.3a_0$, roughly corresponding to the dimensions of the O₂ molecule, then we find $j_1 = 0.416$, $j_3 = 0.104$, and $j_5 = 0.007$. For $L > 5$, the j_L values are all less than 2×10^{-4} and rapidly approach zero as L increases. From this simple energy argument one expects a significant contribution from the $L = 3$ partial wave for $E_e \sim 8.0$ eV

and a negligible contribution from partial waves with $L > 3$. The experimental results are consistent with this interpretation and suggest that the single-term approximation proposed by O'Malley and Taylor¹⁶ does not apply in the energy range for dissociative attachment of O₂. Dissociation via a single ${}^2\Pi_u$ resonance state with inclusion of the $L = 1$ and $L = 3$ partial waves appears to be the simplest and most attractive explanation for the observed energy dependence of the angular distribution of O⁻ ions, because the above argument shows that higher-order partial waves should be important even in a two-state theory. If the data are interpreted in terms of two or more repulsive resonances, one must also examine the possible effect of interactions between the states. This is especially important in this case because the results suggest that the states lie fairly close in energy with a separation possibly as small as 0.15 eV. From the Kronig selection rules²⁵ it is found that a first-order interaction is possible between Π_u and Δ_u states but not between Π_u and Π_g states.

Inclusion of an interaction term introduces a forward-backward asymmetry in the angular dependence of dissociative attachment. This can be understood from a consideration of Eqs. (2), (5), and (13) of O'Malley and Taylor.¹⁸ The angular dependence for dissociative attachment is determined by the electronic matrix element

$$V_A(\vec{R}) = \langle \phi_r | H_{e1} | \phi_{ad} \rangle, \quad (8)$$

where H_{e1} is the electronic Hamiltonian, ϕ_r is the final resonant electronic-state wave function, and ϕ_{ad} is a function containing the initial total molecular electronic state plus the incident electron plane wave [see Eq. (2) of O'Malley and Taylor]. If two interacting resonance states with unperturbed eigenfunctions ϕ_{r1}^0 and ϕ_{r2}^0 contribute to the process of dissociative attachment, then the eigenfunctions of the perturbed levels ϕ_{r1}^1 and ϕ_{r2}^1 which appear in Eq. (9) can be written as

$$\phi_{r1}^1 = c\phi_{r1}^0 + d\phi_{r2}^0, \quad \phi_{r2}^1 = d\phi_{r1}^0 + c\phi_{r2}^0, \quad (9)$$

where the value of d is determined by the magnitude of the interaction term appearing in the negative-ion molecule Hamiltonian (see, for example, Herzberg²⁶). The expansion of $V_A(\vec{R})$ corresponding to a single unperturbed final state is

$$V_A(\vec{R}) = (4)^{1/2} \sum_{L=1, \mu}^{\infty} Y_{L, \mu}(\theta, \phi) V_{L, |\mu|}(k), \quad (10)$$

where μ is given by Eq. (5) and the prime denotes a sum over the allowed L values. The angular dependence of the cross section for this case is proportional to $|V_A(R)|^2$ and given by Eq. (2) assuming validity of the Born-Oppenheimer and related adiabatic approximation.^{18, 27} For perturbed states given by Eq. (9), the expansions (10) have the form

$$V_{a1}(\vec{R}) = (4\pi)^{1/2} \left(c \sum_{L=1, \mu 1}^{\infty} [Y_{L, \mu 1}(\theta, \phi) V_{L, \mu 1}^{(1)}(k)] + d \sum_{L=1, \mu 2}^{\infty} [Y_{L, \mu 2}(\theta, \phi) V_{L, \mu 2}^{(2)}(k)] \right), \quad (11)$$

$$V_{a2}(\vec{R}) = (4\pi)^{1/2} \left(d \sum_{L=1, \mu 1}^{\infty} [Y_{L, \mu 1}(\theta, \phi) V_{L, \mu 1}^{(1)}(k)] + c \sum_{L=1, \mu 2}^{\infty} [Y_{L, \mu 2}(\theta, \phi) V_{L, \mu 2}^{(2)}(k)] \right). \quad (12)$$

The angular distribution for a two-state process, assuming the single-term approximation is valid for each, is then given by

$$I(k, \theta, \phi) \sim |a_{L1, \mu 1}(k) Y_{L1, \mu 1}(\theta, \phi)|^2 + |a_{L2, \mu 2}(k) Y_{L2, \mu 2}(\theta, \phi)|^2 + \gamma Y_{L2, \mu 2}(\theta, \phi) Y_{L1, \mu 1}(\theta, \phi), \quad (13)$$

where $L1, \mu 1$ and $L2, \mu 2$ are the lowest allowed values associated with the unperturbed final resonance states ϕ_{r1}^0 and ϕ_{r2}^0 . The last term in Eq. (13) gives a forward-backward asymmetry, and the value of its coefficient γ depends on the magnitude of the interaction responsible for the mixing of the unperturbed eigenfunctions of Eq. (9). For example, if the two final states involved in dissociative attachment of O_2 had the symmetries Π_u and Δ_u , then the asymmetry term would have the form $\sin^2\theta \times \sin 2\theta$.

A slight degree of forward-backward asymmetry not accounted for by molecular recoil is apparent in the data shown in Fig. 2, and although it may have physical significance, it could easily be a small instrumental effect such as might result from non-uniformities and small deflections of the electron beam. The results obtained here indicate that if

the process is due to two closely separated repulsive states, there is only a very weak interaction.

V. CONCLUSION

The observed electron-energy dependence for the angular distribution of O^- from dissociative attachment of O_2 can be explained most simply in terms of a transition to a single ${}^2\Pi_u$ resonance repulsive state. This interpretation is consistent with previous interpretations of the observed electron energy and temperature dependence of the O_2 dissociative-attachment cross section⁸ as well as measurements of the O^- kinetic energy.^{3,17} Although the dominant term in the expansion of the differential cross section [Eq. (2)] corresponds to the $L=1$ partial wave at all electron energies, a significant contribution from the $L=3$ partial wave becomes apparent at the higher energies. The magnitude of the $L=3$ term increases with energy and is consistent with the wavelength of the incident electron.

If dissociative attachment of O_2 is viewed as a process involving two closely separated resonance states, then the results indicate that higher-order partial waves should again be included. Moreover, the lack of a pronounced forward-backward asymmetry suggests that such states do not interact strongly. The results are not completely incompatible with a two-state theory, but this appears to be a less attractive alternative.

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communication). Their calculations indicate that the ${}^2\Pi_u$ state lies at the proper energy above the $O(^3P) + O(^2P)$ limit in the Franck-Condon region to produce O^- with the observed kinetic energy of 1.5 eV at the peak of the cross section. The ${}^4\Sigma_u^-$ state also has nearly the correct energy, but is predicted to be very broad so that decay from this state occurs predominantly via autodetachment.

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tron energies and referred to a mean value of electron momentum given by $\bar{k} = \int kf(k) dk / \int f(k) dk$, where $f(k)$ is the electron-momentum distribution function. The electron energy spread was found to be about 0.3 eV, and the stated values for the electron energy correspond to \bar{k} .

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Absolute Measurement of Differential Cross Sections for Electron Scattering in Helium[†]

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Absolute measurements of cross sections for electron-impact scattering in helium at an angle of 5° have been made for elastic scattering and excitation of the 2¹P and 2¹S states and for incident energies of 50–400 eV. Cross-section values of $\sigma(2^1P, 5^\circ)$ are found to be lower than theoretical Born values by (9.5 ± 5.4)% at 400 eV, (31.5 ± 4.6)% at 100 eV, and (62.5 ± 3.4)% at 50 eV. Deduced values of total 2¹P excitation cross sections ($E \geq 100$ eV) are in agreement with other experimental values. Our measurements for elastic scattering agree well with recent theoretical calculations.

I. INTRODUCTION

This article reports the absolute¹ measurement of small-angle differential cross sections for electron-impact scattering from He. Measurements were carried out for elastic scattering and excitation of the 2¹P and 2¹S states at fixed angles of 5° for incident energies of 50–400 eV utilizing a high-resolution electron spectrometer with a static gas target. The principal motivation for this work was (a) to provide accurate absolute small-angle cross-section values for normalization of angular-distribution measurements and (b) to study the nature and degree of breakdown of the Born approximation for excitation of the (optically) allowed 2¹P state.

Until recently,² little work appears to have been done on the absolute determinations of differential cross sections for electron scattering in He at small angles (< 15°) and in the energy range of 1–1000 eV.³ Lawson *et al.*,⁴ in determining theoretical zero-angle cross sections for He, pointed out inconsistencies existing in experimental data and emphasized the need for accurate small-angle elastic cross sections. It is desirable, also, to have accurate inelastic cross sections at small angles, where the contribution to the total cross section is the most significant. Excellent work on angular distributions in various gases has been carried out by Lassetre and collaborators^{5–7} who

normalized He (2¹P) excitation to theoretical Born values in order to calibrate their apparatus.

Although it is doubtful that the Born approximation is valid for excitation at 50 eV, evidence has been reported that the Born approximation still applies within a few percent down to 100 eV for differential⁸ and total⁹ n¹P excitations, whereas significant deviations from Born occur at 1500 eV for total⁹ n¹S excitations. In contrast, we find the difference between this experiment and Born theory for 2¹P excitation to be (9.5 ± 5.4)% at 400 eV and (31.5 ± 4.6)% at 100 eV.

In Sec. II, we briefly describe the apparatus. Section III discusses the theory of the experiment, the methods used in obtaining experimental parameters, and calibration and consistency checks made on the apparatus. In Sec. V, analysis of the data is discussed, the results are presented, and sources of systematic error are considered. In Sec. VI, we compare our results to other values of experimental and theoretical cross sections selected from various authors.

II. APPARATUS

Except for minor modifications, the apparatus has been described in detail by Kuyatt and Simpson.^{10,11} It was designed to operate over the energy range 50–400 eV with a resolution range of 0.04 to 0.1 eV. In the interest of completeness, we