

## Calculation of the excitation cross sections for the $B^1\Sigma_u^+$ and $C^1\Pi_u^+$ states in $e$ - $H_2$ scattering at 60 eV

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(Received 16 September 1994)

Integral cross sections have been calculated for the dipole-allowed transitions from  $X^1\Sigma_g^+$  to the  $B^1\Sigma_u^+$  and the  $C^1\Pi_u^+$  states in electron-hydrogen-molecule scattering at 60 eV. Experimental data and a universal function [A. Z. Msezane and I. A. Sakmar, *Phys. Rev. A* **49**, 2405 (1994)], which can extrapolate the generalized oscillator strength through the nonphysical region to  $K^2$  (momentum transfer squared) = 0, have been used in the calculation. A better agreement between theoretical results and experimental data has been achieved by using the universal function in the range of small scattering angles ( $\theta < 10^\circ$ ). The results show that the extrapolation used by experimentalists underestimated the differential cross sections (DCS's) at small scattering angles. Our calculation suggests that the universal function can be used to extrapolate the DCS for the electron-molecule scattering down to  $0^\circ$  at high energy.

PACS number(s): 34.10.+x, 34.50.Fa, 31.50.+w

### I. INTRODUCTION

One of the fundamental problems in molecular physics is electron-hydrogen molecule scattering, since the hydrogen molecule is the simplest with only two electrons and is the most abundant in the universe. The measured cross-section data are often used as the testing ground of the theory and they are important for the studies of plasmas, gas discharges, and astrophysics.

Although theories and experiments to study  $e$ - $H_2$  scattering have been developed in the past two decades, relatively few papers dealing with electronic excitation processes have been published [1–13,15–23]. The most important dipole-allowed transitions in  $e$ - $H_2$  scattering are  $X^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$  and  $X^1\Sigma_g^+ \rightarrow C^1\Pi_u^+$ . They give rise to the well-known Lyman and Werner band in the ultraviolet region of spectroscopy.

The main difficulty in the measurement using electron-energy-loss spectroscopy is resolving the heavily overlapped band structure [1–3]. For the  $B^1\Sigma_u^+$  state only the first three vibrational states are relatively free of other overlapping states. One way to address this problem is to measure the excitation cross section to one vibrational state (for the  $B^1\Sigma_u^+$  state it is usually the second vibrational state) and use the Franck-Condon (FC) factors to obtain the excitation cross section to other vibrational states [3].

Another difficulty in the experiments is measuring the differential cross section (DCS) at small scattering angles where the DCS increases dramatically as the scattering angles decrease. Therefore the experimental DCS data for the transitions to the  $B^1\Sigma_u^+$  and the  $C^1\Pi_u^+$  states in the impact-energy range of interest are only available for scattering angles above  $10^\circ$  [1–3]. However, the integral cross section receives a significant contribution mainly from small angles [24], particularly when impact energies are high. Without reliable data for the DCS's in the range of small scattering angles, the integral cross section

obtained from those DCS's is bound to contain large uncertainties. Several methods have been used in the extrapolation of the DCS to  $0^\circ$  by experimentalists. Srivastava and Jensen [1] extrapolated the DCS data to  $0^\circ$  by continuing the slope between the last two measured points. Khakoo and Trajmar [3] used theoretical DCS's whenever they were available to extrapolate their data to  $0^\circ$ . Their extrapolation, according to our calculation, uses a slope which is approximately three times steeper than the slope achieved from continuing the last two measured points. The difference between the two measurements as shown in Fig. 1 is partly due to the use of different extrapolation methods. This difference reaches a maximum at 60 eV because a larger percentage of the excitation cross section will be contributed from small angles at higher impact energy.

From a theoretical point of view, the wave function of the molecule is more complicated than that of the atom. It includes not only the electronic part but also the vibrational and rotational parts. In most experiments it is impossible to resolve the rotational transitions. Therefore, the summation over the final rotational states and averaging over the initial states is usually carried out in order to compare theoretical results with the experimental data [4–8]. Theoretical results from different calculations, such as the Born, Born-Ochkur, and distorted-wave (DW) calculations [13,5], are closer to each other at angles of  $\theta < 20^\circ$ . When the scattering angle increases, the difference between the calculations appear. This indicates that theoretical calculations are more reliable at small angles than what the experimentalists used in their extrapolation. However, to represent the electronic ground and excited states accurately the expansion of the wave function requires a large number of terms. For example, Kolos [9,10] used a 36-term expansion for the  $X^1\Sigma_g^+$  ground state and 90 terms for the  $B^1\Sigma_u^+$  excited state. This requirement makes the systematic improvement of the theoretical calculations difficult even for the Born ap-

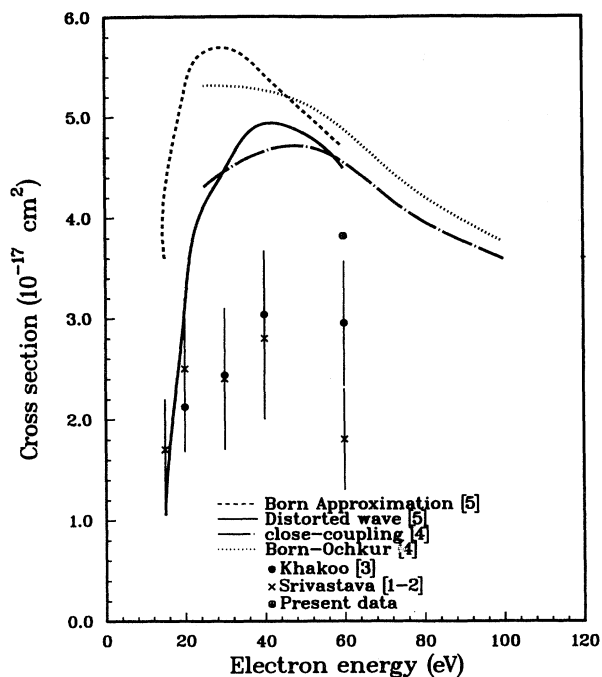


FIG. 1. Integral cross sections for the excitation of the  $B^1\Sigma_u^+$  state in  $e\text{-H}_2$  scattering. Solid line, distorted wave; dashed line, Born approximation; dotted line, Born-Ochkur approximation; dashed-dotted line, close-coupling calculation. Black dots and crosses are the experimental data of Khakoo and Trajmar and of Srivastava and Jensen, respectively.

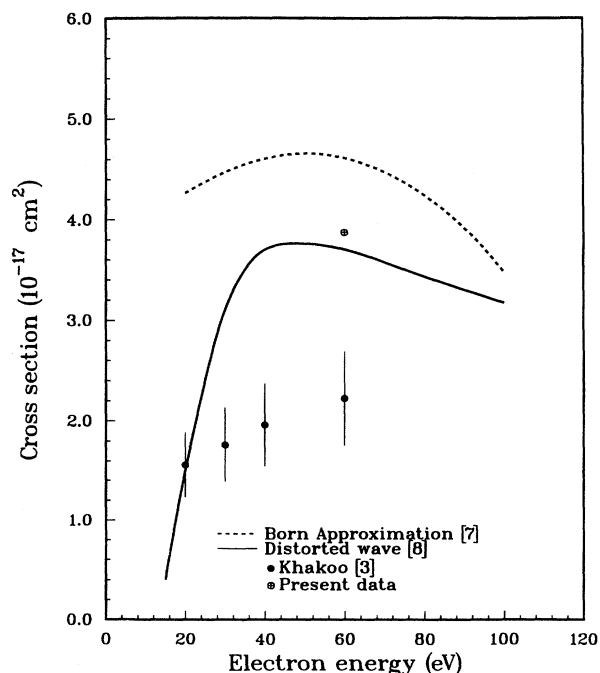


FIG. 2. Integral cross sections for the excitation of the  $C^1\Pi_u^+$  state in  $e\text{-H}_2$  scattering. Symbols are the same as in Fig. 1.

proximation. It is also difficult for the experimentalists to use the theory as a guide to calculate the integral cross sections. Because of these problems the disagreement between theory and experiment for the transitions to the  $B^1\Sigma_u^+$  and the  $C^1\Pi_u^+$  states has lingered for a long time, particularly for the 60-eV impact energy, as shown in Figs. 1 and 2.

## II. THEORY AND RESULT

In this paper we will introduce a practical method to calculate the DCS's for the electronic transitions at small-angle and high-impact energy in electron-molecule collisions. Recently, Msezane and Sakmar [14] have developed an extrapolation formula for the generalized oscillator strength (GOS) using rigorous bounds on  $K^2$  (momentum transfer squared). This formula, which they called the universal function, can be expressed as

$$f^u(K) = -f^o \left[ 1 - \frac{2}{\left[ 1 + \frac{K^2}{2w} \right]} \right], \quad (1)$$

where  $f^u(K)$  is the GOS,  $f^o$  is the optical oscillator strength (OOS), and  $K^2$ , in atomic units, is given by

$$K^2 = 2E \left\{ 2 - \frac{w}{E} - 2 \left[ 1 - \frac{w}{E} \right]^{1/2} \cos\theta \right\}, \quad (2)$$

where  $w$ ,  $\theta$ , and  $E$  are, respectively, the excitation energy, the scattering angle, and the impact energy. The equation is applicable only to optically allowed transitions for small  $K^2$  values. The formula has been used successfully in many electron-atom collision systems [24–28]. In electron-atom scattering, Eq. (1) describes the relationship between the GOS and  $K^2$  for a dipole-allowed transition.

In the electron-molecule collision, Eq. (1) may be used to evaluate the GOS for each rotationally averaged vibrational state. The validity of Eq. (1) in electron-molecule scattering is shown in Fig. 3. Figure 3 gives the relation-

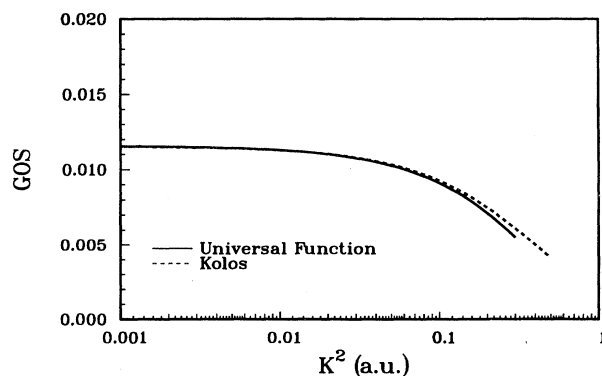


FIG. 3. Relationship between GOS and  $K^2$  for the transition to the second vibrational state of  $B^1\Sigma_u^+$  in the  $e\text{-H}_2$  collision. The solid line is calculated by the universal function. The dashed line is the Born approximation from Kolos [10] with a 36-term expansion for the electronic ground state, and a 90-term expansion for the  $B^1\Sigma_u^+$  state.

ship between the GOS and  $K^2$  for the second vibrational state of the  $B^1\Sigma_u^+$  state in the scattering process  $e\text{-H}_2$ . In Fig. 3 the solid line is the universal function and the dashed line is calculated from the Born approximation [10]. The OOS and the excitation energy are taken from Allison and Dalgarno [11] and Kolos [10], respectively. As in the electron-atom scattering, when  $K^2 \ll 1$  a.u., the universal function merges with the Born calculation. Therefore, Eq. (1) used in electron-molecule scattering will always have a correct asymptotic behavior. It is a good tool, as in the electron-atom scattering, to extrapolate the GOS to the OOS through the unphysical region even for the electron-molecule collision.

The DCS for each rotationally averaged vibrational state may be obtained from

$$\left(\frac{d\sigma}{d\Omega}\right)_i = -2 \left(1 - \frac{w_i}{E}\right)^{1/2} \frac{f_i^o}{K^2 w_i} \left[1 - \frac{2}{1 + \frac{K^2}{2w_i}}\right]. \quad (3)$$

The total DCS to the  $B^1\Sigma_u^+$  or  $C^1\Pi_u^+$  electronic state is the summation of the DCS's of all the vibrational states,

$$\frac{d\sigma}{d\Omega} = \sum_{i=1}^n -2 \left(1 - \frac{w_i}{E}\right)^{1/2} \frac{f_i^o}{K^2 w_i} \left[1 - \frac{2}{1 + \frac{K^2}{2w_i}}\right], \quad (4)$$

where  $f_i^o$  is the OOS for the  $i$ th vibrational state. Using the Franck-Condon factor, Eq. (4) may be expressed approximately as

$$\frac{d\sigma}{d\Omega} = \sum_{i=1}^n -2 \left(1 - \frac{w_i}{E}\right)^{1/2} \frac{f_e^o q_i}{K^2 w_i} \left[1 - \frac{2}{1 + \frac{K^2}{2w_i}}\right], \quad (5)$$

where  $q_i$  is the Franck-Condon factor for the  $i$ th vibrational state and  $f_e^o$  is the OOS calculated by using the electronic wave function. The data needed in the calculation are listed in Table I. The first column is the number of the vibrational states. The second and third columns are, respectively, the OOS and Franck-Condon factors for the  $B^1\Sigma_u^+$  state. The fifth and sixth columns are the same data for the  $C^1\Pi_u^+$  state. The fourth and seventh columns are the excitation energy. The OOS was calcu-

TABLE I. OOS, Frank-Condon factors, and excitation energy for the Lyman and Werner systems. The numbers in brackets denote multiplicative powers of ten.

$v'$	$B^1\Sigma_u^+$			$C^1\Pi_u^+$		
	OOS (a.u.)	FC (a.u.)	$w$ (a.u.)	OOS (a.u.)	FC (a.u.)	$w$ (a.u.)
0	1.689[-3]	4.223[-3]	0.411 198	4.760[-2]	1.226[-1]	0.4518
1	5.790[-3]	1.488[-2]	0.417 205	7.284[-2]	1.926[-1]	0.4621
2	1.156[-2]	3.052[-2]	0.423 046	6.982[-2]	1.892[-1]	0.4720
3	1.755[-2]	4.762[-2]	0.428 727	5.472[-2]	1.518[-1]	0.4812
4	2.250[-2]	6.272[-2]	0.434 255	3.874[-2]	1.098[-1]	0.4901
5	2.571[-2]	7.361[-2]	0.439 632	2.598[-2]	7.524[-2]	0.4981
6	2.704[-2]	7.945[-2]	0.444 863	1.700[-2]	5.016[-2]	0.5059
7	2.673[-2]	8.059[-2]	0.449 947	1.099[-2]	3.301[-2]	0.5128
8	2.523[-2]	7.802[-2]	0.454 889	7.098[-3]	2.168[-2]	0.5191
9	2.298[-2]	7.282[-2]	0.459 688	4.592[-3]	1.423[-2]	0.5246
10	2.035[-2]	6.606[-2]	0.464 348	2.976[-3]	9.347[-3]	0.5298
11	1.764[-2]	5.863[-2]	0.468 871	1.909[-3]	6.059[-3]	0.5338
12	1.504[-2]	5.115[-2]	0.473 259	1.171[-3]	3.749[-3]	0.5371
13	1.266[-2]	4.403[-2]	0.477 515	5.590[-4]	1.800[-3]	0.5393
14	1.055[-2]	3.751[-2]	0.481 640			
15	8.730[-3]	3.169[-2]	0.485 637			
16	7.185[-3]	2.663[-2]	0.489 509			
17	5.891[-3]	2.227[-2]	0.493 258			
18	4.820[-3]	1.858[-2]	0.496 886			
19	3.939[-3]	1.547[-2]	0.500 395			
20	3.219[-3]	1.287[-2]	0.503 786			
21	2.632[-3]	1.071[-2]	0.507 062			
22	2.154[-3]	8.913[-3]	0.510 223			
23	1.766[-3]	7.426[-3]	0.513 272			
24	1.450[-3]	6.194[-3]	0.516 207			
25	1.193[-3]	5.173[-3]	0.519 029			
26	9.815[-4]	4.317[-3]	0.521 735			
27	8.057[-4]	3.593[-3]	0.524 325			

TABLE II. DCS's ( $10^{-16}$  cm<sup>2</sup>/sr) for the transition to the  $B^1\Sigma_u^+$  state.

Angle (deg)	DCS <sub>oos</sub>	DCS <sub>FC</sub>	DW
0.0	5.700	5.314	5.200
0.2	5.694	5.308	
0.4	5.677	5.292	
0.6	5.648	5.265	
0.8	5.608	5.228	
1.0	5.557	5.182	
2.0	5.164	4.819	
3.0	4.610	4.307	
4.0	3.994	3.736	
5.0	3.388	3.175	
6.0	2.837	2.662	
7.0	2.358	2.214	
8.0	1.951	1.834	
9.0	1.611	1.516	
10.0	1.329	1.252	1.210

TABLE III. DCS's ( $10^{-16}$  cm<sup>2</sup>/sr) for the transition to the  $C^1\Pi_u^+$  state.

Angle (deg)	DCS <sub>oos</sub>	DCS <sub>FC</sub>	DW	
0.0	5.658	5.107	4.1	
0.2	5.653	5.104		
0.4	5.637	5.088		
0.6	5.611	5.065		
0.8	5.575	5.032		
1.0	5.529	4.991		
2.0	5.174	4.670		
3.0	4.662	4.211		
4.0	4.083	3.689		
5.0	3.502	3.166		2.4
6.0	2.964	2.680		
7.0	2.486	2.250		
8.0	2.075	1.878		
9.0	1.727	1.564		
10.0	1.436	1.300	0.92	

lated by Allison and Dalgarno [11] and the excitation energies of the vibrational states were taken from Kolos [10] for the  $B^1\Sigma_u^+$  state and Arrighini and Biondi [13] for the  $C^1\Pi_u^+$  state. Using Eq. (3) we calculated the DCS's of the  $B^1\Sigma_u^+$  and  $C^1\Pi_u^+$  states from  $\theta=0^\circ$  to  $10^\circ$  and compared them with the DW calculation. Twenty-eight vibrational states for the  $B^1\Sigma_u^+$  state and 14 vibrational states for the  $C^1\Pi_u^+$  state have been used in the calculations.

Table II lists our calculation for the  $B^1\Sigma_u^+$  state. The DW calculations [5] at  $0^\circ$  and  $10^\circ$  are also given in Table II. The data in the second column are obtained by using OOS's, and in the third column they are calculated from the Franck-Condon factors. Excellent agreement has been obtained between our calculations and the DW results at  $0^\circ$  and  $10^\circ$ . As the DW also used the Franck-Condon factors in the calculation, it is not surprising that the data in the third column are closer to the DW results. Table III is the same calculation but for the  $C^1\Pi_u^+$  state. The DW results in the fourth column were obtained from Fig. 6 of Lee, Luocheese, and McKoy [8]. Their results of the  $C^1\Pi_u^+$  state agree reasonably well with our data but are a little bit lower than our calculation. After comparing the cross sections of the  $B^1\Sigma_u^+$  state, we found that the data of Lee, Luocheese, and McKoy are also lower than that of Flifet [5]. Tables IV and V give the results of the integral cross sections for the transitions to the  $B^1\Sigma_u^+$  and  $C^1\Pi_u^+$  states at 60 eV. The tables show that the largest contribution to the excitation cross section comes from angles of  $\theta < 10^\circ$ . Therefore a large error will be introduced to the integral cross sections if the experimentalists calculate it without the appropriate theory to guide them.

In our calculation, the DCS's for the scattering angles between  $10^\circ$  and  $120^\circ$  were taken from the measurement of Khakoo and Trajmar [3]. Data of less than  $10^\circ$  came from Tables II and III. Since the cross sections contributed from  $120^\circ$  to  $180^\circ$  are small, we used a linear equation in this region to represent the DCS data and obtained the contribution to the integral cross section by integrating the equation from  $120^\circ$  to  $180^\circ$  analytically. The data from  $0^\circ$  to  $120^\circ$  were fitted by a cubic spline.

In Tables IV and V we also listed the results of the distorted-wave (DW) approximation, the Born approximation (Born), the Born-Ochkur approximation (BO), and the close-coupling (CC) method. The experimental

TABLE IV. Integral cross sections ( $10^{-17}$  cm<sup>2</sup>) for the transition to the  $B^1\Sigma_u^+$  state at 60 eV.

Angle (deg)	Eq. (4)	Eq. (5)	DW Ref. [5]	Born Ref. [5]	CC Ref. [4]	BO Ref. [4]	Ref. [3]	Ref. [2]
0-10	2.51	2.34						
10-120	1.29	1.29						
120-180	0.0246	0.0246						
Total	3.82	3.65	4.49	4.70	4.62	4.96	2.951	1.8

TABLE V. Integral cross sections ( $10^{-17}$  cm<sup>2</sup>) for the transition to the  $C^1\Pi_u^+$  state at 60 eV.

Angle (deg)	Eq. (4)	Eq. (5)	DW Ref. [8]	Born Ref. [7]	BO Ref. [13]	Ref. [3]
0-10	2.63	2.38				
10-120	1.22	1.22				
120-180	0.0265	0.0265				
Total	3.87	3.63	3.71	4.42	4.30	2.220

and the close-coupling (CC) method. The experimental data from Khakoo and Trajmar [3] and Srivastava and Jensen [1], including cross sections extrapolated to  $0^\circ$ , are also given in the tables. The integral cross sections for  $B^1\Sigma_u^+$  and  $C^1\Pi_u^+$  at 60 eV are  $3.82 \times 10^{-17}$  cm<sup>2</sup> and  $3.87 \times 10^{-17}$  cm<sup>2</sup>, respectively. A better agreement between measurement and the theory has thus been obtained by using the universal function (Tables II and III) in the calculation involving small scattering angles. Khakoo and Trajmar used their experimental data and obtained  $2.2 \times 10^{-17}$  cm<sup>2</sup> for the integral cross section of the  $C^1\Pi_u^+$  state at 60 eV. We obtained  $3.87 \times 10^{-17}$  cm<sup>2</sup> for the same state by using the same data except that the universal function was used for the small angles. The calculation we used is approximately equivalent to the appropriate extrapolation of the measurement to  $0^\circ$  using the distorted-wave approximation as a guide. It brings the experimental results closer to the theoretical evaluation ( $3.7 \times 10^{-17}$  cm<sup>2</sup>). A better agreement between theory and measurement is also obtained for the  $B^1\Sigma_u^+$  state. The use of the universal function at small angles thus brings the experimental data from  $2.95 \times 10^{-17}$  cm<sup>2</sup> to  $3.82 \times 10^{-17}$  cm<sup>2</sup>, which is closer to the theoretical result (DW) of  $4.49 \times 10^{-17}$  cm<sup>2</sup>.

The calculations show that a long-standing discrepancy between theory and measurement can be resolved, or improved, by using the appropriate method to extrapolate the experimental data to  $0^\circ$  scattering angle. These calculations further demonstrate that Eq. (3) can be useful to the experimentalists to evaluate the small-angle DCS at high energies. We note that this is the first time that the universal function has been employed in electron-molecule scattering.

### III. CONCLUSION

Integral cross sections for the transitions from the ground state to the  $B^1\Sigma_u^+$  and the  $C^1\Pi_u^+$  states in  $e$ -H<sub>2</sub> scattering have been evaluated at 60 eV by using the experimental data of Khakoo and Trajmar for the scattering angles of  $10^\circ$  to  $120^\circ$  and the universal function for the angles less than  $10^\circ$ . The results for the  $C^1\Pi_u^+$  state show excellent agreement with the distorted-wave calculation. A better agreement between theory [4,5] and experiment [3] has also been achieved for the  $B^1\Sigma_u^+$  state. The results demonstrate that the extrapolation of the experimental data to  $0^\circ$  in the evaluation of the integral cross sections used by the experimentalists before underestimated the contribution between  $0^\circ$  and  $10^\circ$ . Our universal formula thus provides a useful extrapolation procedure for use by the experimentalists at small scattering angles even for electron-molecule scattering.

### ACKNOWLEDGMENTS

This work was supported in part by the DOE, Office of Basic Energy Sciences, Division of Chemical Sciences, and The National Science Foundation. Supercomputer time on the National Energy Research Supercomputer Center (NERSC) Cray Computers provided by the U.S. DOE, Office of Fusion Energy, is appreciated. A.Z.M. is grateful to Professor Alex Dalgarno for the invitation and for suggesting the problem and to his colleagues for fruitful discussions and assistance while visiting the Institute for Theoretical Atomic and Molecular Physics at Harvard University.

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