Effect of ground-state electron correlation on the $(e,2e)$ reaction spectroscopy of $H_2(^1\Sigma_g^+)$

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A critical theoretical study of the electron momentum distributions obtained for $H_2(^1\Sigma_g^+)$ by the $(e,2e)$ reaction technique for transitions to the ground and the $n=2$ and 3 excited states is presented. Two very highly electron correlated ground-state wave functions for $H_2(^1\Sigma_g^+)$, each of which yields more than 96% of the "exact" binding energy and the exact Born-Oppenheimer wave function of H_2^+ in the ground and various excited states, were employed in the computations within the context of the plane-wave impulse approximation (PWIA). For the ground-state transition, the theoretical values computed from both correlated wave functions and a self-consistent-field wave function are all in relatively good agreement with experiments for recoil momenta in the range $(0.11 \leq q \leq 1.0$ a.u.). In order to discriminate among the various theoretical values reported here it is highly desirable to have experimental data for q smaller than 0.05 a.u. The experimental data for the transitions to the $n=2$ excited states ($2s\sigma_g$, $2p\pi_u$, and $2p\sigma_u$) are surprisingly not in as good agreement with the theoretical values reported here. The failure to describe the transition to excited states with a molecular angular momentum quantum number other than zero may be due to (i) the inadequacy of the ground-state wave function, (ii) the failure of the Franck-Condon principle in the transition, (iii) the need to go beyond the PWIA, and/or (iv) the need for improved and extended experimental data.

I. INTRODUCTION

For many decades quantum chemists have been very interested in obtaining accurate *ab inito* wave functions for atoms and molecules; however, the usual criterion to assess the accuracy of these computed wave functions has been based on the energy variation principle, i.e., by comparing the total electron energies obtained.[1] Recently studies of electron momentum distributions in atoms and molecules by means of the $(e,2e)$ reaction technique[2,3] have been of considerable interest to both experimentalists and theoreticians. In particular in the studies on helium reported recently,[4] it is clearly shown that the $(e,2e)$ reaction (for transitions to excited states of the residual ion) is able to discriminate among various correlated helium wave functions which contain a comparable amount of the electron correlation energy. Fairly good agreement between theory and experiment in studies of the Compton profile and ground-state momentum distribution obtained from photon scattering, or high-energy electron scattering,[5] as well as $(e,2e)$ reaction spectroscopy for He have been reported elsewhere.[3,4] However, for molecular hydrogen, the discrepancies found in studies of the Compton profile[6-8] by high-energy photon scattering, high-energy electron scattering,[5] and $(e,2e)$ reaction spectroscopy in the transition to the ground state of H_2^+ reported recently[9] are quite disturbing and led us to the present investigation. Calculations of the $(e,2e)$ reaction cross-section for the ground- and excited-state transitions ($n=2$ and 3) are presented here. A self-consistent-field (SCF) and two configuration-interaction (CI) wave functions[10,11] for H_2 and the exact [Born-Oppenheimer (BO)] wave function[12] of H_2^+ were used in the computations. Since the hydrogen molecular ion wave function is known exactly,[12] the effect of electron correlation on the $(e,2e)$ reaction cross section can be demonstrated by using various correlated wave functions for molecular hydrogen. A discussion of the comparison between theory and experiment is made for the transition to the ground state and to those excited states for which experimental data is available, namely, $2p\sigma_u$, $2p\pi_u$, and $2s\sigma_g$.

II. CALCULATIONS AND RESULTS

In the plane-wave impulse approximation[3] (PWIA) the $(e,2e)$ reaction cross section is proportional to the square of the Fourier transform of the overlap amplitude between the ground state of the target and the residual ion state,

$$\sigma(q) \propto |T_M|^2 \int d\hat{q}\, |F_{if}(\mathbf{q})|^2 \delta(E_0 - E_A - E_B - \epsilon) , \quad (1)$$

where T_M is the off-shell Coulombic T matrix which is a constant for the noncoplanar experimental geometry usually employed; E_0, E_A, and E_B are, respectively, the incident, scattered, and ejected electron energies; ϵ is the separation energy of the reaction, and $\mathbf{q}$ $(=\mathbf{k}_0 - \mathbf{k}_A - \mathbf{k}_B)$ is the recoil momentum. The overlap amplitude in momentum space $F_{if}(\mathbf{q})$ may be written as

$$F_{if}(\mathbf{q}) = (2\pi)^{-3/2} \langle \Psi_f | \exp(i\mathbf{q}\cdot\mathbf{r}) | \Psi_i \rangle , \quad (2)$$

where the wave function Ψ_i of the target molecule (H_2) may be written[13] as an expansion in natural molecular or-

TABLE I. Values of $O_{if}(q)$ for various transitions: $H_2(e,2e)H_2^+$ calculated using a SCF and the DJ and HS CI wave functions.

	$1s\sigma_g$			$2s\sigma_g$		
q	SCF	DJ	HS	SCF	DJ	HS
0.000 00	1.182 3	1.134 5	1.111 7	$0.301\,51\times10^{-1}$	$0.290\,34\times10^{-1}$	$0.281\,54\times10^{-1}$
0.050 00	1.172 6	1.125 4	1.103 2	$0.299\,03\times10^{-1}$	$0.297\,68\times10^{-1}$	$0.289\,19\times10^{-1}$
0.100 00	1.144 8	1.099 9	1.078 4	$0.291\,93\times10^{-1}$	$0.290\,06\times10^{-1}$	$0.282\,07\times10^{-1}$
0.150 00	1.099 9	1.056 7	1.038 4	$0.280\,48\times10^{-1}$	$0.277\,78\times10^{-1}$	$0.270\,59\times10^{-1}$
0.200 00	1.040 6	1.000 7	0.985 38	$0.265\,37\times10^{-1}$	$0.261\,68\times10^{-1}$	$0.255\,45\times10^{-1}$
0.250 00	0.970 17	0.934 06	0.921 96	$0.247\,40\times10^{-1}$	$0.242\,65\times10^{-1}$	$0.237\,47\times10^{-1}$
0.300 00	0.891 93	0.859 92	0.851 04	$0.227\,45\times10^{-1}$	$0.221\,69\times10^{-1}$	$0.217\,53\times10^{-1}$
0.350 00	0.809 33	0.781 50	0.775 59	$0.206\,39\times10^{-1}$	$0.199\,76\times10^{-1}$	$0.196\,53\times10^{-1}$
0.400 00	0.725 51	0.701 78	0.698 40	$0.185\,01\times10^{-1}$	$0.177\,73\times10^{-1}$	$0.175\,29\times10^{-1}$
0.450 00	0.643 17	0.623 30	0.621 92	$0.164\,01\times10^{-1}$	$0.156\,32\times10^{-1}$	$0.154\,52\times10^{-1}$
0.500 00	0.564 45	0.548 09	0.548 17	$0.143\,94\times10^{-1}$	$0.136\,10\times10^{-1}$	$0.134\,77\times10^{-1}$
0.550 00	0.490 90	0.477 66	0.478 70	$0.125\,19\times10^{-1}$	$0.117\,44\times10^{-1}$	$0.116\,45\times10^{-1}$
0.600 00	0.423 53	0.412 97	0.414 54	$0.108\,01\times10^{-1}$	$0.100\,56\times10^{-1}$	$0.998\,07\times10^{-2}$
0.650 00	0.362 85	0.354 56	0.356 33	$0.925\,31\times10^{-2}$	$0.855\,57\times10^{-1}$	$0.849\,59\times10^{-2}$
0.700 00	0.308 98	0.302 57	0.304 30	$0.787\,94\times10^{-2}$	$0.724\,13\times10^{-2}$	$0.719\,15\times10^{-2}$
0.750 00	0.261 75	0.256 86	0.258 41	$0.667\,49\times10^{-2}$	$0.610\,38\times10^{-2}$	$0.606\,08\times10^{-2}$
0.800 00	0.220 77	0.217 09	0.218 38	$0.562\,99\times10^{-2}$	$0.512\,96\times10^{-2}$	$0.509\,15\times10^{-2}$
0.850 00	0.185 53	0.182 81	0.183 81	$0.473\,13\times10^{-2}$	$0.430\,23\times10^{-2}$	$0.426\,79\times10^{-2}$
0.900 00	0.155 46	0.153 48	0.154 20	$0.396\,44\times10^{-2}$	$0.360\,44\times10^{-2}$	$0.357\,33\times10^{-2}$
0.950 00	0.129 96	0.128 54	0.129 02	$0.331\,41\times10^{-2}$	$0.301\,88\times10^{-2}$	$0.299\,08\times10^{-2}$
1.000 00	0.108 45	0.107 45	0.107 73	$0.276\,55\times10^{-2}$	$0.252\,94\times10^{-2}$	$0.250\,42\times10^{-2}$
1.100 00	$0.752\,49\times10^{-1}$	$0.748\,01\times10^{-1}$	$0.748\,04\times10^{-1}$	$0.191\,89\times10^{-2}$	$0.178\,18\times10^{-2}$	$0.176\,18\times10^{-2}$
1.200 00	$0.521\,01\times10^{-1}$	$0.519\,39\times10^{-1}$	$0.518\,09\times10^{-1}$	$0.132\,86\times10^{-2}$	$0.126\,40\times10^{-2}$	$0.124\,83\times10^{-2}$
1.300 00	$0.360\,81\times10^{-1}$	$0.360\,59\times10^{-1}$	$0.358\,88\times10^{-1}$	$0.920\,10\times10^{-3}$	$0.904\,44\times10^{-3}$	$0.892\,14\times10^{-3}$
1.400 00	$0.250\,37\times10^{-1}$	$0.250\,75\times10^{-1}$	$0.249\,14\times10^{-1}$	$0.638\,46\times10^{-3}$	$0.652\,85\times10^{-3}$	$0.643\,19\times10^{-3}$
1.500 00	$0.174\,30\times10^{-1}$	$0.174\,88\times10^{-1}$	$0.173\,58\times10^{-1}$	$0.444\,48\times10^{-3}$	$0.475\,04\times10^{-3}$	$0.467\,48\times10^{-3}$
1.600 00	$0.121\,85\times10^{-1}$	$0.122\,45\times10^{-1}$	$0.121\,49\times10^{-1}$	$0.310\,73\times10^{-3}$	$0.348\,03\times10^{-3}$	$0.342\,17\times10^{-3}$
1.700 00	$0.855\,98\times10^{-2}$	$0.861\,35\times10^{-2}$	$0.854\,73\times10^{-2}$	$0.218\,28\times10^{-3}$	$0.256\,41\times10^{-3}$	$0.251\,95\times10^{-3}$
1.800 00	$0.604\,53\times10^{-2}$	$0.609\,03\times10^{-2}$	$0.604\,78\times10^{-2}$	$0.154\,16\times10^{-3}$	$0.189\,75\times10^{-3}$	$0.186\,45\times10^{-3}$
1.900 00	$0.429\,38\times10^{-2}$	$0.433\,03\times10^{-2}$	$0.430\,49\times10^{-2}$	$0.109\,50\times10^{-3}$	$0.140\,93\times10^{-3}$	$0.138\,56\times10^{-3}$
2.000 00	$0.306\,80\times10^{-2}$	$0.309\,71\times10^{-2}$	$0.308\,35\times10^{-2}$	$0.782\,38\times10^{-4}$	$0.104\,99\times10^{-3}$	$0.103\,35\times10^{-3}$
2.200 00	$0.159\,64\times10^{-2}$	$0.161\,44\times10^{-2}$	$0.161\,31\times10^{-2}$	$0.407\,10\times10^{-4}$	$0.587\,26\times10^{-4}$	$0.580\,61\times10^{-4}$
2.400 00	$0.853\,57\times10^{-3}$	$0.864\,79\times10^{-3}$	$0.867\,50\times10^{-3}$	$0.217\,67\times10^{-4}$	$0.331\,98\times10^{-4}$	$0.330\,26\times10^{-4}$
2.600 00	$0.470\,24\times10^{-3}$	$0.477\,36\times10^{-3}$	$0.480\,66\times10^{-3}$	$0.119\,92\times10^{-4}$	$0.189\,98\times10^{-4}$	$0.190\,37\times10^{-4}$
2.800 00	$0.267\,80\times10^{-3}$	$0.272\,43\times10^{-3}$	$0.275\,18\times10^{-3}$	$0.682\,92\times10^{-5}$	$0.110\,38\times10^{-4}$	$0.111\,41\times10^{-4}$
3.000 00	$0.158\,19\times10^{-3}$	$0.161\,27\times10^{-3}$	$0.163\,26\times10^{-3}$	$0.403\,40\times10^{-5}$	$0.653\,45\times10^{-5}$	$0.663\,85\times10^{-5}$
3.200 00	$0.971\,82\times10^{-4}$	$0.992\,73\times10^{-4}$	$0.100\,60\times10^{-3}$	$0.247\,82\times10^{-5}$	$0.395\,64\times10^{-5}$	$0.403\,94\times10^{-5}$
3.400 00	$0.621\,61\times10^{-4}$	$0.636\,02\times10^{-4}$	$0.644\,40\times10^{-4}$	$0.158\,52\times10^{-5}$	$0.245\,85\times10^{-5}$	$0.251\,77\times10^{-5}$
3.600 00	$0.413\,49\times10^{-4}$	$0.423\,51\times10^{-4}$	$0.428\,53\times10^{-4}$	$0.105\,44\times10^{-5}$	$0.157\,21\times10^{-5}$	$0.161\,15\times10^{-5}$
3.800 00	$0.285\,03\times10^{-4}$	$0.292\,05\times10^{-4}$	$0.294\,87\times10^{-4}$	$0.726\,86\times10^{-6}$	$0.103\,58\times10^{-5}$	$0.106\,08\times10^{-5}$
4.000 00	$0.202\,54\times10^{-4}$	$0.207\,48\times10^{-4}$	$0.208\,93\times10^{-4}$	$0.516\,50\times10^{-6}$	$0.702\,86\times10^{-6}$	$0.718\,25\times10^{-6}$

TABLE I. (*Continued*).

	$2p\sigma_u$		$2p\pi_u$	
q	DJ	HS	DJ	HS
0.000 00	0.000 00	0.000 00	0.000 00	0.000 00
0.050 00	$0.122\,37\times10^{-4}$	$0.921\,43\times10^{-5}$	$0.501\,39\times10^{-7}$	$0.446\,93\times10^{-7}$
0.100 00	$0.475\,68\times10^{-4}$	$0.369\,97\times10^{-4}$	$0.196\,92\times10^{-6}$	$0.174\,90\times10^{-6}$
0.150 00	$0.104\,50\times10^{-3}$	$0.819\,98\times10^{-4}$	$0.436\,05\times10^{-6}$	$0.384\,90\times10^{-6}$
0.200 00	$0.179\,58\times10^{-3}$	$0.142\,73\times10^{-3}$	$0.756\,66\times10^{-6}$	$0.663\,21\times10^{-6}$
0.250 00	$0.268\,64\times10^{-3}$	$0.216\,90\times10^{-3}$	$0.114\,38\times10^{-5}$	$0.995\,64\times10^{-6}$
0.300 00	$0.366\,83\times10^{-3}$	$0.301\,63\times10^{-3}$	$0.157\,78\times10^{-5}$	$0.136\,57\times10^{-5}$
0.350 00	$0.469\,04\times10^{-3}$	$0.393\,53\times10^{-3}$	$0.203\,53\times10^{-5}$	$0.175\,57\times10^{-5}$
0.400 00	$0.570\,24\times10^{-3}$	$0.488\,85\times10^{-3}$	$0.249\,13\times10^{-5}$	$0.214\,79\times10^{-5}$
0.450 00	$0.665\,80\times10^{-3}$	$0.583\,69\times10^{-3}$	$0.292\,12\times10^{-5}$	$0.252\,58\times10^{-5}$
0.500 00	$0.751\,83\times10^{-3}$	$0.674\,21\times10^{-3}$	$0.330\,39\times10^{-5}$	$0.287\,46\times10^{-5}$
0.550 00	$0.825\,32\times10^{-3}$	$0.756\,88\times10^{-3}$	$0.362\,33\times10^{-5}$	$0.318\,20\times10^{-5}$
0.600 00	$0.884\,26\times10^{-3}$	$0.828\,72\times10^{-3}$	$0.386\,93\times10^{-5}$	$0.343\,89\times10^{-5}$
0.650 00	$0.927\,59\times10^{-3}$	$0.887\,41\times10^{-3}$	$0.403\,82\times10^{-5}$	$0.363\,90\times10^{-5}$
0.700 00	$0.955\,16\times10^{-3}$	$0.931\,47\times10^{-3}$	$0.413\,14\times10^{-5}$	$0.377\,95\times10^{-5}$
0.750 00	$0.967\,59\times10^{-3}$	$0.960\,20\times10^{-3}$	$0.415\,47\times10^{-5}$	$0.386\,02\times10^{-5}$
0.800 00	$0.966\,03\times10^{-3}$	$0.973\,71\times10^{-3}$	$0.411\,61\times10^{-5}$	$0.388\,33\times10^{-5}$
0.850 00	$0.952\,08\times10^{-3}$	$0.972\,77\times10^{-3}$	$0.402\,54\times10^{-5}$	$0.385\,32\times10^{-5}$
0.900 00	$0.927\,53\times10^{-3}$	$0.958\,70\times10^{-3}$	$0.389\,23\times10^{-5}$	$0.377\,57\times10^{-5}$
0.950 00	$0.894\,30\times10^{-3}$	$0.933\,18\times10^{-3}$	$0.372\,63\times10^{-5}$	$0.365\,76\times10^{-5}$
1.000 00	$0.854\,26\times10^{-3}$	$0.898\,15\times10^{-3}$	$0.353\,59\times10^{-5}$	$0.350\,61\times10^{-5}$
1.100 00	$0.760\,73\times10^{-3}$	$0.807\,51\times10^{-3}$	$0.311\,14\times10^{-5}$	$0.313\,20\times10^{-5}$
1.200 00	$0.659\,09\times10^{-3}$	$0.701\,85\times10^{-3}$	$0.266\,66\times10^{-5}$	$0.270\,78\times10^{-5}$
1.300 00	$0.558\,31\times10^{-3}$	$0.593\,40\times10^{-3}$	$0.223\,46\times10^{-5}$	$0.227\,71\times10^{-5}$
1.400 00	$0.464\,23\times10^{-3}$	$0.490\,62\times10^{-3}$	$0.183\,65\times10^{-5}$	$0.187\,07\times10^{-5}$
1.500 00	$0.380\,13\times10^{-3}$	$0.398\,47\times10^{-3}$	$0.148\,39\times10^{-5}$	$0.150\,72\times10^{-5}$
1.600 00	$0.307\,37\times10^{-3}$	$0.319\,10\times10^{-3}$	$0.118\,16\times10^{-5}$	$0.119\,47\times10^{-5}$
1.700 00	$0.245\,97\times10^{-3}$	$0.252\,76\times10^{-3}$	$0.929\,28\times10^{-6}$	$0.934\,52\times10^{-6}$
1.800 00	$0.195\,18\times10^{-3}$	$0.198\,54\times10^{-3}$	$0.723\,37\times10^{-6}$	$0.723\,12\times10^{-6}$
1.900 00	$0.153\,84\times10^{-3}$	$0.154\,99\times10^{-3}$	$0.558\,40\times10^{-6}$	$0.554\,76\times10^{-6}$
2.000 00	$0.120\,60\times10^{-3}$	$0.120\,45\times10^{-3}$	$0.428\,22\times10^{-6}$	$0.422\,78\times10^{-6}$
2.200 00	$0.732\,66\times10^{-4}$	$0.721\,59\times10^{-4}$	$0.248\,38\times10^{-6}$	$0.242\,25\times10^{-6}$
2.400 00	$0.440\,94\times10^{-4}$	$0.430\,50\times10^{-4}$	$0.142\,59\times10^{-6}$	$0.137\,52\times10^{-6}$
2.600 00	$0.264\,31\times10^{-4}$	$0.257\,12\times10^{-4}$	$0.815\,85\times10^{-7}$	$0.779\,46\times10^{-7}$
2.800 00	$0.158\,41\times10^{-4}$	$0.154\,25\times10^{-4}$	$0.467\,56\times10^{-7}$	$0.443\,61\times10^{-7}$
3.000 00	$0.951\,73\times10^{-5}$	$0.931\,24\times10^{-5}$	$0.269\,24\times10^{-7}$	$0.254\,44\times10^{-7}$
3.200 00	$0.574\,38\times10^{-5}$	$0.566\,48\times10^{-5}$	$0.156\,13\times10^{-7}$	$0.147\,44\times10^{-7}$
3.400 00	$0.348\,74\times10^{-5}$	$0.347\,47\times10^{-5}$	$0.913\,06\times10^{-8}$	$0.864\,41\times10^{-8}$
3.600 00	$0.213\,32\times10^{-5}$	$0.215\,06\times10^{-5}$	$0.539\,06\times10^{-8}$	$0.513\,15\times10^{-8}$
3.800 00	$0.131\,65\times10^{-5}$	$0.134\,42\times10^{-5}$	$0.321\,58\times10^{-8}$	$0.308\,60\times10^{-8}$
4.000 00	$0.821\,15\times10^{-6}$	$0.849\,32\times10^{-6}$	$0.194\,00\times10^{-8}$	$0.188\,05\times10^{-8}$

TABLE I. (*Continued*).

	$3s\sigma_g$			$3p\sigma_u$		$3p\pi_u$	
q	SCF	DJ	HS	DJ	HS	DJ	HS
0.000 00	$0.280\,18\times10^{-2}$	$0.285\,44\times10^{-2}$	$0.300\,38\times10^{-2}$	0.000 00	0.000 00	0.000 00	0.000 00
0.050 00	$0.277\,87\times10^{-2}$	$0.296\,57\times10^{-2}$	$0.311\,81\times10^{-2}$	$0.700\,15\times10^{-6}$	$0.696\,34\times10^{-6}$	$0.217\,62\times10^{-9}$	$0.224\,34\times10^{-9}$
0.100 00	$0.271\,28\times10^{-2}$	$0.288\,71\times10^{-2}$	$0.303\,95\times10^{-2}$	$0.272\,25\times10^{-5}$	$0.273\,70\times10^{-5}$	$0.850\,57\times10^{-9}$	$0.878\,29\times10^{-9}$
0.150 00	$0.260\,63\times10^{-2}$	$0.276\,07\times10^{-2}$	$0.291\,25\times10^{-2}$	$0.597\,47\times10^{-5}$	$0.602\,30\times10^{-5}$	$0.188\,21\times10^{-8}$	$0.193\,32\times10^{-8}$
0.200 00	$0.246\,59\times10^{-2}$	$0.259\,52\times10^{-2}$	$0.274\,54\times10^{-2}$	$0.102\,54\times10^{-4}$	$0.103\,81\times10^{-4}$	$0.326\,28\times10^{-8}$	$0.333\,22\times10^{-8}$
0.250 00	$0.229\,90\times10^{-2}$	$0.240\,02\times10^{-2}$	$0.254\,72\times10^{-2}$	$0.153\,17\times10^{-4}$	$0.155\,95\times10^{-4}$	$0.492\,69\times10^{-8}$	$0.500\,45\times10^{-8}$
0.300 00	$0.211\,36\times10^{-2}$	$0.218\,61\times10^{-2}$	$0.232\,79\times10^{-2}$	$0.208\,83\times10^{-4}$	$0.214\,15\times10^{-4}$	$0.678\,94\times10^{-8}$	$0.686\,79\times10^{-8}$
0.350 00	$0.191\,78\times10^{-2}$	$0.196\,29\times10^{-2}$	$0.209\,75\times10^{-2}$	$0.266\,61\times10^{-4}$	$0.275\,78\times10^{-4}$	$0.875\,05\times10^{-8}$	$0.883\,45\times10^{-8}$
0.400 00	$0.171\,92\times10^{-2}$	$0.173\,96\times10^{-2}$	$0.186\,52\times10^{-2}$	$0.323\,65\times10^{-4}$	$0.338\,17\times10^{-4}$	$0.107\,04\times10^{-7}$	$0.108\,16\times10^{-7}$
0.450 00	$0.152\,41\times10^{-2}$	$0.152\,38\times10^{-2}$	$0.163\,88\times10^{-2}$	$0.377\,38\times10^{-4}$	$0.398\,81\times10^{-4}$	$0.125\,48\times10^{-7}$	$0.127\,29\times10^{-7}$
0.500 00	$0.133\,76\times10^{-2}$	$0.132\,10\times10^{-2}$	$0.142\,43\times10^{-2}$	$0.425\,63\times10^{-4}$	$0.455\,41\times10^{-4}$	$0.141\,92\times10^{-7}$	$0.144\,98\times10^{-7}$
0.550 00	$0.116\,33\times10^{-2}$	$0.113\,50\times10^{-2}$	$0.122\,63\times10^{-2}$	$0.466\,73\times10^{-4}$	$0.506\,01\times10^{-4}$	$0.155\,70\times10^{-7}$	$0.160\,64\times10^{-7}$
0.600 00	$0.100\,36\times10^{-2}$	$0.967\,72\times10^{-3}$	$0.104\,72\times10^{-2}$	$0.499\,59\times10^{-4}$	$0.549\,06\times10^{-4}$	$0.166\,39\times10^{-7}$	$0.173\,78\times10^{-7}$
0.650 00	$0.859\,85\times10^{-3}$	$0.819\,97\times10^{-3}$	$0.888\,29\times10^{-3}$	$0.523\,63\times10^{-4}$	$0.583\,41\times10^{-4}$	$0.173\,81\times10^{-7}$	$0.184\,09\times10^{-7}$
0.700 00	$0.732\,19\times10^{-3}$	$0.691\,39\times10^{-3}$	$0.749\,46\times10^{-3}$	$0.538\,80\times10^{-4}$	$0.608\,41\times10^{-4}$	$0.178\,02\times10^{-7}$	$0.191\,41\times10^{-7}$
0.750 00	$0.620\,27\times10^{-3}$	$0.580\,86\times10^{-3}$	$0.629\,81\times10^{-3}$	$0.545\,42\times10^{-4}$	$0.623\,84\times10^{-4}$	$0.179\,23\times10^{-7}$	$0.195\,73\times10^{-7}$
0.800 00	$0.523\,16\times10^{-3}$	$0.486\,84\times10^{-3}$	$0.527\,84\times10^{-3}$	$0.544\,19\times10^{-4}$	$0.629\,89\times10^{-4}$	$0.177\,79\times10^{-7}$	$0.197\,14\times10^{-7}$
0.850 00	$0.439\,65\times10^{-3}$	$0.407\,53\times10^{-3}$	$0.441\,73\times10^{-3}$	$0.535\,98\times10^{-4}$	$0.627\,13\times10^{-4}$	$0.174\,08\times10^{-7}$	$0.195\,86\times10^{-7}$
0.900 00	$0.368\,39\times10^{-3}$	$0.341\,07\times10^{-3}$	$0.369\,51\times10^{-3}$	$0.521\,81\times10^{-4}$	$0.616\,41\times10^{-4}$	$0.168\,52\times10^{-7}$	$0.192\,16\times10^{-7}$
0.950 00	$0.307\,96\times10^{-3}$	$0.285\,65\times10^{-3}$	$0.309\,28\times10^{-3}$	$0.502\,78\times10^{-4}$	$0.598\,77\times10^{-4}$	$0.161\,51\times10^{-7}$	$0.186\,39\times10^{-7}$
1.000 00	$0.256\,99\times10^{-3}$	$0.239\,59\times10^{-3}$	$0.259\,21\times10^{-3}$	$0.479\,93\times10^{-4}$	$0.575\,40\times10^{-4}$	$0.153\,43\times10^{-7}$	$0.178\,90\times10^{-7}$
1.100 00	$0.178\,32\times10^{-3}$	$0.169\,69\times10^{-3}$	$0.183\,23\times10^{-3}$	$0.426\,77\times10^{-4}$	$0.516\,29\times10^{-4}$	$0.135\,27\times10^{-7}$	$0.160\,20\times10^{-7}$
1.200 00	$0.123\,46\times10^{-3}$	$0.121\,56\times10^{-3}$	$0.130\,91\times10^{-3}$	$0.369\,22\times10^{-4}$	$0.448\,26\times10^{-4}$	$0.116\,13\times10^{-7}$	$0.138\,81\times10^{-7}$
1.300 00	$0.855\,01\times10^{-4}$	$0.881\,42\times10^{-4}$	$0.946\,00\times10^{-4}$	$0.312\,35\times10^{-4}$	$0.378\,79\times10^{-4}$	$0.974\,54\times10^{-8}$	$0.116\,95\times10^{-7}$
1.400 00	$0.593\,29\times10^{-4}$	$0.646\,30\times10^{-4}$	$0.690\,82\times10^{-4}$	$0.259\,45\times10^{-4}$	$0.313\,11\times10^{-4}$	$0.801\,75\times10^{-8}$	$0.962\,10\times10^{-8}$
1.500 00	$0.413\,03\times10^{-4}$	$0.478\,29\times10^{-4}$	$0.508\,96\times10^{-4}$	$0.212\,32\times10^{-4}$	$0.254\,29\times10^{-4}$	$0.648\,19\times10^{-8}$	$0.775\,66\times10^{-8}$
1.600 00	$0.288\,75\times10^{-4}$	$0.356\,43\times10^{-4}$	$0.377\,61\times10^{-4}$	$0.171\,65\times10^{-4}$	$0.203\,67\times10^{-4}$	$0.516\,11\times10^{-8}$	$0.614\,78\times10^{-8}$
1.700 00	$0.202\,84\times10^{-4}$	$0.266\,91\times10^{-4}$	$0.281\,64\times10^{-4}$	$0.137\,43\times10^{-4}$	$0.161\,38\times10^{-4}$	$0.405\,56\times10^{-8}$	$0.480\,36\times10^{-8}$
1.800 00	$0.143\,25\times10^{-4}$	$0.200\,52\times10^{-4}$	$0.210\,89\times10^{-4}$	$0.109\,16\times10^{-4}$	$0.126\,85\times10^{-4}$	$0.315\,12\times10^{-8}$	$0.370\,90\times10^{-8}$
1.900 00	$0.101\,75\times10^{-4}$	$0.150\,95\times10^{-4}$	$0.158\,39\times10^{-4}$	$0.861\,71\times10^{-5}$	$0.991\,20\times10^{-5}$	$0.242\,56\times10^{-8}$	$0.283\,61\times10^{-8}$
2.000 00	$0.727\,02\times10^{-5}$	$0.113\,79\times10^{-4}$	$0.119\,24\times10^{-4}$	$0.676\,92\times10^{-5}$	$0.771\,41\times10^{-5}$	$0.185\,26\times10^{-8}$	$0.215\,17\times10^{-8}$
2.200 00	$0.378\,30\times10^{-5}$	$0.648\,40\times10^{-5}$	$0.679\,88\times10^{-5}$	$0.413\,48\times10^{-5}$	$0.464\,23\times10^{-5}$	$0.106\,23\times10^{-8}$	$0.121\,74\times10^{-8}$
2.400 00	$0.202\,27\times10^{-5}$	$0.371\,07\times10^{-5}$	$0.390\,69\times10^{-5}$	$0.250\,55\times10^{-5}$	$0.278\,89\times10^{-5}$	$0.600\,25\times10^{-9}$	$0.679\,26\times10^{-9}$
2.600 00	$0.111\,43\times10^{-5}$	$0.213\,79\times10^{-5}$	$0.226\,59\times10^{-5}$	$0.151\,36\times10^{-5}$	$0.168\,17\times10^{-5}$	$0.336\,86\times10^{-9}$	$0.376\,95\times10^{-9}$
2.800 00	$0.634\,60\times10^{-6}$	$0.124\,45\times10^{-5}$	$0.132\,95\times10^{-5}$	$0.914\,76\times10^{-6}$	$0.102\,12\times10^{-5}$	$0.188\,86\times10^{-9}$	$0.209\,46\times10^{-9}$
3.000 00	$0.374\,86\times10^{-6}$	$0.734\,89\times10^{-6}$	$0.791\,64\times10^{-6}$	$0.554\,42\times10^{-6}$	$0.625\,52\times10^{-6}$	$0.106\,23\times10^{-9}$	$0.117\,14\times10^{-9}$
3.200 00	$0.230\,29\times10^{-6}$	$0.442\,15\times10^{-6}$	$0.479\,96\times10^{-6}$	$0.337\,58\times10^{-6}$	$0.386\,85\times10^{-6}$	$0.601\,41\times10^{-10}$	$0.661\,93\times10^{-10}$
3.400 00	$0.147\,30\times10^{-6}$	$0.272\,16\times10^{-6}$	$0.297\,31\times10^{-6}$	$0.206\,78\times10^{-6}$	$0.241\,63\times10^{-6}$	$0.343\,47\times10^{-10}$	$0.378\,99\times10^{-10}$
3.600 00	$0.979\,84\times10^{-7}$	$0.171\,99\times10^{-6}$	$0.188\,75\times10^{-6}$	$0.127\,57\times10^{-6}$	$0.152\,46\times10^{-6}$	$0.198\,24\times10^{-10}$	$0.220\,28\times10^{-10}$
3.800 00	$0.675\,43\times10^{-7}$	$0.111\,85\times10^{-6}$	$0.123\,09\times10^{-6}$	$0.793\,70\times10^{-7}$	$0.971\,85\times10^{-7}$	$0.115\,82\times10^{-10}$	$0.130\,14\times10^{-10}$
4.000 00	$0.479\,95\times10^{-7}$	$0.749\,04\times10^{-7}$	$0.825\,25\times10^{-7}$	$0.498\,74\times10^{-7}$	$0.626\,17\times10^{-7}$	$0.685\,74\times10^{-11}$	$0.782\,10\times10^{-11}$

TABLE II. Total momentum density of $H_2(^1\Sigma_g^+)$ at $q=0$.

Wave function	Binding energy (eV)	$\Pi(0)$
Minimum basis set	3.17	1.1474
CI Das and Wahl (Ref. 16)	4.62	1.2299
SCF DJ	3.63	1.2205
CI DJ	4.71	1.1724
CI HS	4.71	1.1490
CI Liu (Ref. 17)	4.72	1.1636
CI Liu (vib. av.)	4.74	1.1990
Expt. (Refs. 5 and 15)	4.74	1.254±0.251

bitals $\psi_k(\mathbf{r},R)$,

$$\Psi_i(1,2)=\sum_k \mu_k \psi_k(\mathbf{r}_1,R)\psi_k(\mathbf{r}_2,R)\ , \tag{3}$$

and Ψ_f is the exact BO wave function[12] of the final residual ion (H_2^+) in the ground or excited state. The H_2^+ wave functions were calculated for the present study by the method of Bates, Ledsham, and Stewart[12] with more significant figures than in the original tabulation.[12]

The (spherically averaged) momentum density function corresponding to the overlap function $F_{if}(\mathbf{q})$ is

TABLE III. The sum of the $O(q)$ for the transitions to the $n=2$ and $n=3$ ion states.

	$n=2$		$n=3$	
q	DJ	HS	DJ	HS
0.000 00	$0.290\,34\times10^{-1}$	$0.281\,54\times10^{-1}$	$0.285\,44\times10^{-2}$	$0.300\,38\times10^{-2}$
0.050 00	$0.297\,80\times10^{-1}$	$0.289\,28\times10^{-1}$	$0.296\,64\times10^{-2}$	$0.311\,88\times10^{-2}$
0.100 00	$0.290\,53\times10^{-1}$	$0.282\,44\times10^{-1}$	$0.288\,99\times10^{-2}$	$0.304\,22\times10^{-2}$
0.150 00	$0.278\,83\times10^{-1}$	$0.271\,41\times10^{-1}$	$0.276\,67\times10^{-2}$	$0.291\,86\times10^{-2}$
0.200 00	$0.263\,48\times10^{-1}$	$0.256\,88\times10^{-1}$	$0.260\,55\times10^{-2}$	$0.275\,58\times10^{-2}$
0.250 00	$0.245\,35\times10^{-1}$	$0.239\,65\times10^{-1}$	$0.241\,55\times10^{-2}$	$0.256\,28\times10^{-2}$
0.300 00	$0.225\,37\times10^{-1}$	$0.220\,56\times10^{-1}$	$0.220\,69\times10^{-2}$	$0.234\,94\times10^{-2}$
0.350 00	$0.204\,47\times10^{-1}$	$0.200\,48\times10^{-1}$	$0.198\,95\times10^{-2}$	$0.212\,51\times10^{-2}$
0.400 00	$0.183\,45\times10^{-1}$	$0.180\,20\times10^{-1}$	$0.177\,20\times10^{-2}$	$0.189\,90\times10^{-2}$
0.450 00	$0.163\,01\times10^{-1}$	$0.160\,38\times10^{-1}$	$0.156\,16\times10^{-2}$	$0.167\,87\times10^{-2}$
0.550 00	$0.143\,65\times10^{-1}$	$0.141\,54\times10^{-1}$	$0.136\,36\times10^{-2}$	$0.146\,99\times10^{-2}$
0.550 00	$0.125\,73\times10^{-1}$	$0.124\,05\times10^{-1}$	$0.118\,17\times10^{-2}$	$0.127\,69\times10^{-2}$
0.600 00	$0.109\,44\times10^{-1}$	$0.108\,13\times10^{-1}$	$0.101\,77\times10^{-2}$	$0.110\,21\times10^{-2}$
0.650 00	$0.948\,74\times10^{-2}$	$0.938\,69\times10^{-2}$	$0.872\,35\times10^{-3}$	$0.946\,65\times10^{-3}$
0.700 00	$0.820\,06\times10^{-2}$	$0.812\,68\times10^{-2}$	$0.745\,29\times10^{-3}$	$0.810\,33\times10^{-3}$
0.750 00	$0.707\,56\times10^{-2}$	$0.702\,49\times10^{-2}$	$0.635\,43\times10^{-3}$	$0.692\,22\times10^{-3}$
0.800 00	$0.609\,98\times10^{-2}$	$0.606\,91\times10^{-2}$	$0.541\,28\times10^{-3}$	$0.590\,86\times10^{-3}$
0.850 00	$0.525\,84\times10^{-2}$	$0.524\,45\times10^{-2}$	$0.461\,15\times10^{-3}$	$0.504\,46\times10^{-3}$
0.900 00	$0.453\,58\times10^{-2}$	$0.453\,58\times10^{-2}$	$0.393\,27\times10^{-3}$	$0.431\,18\times10^{-3}$
0.950 00	$0.391\,68\times10^{-2}$	$0.302\,76\times10^{-2}$	$0.335\,94\times10^{-3}$	$0.369\,18\times10^{-3}$
1.000 00	$0.338\,72\times10^{-2}$	$0.340\,59\times10^{-2}$	$0.287\,60\times10^{-3}$	$0.316\,77\times10^{-3}$
1.100 00	$0.254\,56\times10^{-2}$	$0.257\,24\times10^{-2}$	$0.212\,38\times20^{-3}$	$0.234\,88\times10^{-3}$
1.200 00	$0.192\,58\times10^{-2}$	$0.195\,29\times10^{-2}$	$0.158\,49\times10^{-3}$	$0.175\,76\times10^{-3}$
1.300 00	$0.146\,50\times10^{-2}$	$0.148\,78\times10^{-2}$	$0.119\,39\times10^{-3}$	$0.132\,50\times10^{-3}$
1.400 00	$0.111\,89\times10^{-2}$	$0.113\,57\times10^{-2}$	$0.905\,86\times10^{-4}$	$0.100\,41\times10^{-3}$
1.500 00	$0.856\,66\times10^{-3}$	$0.867\,46\times10^{-3}$	$0.690\,70\times10^{-4}$	$0.763\,39\times10^{-4}$
1.600 00	$0.656\,58\times10^{-3}$	$0.662\,47\times10^{-3}$	$0.528\,16\times10^{-4}$	$0.581\,39\times10^{-4}$
1.700 00	$0.503\,30\times10^{-3}$	$0.505\,65\times10^{-3}$	$0.404\,40\times10^{-4}$	$0.443\,12\times10^{-4}$
1.800 00	$0.385\,66\times10^{-3}$	$0.385\,72\times10^{-3}$	$0.309\,74\times10^{-4}$	$0.337\,82\times10^{-4}$
1.900 00	$0.295\,33\times10^{-3}$	$0.294\,10\times10^{-3}$	$0.237\,17\times10^{-4}$	$0.257\,58\times10^{-4}$
2.000 00	$0.226\,01\times10^{-3}$	$0.224\,22\times10^{-3}$	$0.181\,52\times10^{-4}$	$0.196\,44\times10^{-4}$
2.200 00	$0.132\,24\times10^{-3}$	$0.130\,46\times10^{-3}$	$0.106\,21\times10^{-4}$	$0.114\,45\times10^{-4}$
2.400 00	$0.774\,34\times10^{-4}$	$0.762\,13\times10^{-4}$	$0.621\,78\times10^{-5}$	$0.669\,80\times10^{-5}$
2.600 00	$0.455\,11\times10^{-4}$	$0.448\,27\times10^{-4}$	$0.365\,25\times10^{-5}$	$0.394\,90\times10^{-5}$
2.800 00	$0.269\,25\times10^{-4}$	$0.266\,10\times10^{-4}$	$0.215\,99\times10^{-5}$	$0.235\,16\times10^{-5}$
3.000 00	$0.160\,79\times10^{-4}$	$0.159\,76\times10^{-4}$	$0.128\,97\times10^{-5}$	$0.141\,78\times10^{-5}$
3.200 00	$0.971\,58\times10^{-5}$	$0.971\,89\times10^{-5}$	$0.779\,98\times10^{-6}$	$0.867\,24\times10^{-6}$
3.400 00	$0.595\,51\times10^{-5}$	$0.600\,11\times10^{-5}$	$0.479\,09\times10^{-6}$	$0.539\,24\times10^{-6}$
3.600 00	$0.371\,07\times10^{-5}$	$0.376\,73\times10^{-5}$	$0.299\,65\times10^{-6}$	$0.341\,41\times10^{-6}$
3.800 00	$0.235\,55\times10^{-5}$	$0.240\,81\times10^{-5}$	$0.191\,28\times10^{-6}$	$0.220\,41\times10^{-6}$
4.000 00	$0.152\,60\times10^{-5}$	$0.156\,95\times10^{-5}$	$0.124\,82\times10^{-6}$	$0.145\,24\times10^{-6}$

TABLE IV. The $n=2$ to $n=1$ and $n=3$ to $n=1$ cross-section ratios.

q	HS $\sigma(n=2)/\sigma(n=1)$	HS $\sigma(n=3)/\sigma(n=1)$	DJ $\sigma(n=2)/\sigma(n=1)$	DJ $\sigma(n=3)/\sigma(n=1)$
0.000 00	$0.253\,25\times10^{-1}$	$0.270\,19\times10^{-2}$	$0.255\,91\times10^{-1}$	$0.251\,59\times10^{-2}$
0.050 00	$0.262\,23\times10^{-1}$	$0.282\,72\times10^{-2}$	$0.264\,62\times10^{-1}$	$0.262\,59\times10^{-2}$
0.100 00	$0.261\,91\times10^{-1}$	$0.282\,10\times10^{-2}$	$0.264\,33\times10^{-1}$	$0.262\,92\times10^{-2}$
0.150 00	$0.261\,37\times10^{-1}$	$0.281\,06\times10^{-2}$	$0.263\,87\times10^{-1}$	$0.261\,82\times10^{-2}$
0.200 00	$0.260\,70\times10^{-1}$	$0.279\,67\times10^{-2}$	$0.263\,29\times10^{-1}$	$0.260\,36\times10^{-2}$
0.250 00	$0.259\,93\times10^{-1}$	$0.277\,98\times10^{-2}$	$0.262\,67\times10^{-1}$	$0.258\,61\times10^{-2}$
0.300 00	$0.259\,16\times10^{-1}$	$0.276\,06\times10^{-2}$	$0.262\,09\times10^{-1}$	$0.256\,65\times10^{-2}$
0.350 00	$0.258\,49\times10^{-1}$	$0.274\,00\times10^{-2}$	$0.261\,63\times10^{-1}$	$0.254\,58\times10^{-2}$
0.400 00	$0.258\,01\times10^{-1}$	$0.271\,91\times20^{-2}$	$0.261\,41\times10^{-1}$	$0.252\,50\times10^{-2}$
0.450 00	$0.257\,88\times10^{-1}$	$0.269\,92\times10^{-2}$	$0.261\,53\times10^{-1}$	$0.250\,54\times10^{-2}$
0.500 00	$0.258\,20\times10^{-1}$	$0.268\,14\times10^{-2}$	$0.262\,09\times10^{-1}$	$0.248\,79\times10^{-2}$
0.550 00	$0.259\,14\times10^{-1}$	$0.266\,75\times10^{-2}$	$0.263\,22\times10^{-1}$	$0.247\,39\times10^{-2}$
0.600 00	$0.260\,84\times10^{-1}$	$0.265\,87\times10^{-2}$	$0.265\,01\times10^{-1}$	$0.246\,43\times10^{-2}$
0.650 00	$0.263\,43\times10^{-1}$	$0.265\,67\times10^{-2}$	$0.267\,58\times10^{-1}$	$0.246\,04\times10^{-2}$
0.700 00	$0.267\,06\times10^{-1}$	$0.266\,29\times10^{-2}$	$0.271\,03\times10^{-1}$	$0.246\,32\times10^{-2}$
0.750 00	$0.271\,86\times10^{-1}$	$0.267\,88\times10^{-2}$	$0.275\,47\times10^{-1}$	$0.247\,39\times10^{-2}$
0.800 00	$0.277\,91\times10^{-1}$	$0.270\,56\times10^{-2}$	$0.280\,98\times10^{-1}$	$0.249\,33\times10^{-2}$
0.850 00	$0.285\,32\times10^{-1}$	$0.274\,45\times10^{-2}$	$0.287\,64\times10^{-1}$	$0.252\,26\times10^{-2}$
0.900 00	$0.294\,14\times10^{-1}$	$0.279\,62\times10^{-2}$	$0.295\,54\times10^{-1}$	$0.256\,24\times10^{-2}$
0.950 00	$0.304\,42\times10^{-1}$	$0.286\,14\times10^{-2}$	$0.304\,72\times10^{-1}$	$0.261\,36\times10^{-2}$
1.000 00	$0.316\,15\times10^{-1}$	$0.294\,04\times10^{-2}$	$0.315\,23\times10^{-1}$	$0.267\,65\times10^{-2}$
1.100 00	$0.343\,89\times10^{-1}$	$0.314\,00\times10^{-2}$	$0.340\,32\times10^{-1}$	$0.283\,93\times10^{-2}$
1.200 00	$0.376\,94\times10^{-1}$	$0.339\,24\times20^{-2}$	$0.370\,78\times10^{-1}$	$0.305\,15\times10^{-2}$
1.300 00	$0.414\,57\times10^{-1}$	$0.369\,19\times10^{-2}$	$0.406\,28\times10^{-1}$	$0.331\,10\times10^{-2}$
1.400 00	$0.455\,84\times10^{-1}$	$0.403\,02\times20^{-2}$	$0.446\,24\times10^{-1}$	$0.361\,27\times10^{-2}$
1.500 00	$0.499\,75\times10^{-1}$	$0.439\,80\times10^{-2}$	$0.489\,85\times10^{-1}$	$0.394\,96\times10^{-2}$
1.600 00	$0.545\,31\times10^{-1}$	$0.478\,57\times10^{-2}$	$0.536\,21\times10^{-1}$	$0.431\,33\times10^{-2}$
1.700 00	$0.591\,59\times10^{-1}$	$0.518\,43\times10^{-2}$	$0.584\,32\times10^{-1}$	$0.469\,50\times10^{-2}$
1.800 00	$0.637\,78\times10^{-1}$	$0.558\,58\times10^{-2}$	$0.633\,23\times10^{-1}$	$0.508\,57\times10^{-2}$
1.900 00	$0.683\,18\times10^{-1}$	$0.598\,33\times10^{-2}$	$0.682\,00\times10^{-1}$	$0.547\,69\times10^{-2}$
2.000 00	$0.727\,16\times10^{-1}$	$0.637\,06\times10^{-2}$	$0.729\,77\times10^{-1}$	$0.586\,09\times10^{-2}$
2.200 00	$0.808\,75\times10^{-1}$	$0.709\,47\times10^{-2}$	$0.819\,11\times10^{-1}$	$0.657\,90\times10^{-2}$
2.400 00	$0.878\,55\times10^{-1}$	$0.772\,11\times10^{-2}$	$0.895\,42\times10^{-1}$	$0.719\,00\times10^{-2}$
2.600 00	$0.932\,60\times10^{-1}$	$0.821\,58\times10^{-2}$	$0.953\,38\times10^{-1}$	$0.765\,14\times10^{-2}$
2.800 00	$0.967\,01\times10^{-1}$	$0.854\,57\times10^{-2}$	$0.988\,34\times10^{-1}$	$0.792\,81\times10^{-2}$
3.000 00	$0.978\,58\times10^{-1}$	$0.868\,43\times10^{-2}$	$0.997\,00\times10^{-1}$	$0.799\,72\times10^{-2}$
3.200 00	$0.966\,07\times10^{-1}$	$0.862\,05\times10^{-2}$	$0.978\,69\times10^{-1}$	$0.785\,69\times10^{-2}$
3.400 00	$0.931\,27\times10^{-1}$	$0.836\,81\times10^{-2}$	$0.936\,30\times10^{-1}$	$0.753\,26\times10^{-2}$
3.600 00	$0.879\,12\times10^{-1}$	$0.796\,71\times10^{-2}$	$0.876\,17\times10^{-1}$	$0.707\,54\times10^{-2}$
3.800 00	$0.816\,66\times10^{-1}$	$0.747\,48\times10^{-2}$	$0.806\,55\times10^{-1}$	$0.654\,95\times10^{-2}$
4.000 00	$0.751\,19\times10^{-1}$	$0.695\,16\times10^{-2}$	$0.735\,48\times10^{-1}$	$0.601\,60\times10^{-2}$

$$O_{if}(q)=\int d\hat{q}\,|F_{if}(q)|^2=\sum_k S_{kf}^2(R)\lambda_k\Pi_k(q)\,, \tag{4}$$

where $S_{kf}(R)$ is the overlap integral between the residual ion in state Ψ_f and the kth natural orbital of the molecule at the internuclear distance R. It is also known as a spectroscopic factor according to the Franck-Condon (FC) principle, and can be written as

$$S_{kf}(R)=\int \psi_k(\mathbf{r},R)\Psi_f(\mathbf{r},R)\,. \tag{5}$$

In Eq. (4) λ_k $(=|\mu_k|^2)$ is the occupation number for the kth natural orbital $\psi_k(\mathbf{r},R)$ of the molecule and $\Pi_k(q,R)$ is the (spherically averaged) momentum density function for the kth natural orbital.

The $\Pi_k(q,R)$,

$$\Pi_k(q,R)=\int d\hat{q}\,|\chi_k(\mathbf{q},R)|^2\,, \tag{6}$$

where $\chi_k(\mathbf{q},R)$ is the Dirac-Fourier transform[13] of $\psi_k(\mathbf{r},R)$, were calculated by the previously developed computational technique[14] for wave functions based on elliptical-type basis functions. The SCF and CI functions of Davidson and Jones[10] and the CI function of Hagstrom and Shull[11] are of this type. The values of the $O_{if}(q)$ for excitation to the ground and various excited states of the residual ion are presented in Table I. It should be noted that the united-atom notation for the states of the molecule and its residual ion is used throughout this paper. Atomic units are used in the tables unless another unit is explicitly indicated.

From Table I, it is interesting to note that electron

correlation is not very important for the ground-state transition. The difference between the values calculated by using the CI and SCF wave functions of Davidson and Jones[10] (DJ) is 5% at $q=0$ and decreases to 3% at $q=0.3$ a.u. The differences are negligible for q greater than 1 a.u. It is also seen that the differences between the values calculated by using the SCF and CI DJ wave functions are also very small for the transitions to the $2s\sigma_g$ and $3s\sigma_g$ states of H_2^+. The results calculated by using the CI wave function of Hagstrom and Shull[11] (HS) which contains about the same amount of correlation energy are shown in Table I for comparison with those calculated from the CI DJ wave function. The two sets of CI values differ by less than 2%.

In order to ascertain the accuracy of the momentum densities calculated from the various wave functions, the total spherically averaged momentum density function $[\Pi(q)=\sum_k \Pi_k(q)]$ for H_2 at $q=0$ were calculated and are presented in Table II for comparison with each other and with the values extracted from high-energy electron scattering data.[5,15] Our calculated difference for $\Pi(0)$ between the CI Liu wave function[17] and the CI DJ wave function is less than 1%, although the binding energies differ by 2%. It is believed that the present theoretical values computed from the CI DJ wave function for the ground-state transition should be accurate enough to distinguish among the various experimental results.

It is not surprising that electron correlation in the ground-state wave function is extremely important for excitation of the molecular ion to an excited state when the angular momentum quantum number l of the molecular ion is greater than zero. The difference between the values calculated by employing the CI DJ and CI HS wave functions is 15% for $2p\sigma_u$ and 13% for $2p\pi_u$ in the small recoil momentum region and decreases to a few percent in the larger recoil momentum region. The sum of the cross sections leading to the $n=2$ and 3 ion states were calculated and are given in Table III. The $n=2$ to $n=1$ and $n=3$ to $n=1$ cross-section ratios are given as well in Table IV, where n is the molecular ion principal quantum number.

III. COMPARISON WITH EXPERIMENTS

At the incident energy of 1200, Cook *et al.*[4] have shown that the PWIA is unable to account for the $(e,2e)$ cross section for He at recoil momenta greater than 1.2 a.u. This is because the distortion of incoming and outgoing electron waves plays a significant role in the region where target electrons are near the nucleus. A more realistic theoretical model to describe the collision process is needed. In the present work we are more interested in the region where q is small, i.e., the target electrons are far out from the nucleus and where electron correlation may be most important.

Since all experimental data are reported on a relative scale and since the experimental data for Weigold *et al.*[18] and Leung and Brion[19] for the ground-state transition are identical, only data from Weigold *et al.*[18] and Migdall *et al.*[9] are shown in Fig. 1. All the theoretical values and the experimental data are scaled relatively at $q=0.2$ a.u. to the experimental raw data provided by Weigold (private communication). In Fig. 1 the relative scale comparison between theory and experiment is shown for recoil momenta q in the range between 0.11 and 1.0 a.u. The agreement is good in this region. As expected, disagreement due to distortions appears when q is greater than 1 a.u. Since experimental values are not available for q smaller than 0.11 a.u. and the Weigold data at $q=0.11$ a.u. were measured at 300 eV with large experimental uncertainty, it is not known whether or not the agreement between theory and experiment should remain as good for recoil momenta smaller than 0.11 a.u. Therefore, it is desirable to obtain experimental data at higher incident electron energies around $q=0.05$ a.u. in order to compare with the present theoretical values.

Inasmuch as it has been shown[6,8] that consideration of molecular vibration is important in the comparison between theory and experiment for the Compton profile [and the underlying $\Pi(\mathbf{q})$] for H_2 and D_2, theoretical calculations including vibrational averaging are in progress in this laboratory using the CI Liu wave functions. The results will provide better theoretical values for a proper consideration of FC distribution in the smaller recoil momentum region.

Weigold *et al.*[18] also reported results for transitions to three excited states, i.e., $2s\sigma_g$, $2p\sigma_u$, and $2p\pi_u$. In Fig. 2 the reaction cross sections for the transition to these three excited states are shown relative to the present theoretical values. The agreement is not as good as reported for the ground-state transition. Since the theoretical values listed in Table I show that the contribution from $2p\pi_u$ is quite small as compared with $2s\sigma_g$, it may be difficult to measure this transition cross section very accurately. The experimental value for the $n=2$ to $n=1$ cross-section ratio for $0.37\geq q\geq 0.27$ a.u. and $0.67\geq q\geq 0.65$ a.u. are 0.023 ± 0.0015 and 0.038 ± 0.002, respectively. The experimental value for $0.37\geq q\geq 0.27$ a.u. is smaller by about 6% than the theoretical value, while for $0.67\geq q\geq 0.65$ a.u. the experimental is larger than the theoretical value by 30%. The absolute cross-section ratio for $1s\sigma_g$:$2s\sigma_g$:$2p\pi_u$:$2p\sigma_u$ at $q=0.3$ a.u. calculated from the CI DJ wave function is 1:0.016:0.002:0.005. It is surpris-

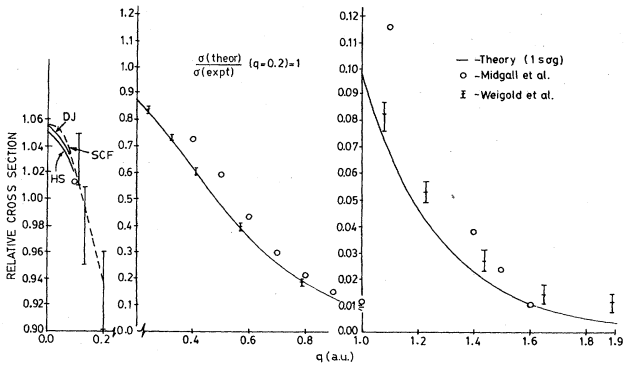

FIG. 1. The $(e,2e)$ noncoplanar differential cross section at incident energies from 300 to 1200 eV plotted as a function of the recoil momentum q. The theoretical values are scaled relative to the experimental values such that their ratio at $q=0.2$ a.u. is equal to 1, i.e., $[\sigma(\text{theor})/\sigma(\text{expt})](q=0.2\text{ a.u})=1$.

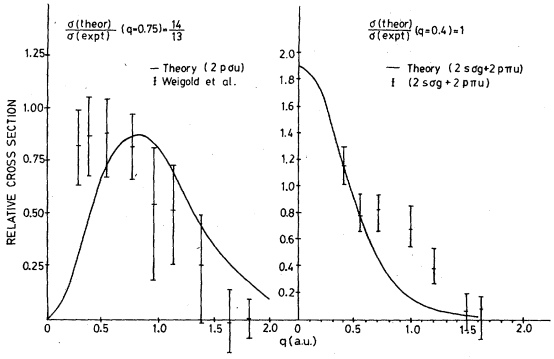

FIG. 2. The $(e,2e)$ noncoplanar differential cross section at 1200 eV plotted as a function of the recoil momentum q. The theoretical values for the CI DJ wave functions for the excited-state transitions are scaled relative to the experimental data taken from Ref. 12. The ratio $\sigma(\text{theor})/\sigma(\text{expt})$ at a particular recoil momentum for the transition is indicated in the figures.

ing to learn that the values for $2p\sigma_u$ and $2p\pi_u$ are underestimated too far by theory. If we assume that the overlap integrals between the molecule and its ion for the same symmetry and same quantum number are 1, and for either different symmetry or quantum number are zero, for example, $S_k(R)$ for $\langle 1s\sigma_g(H_2) \mid 1s\sigma_g(H_2^+)\rangle$ is 1, and for $\langle 1s\sigma_g(H_2) \mid 2s\sigma_g(H_2^+)\rangle$ is 0. Then the ratio is 1:0.0037:0.00011:0.0006 which is still far from agreement with experiment. This disagreement can be the result of the failure of the Franck-Condon principle in the theoretical model or the inadequacy of the ground-state wave function. It appears that these correlated ground-state wave functions can only qualitatively describe the excited-state transitions. It is clearly demonstrated that electron correlation as well as the overlap between the molecule and its ion play equally important roles in $(e,2e)$ reaction spectroscopy. In the present theoretical study, the overlap $S_{kf}(R)$ between the molecular wave function and the residual ion wave function for the $2p\pi_u$ transition is very small and is about 100 times smaller than the expected overlap integral.

IV. CONCLUSIONS

The present theoretical study of the $(e,2e)$ reaction on molecular hydrogen within the PWIA and the FC principle employed several highly electron correlated wave functions for transitions to the ground and all the $n=2$ and $n=3$ excited states of H_2^+. The results for the transition to the ground state are in moderately good agreement with experiment within experimental resolution. Nevertheless, it is desirable to have much higher-resolution (better than 1%) experimental data in order to distinguish among the present theoretical values for smaller q values. Experimental data for the transitions to all the $n=2$ and $n=3$ excited states are urgently needed in order to compare with the present theoretical values since there is only one experimental data set published for a limited number of excited-state transitions ($n=2$). To account for the excited-state transitions a more accurate ground-state wave function for H_2 may be needed and consideration of the possible breakdown of the FC principle for the excited-state transitions may also be necessary as well as consideration of distorted-wave effects. As mentioned earlier, calculations are in progress using the CI Liu wave functions in order to assess the role of molecular rotation and vibration. Since these wave functions are constructed from Slater-type orbitals on the two atomic sites (i.e., linear combination of atomic orbitals, instead of the ellipitical basis functions of the present wave functions, their use will permit an assessment to be made of the effect of the use of finite sets of basis functions of different types.

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