PHYSICAL REVIEW A VOLUME 11, NUMBER 4 APRIL 1975

Ab initio cross sections for excitation of the $2p$ state of hydrogen by electron impact at intermediate energies*

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(Received 15 October 1974)

Differential and integral cross sections for the $1s$-$2p$ excitation of the hydrogen atom at impact energies of 0.81, 1.00, 1.21, 1.44, 2.25, and 4.00 Ry are calculated from the most accurate available *ab initio* calculations and tabulated.

Electron–hydrogen-atom scattering provides an important test case because the most accurate *ab initio* electron-scattering calculations can be performed for this case. There is much interest lately in new experimental and approximate quantum mechanical methods for obtaining electron-scattering differential and integral cross sections at intermediate energies and it is useful to have accurate quantum mechanical results available for comparison.

The excitation of the $2p$ state is one of the most important processes in electron scattering by ground-state hydrogen. Experimental measurements of the integral cross section for this process were reviewed by Moiseiwitsch and Smith[1] and Williams and Willis.[2] The most accurate available results at the time of the former review were the relative measurement of Long *et al.*[3] Since then additional relative measurements with better energy resolution have been carried out by McGowan *et al.*[4] and these agree with the results of Long *et al.* within a few percent. Recently Williams and Willis[2] gave another review of previous work and measured an absolute value for this cross section of $0.867\,^{+0.082}_{-0.050}a_0^2$ ($1a_0 = 5.2918\times 10^{-11}$ m) at an impact energy of 0.81 Ry ($1\text{ Ry} = 2.1799\times 10^{-18}\text{ J} = 13.606$ eV) and suggested the previous experiments be normalized using this value. This has been done and the results are given as the experimental results in Table I. No experimental differential cross sections have been published.

The most accurate published calculations of reactance matrices for inelastic scattering are those performed by Burke and co-workers using the close-coupling approximation[5,6] and the correlation method[7,8] and those performed by Callaway and Wooten using algebraic variational methods.[9] These calculations assume LS coupling (where L and S are the quantum numbers for total orbital and spin angular momentum of the two-electron system) and have been carried out for both $S=0$ and $S=1$. They yield reactance matrices $\underline{\mathrm{R}}^{LS}$ from which differential cross sections $I(\overline{E},\Theta)$, partial integral cross sections, and integral cross sections $Q(E)$ may be obtained at each impact energy E using equations given elsewhere.[10–13] In some cases, however, the partial integral cross sections and/or integral cross sections are published but the reactance matrices are not published. In such cases there is not enough information published to calculate the differential cross section. Our experience with algebraic variational calculations on electron-hydrogen scattering has shown that the reactance matrices (and hence the differential cross sections) are more sensitive to the quality of the calculation than are the partial integral cross sections and integral cross sections,[14] and it is more difficult to judge the quality of a calculation when the reactance matrix is not published. The cross sections involve infinite sums over L, but in practice the reactance matrices are assumed to be nonzero only for $0 \leq L \leq L_{\max}$ where $L_{\max}$ should be large enough that the truncation error is small (see below).

Calculations of different quality are available at various energies. At low-enough energies the quality of the various calculations may be judged using a minimum principle[15] or a quasiminimum principle[16] for the eigenphase sums; in the present article this is useful only at an impact energy of 0.81 Ry. The most accurate calculations available for $L=0$ at 0.81, 1.0, and 1.21 Ry are those obtained by the correlation method[7] and by algebraic variational methods[9] and these calculations are in good agreement at the one energy (0.81 Ry) where comparison can be made.[9] The correlation-method reactance matrices are available[7] for $L=0$ at impact energies of 0.81, 1.00, and 1.21 Ry and were used in the calculations presented here. For higher L at 0.81 Ry calculations have been reported for $1 \leq L \leq 3$ using the six-state close-coupling method,[6] the correlation method,[8] and the three-state plus three-pseudostate close-coupling method[17] and the eigenphase sums have been compared for each symmetry (i.e., for each of the six sets of L and S). In one case the first method was best, in two cases the second method was best, and in three cases the third method was best. At energies 1 Ry and greater, calculations have not been performed using the second method for $L \geqslant 1$ and the eigenphase sums have not been published or compared. For $L \geqslant 1$ the reactance matrices have been made available only for the first method and so it was used for $L \geqslant 1$ for the present calculations. The six-state close-coupling reactance matrices are available for $L \leqslant 7$ at 0.81 and 1.00 Ry and for $L \leqslant 4$ at 1.21 Ry. For $L = 5$ at 1.21 Ry we used the three-state close-coupling results.[5] For $L > 7$ at 0.81 and 1.00 Ry and for $L > 5$ at 1.21 Ry no calculations more accurate than the Born II approximation[18] have been reported. For these L (up to $L = 13$) we calculated three-state Born-II-approximation[18] reactance matrices using a program described elsewhere.[19]

The calculations just described for 0.81 Ry, 1.00 Ry, and 1.21 Ry use the most accurate reactance matrices which have been made available at these energies and will be called the "present" calculations. The integral cross sections are given in Table I where they are compared with the three-state close-coupling approximation[5] (these results actually correspond to three-state close coupling for $0 \leqslant L \leqslant 5$ and to Born II for $6 \leqslant L \leqslant 13$). At 0.81 Ry, the partial integral cross sections from correlation-method calculations[8] and three-state plus three-pseudostate close-coupling method calculations[17] are available for $1 \leqslant L \leqslant 3$. These are substituted for the ones used in the present calculation to yield integral cross sections of $0.855 a_0^2$ and $0.93 a_0^2$, respectively. Comparison with Table I shows the former is in very good agreement with experiment but the smallness of the deviation is probably just fortuitous since combining the three methods by using the best partial cross section for each of these six symmetries (and treating $L = 0$ and $L \geqslant 4$ using the most accurate calculations which have been performed as in the present calculation) yields an integral cross section of $0.924 \times a_0^2$. These three estimates of the integral cross section are within the experimental error bounds.

For higher energies, calculations using the same methods are not available. We again used the most accurate available reactance matrices for low L supplemented by our own calculations at higher L. At impact energies of 1.44, 2.25, and 4.00 Ry we used the three-state close-coupling calculations[5] where available ($L \leqslant 6$ at impact energies of 1.44 and 2.25 Ry, $L \leqslant 7$ at an impact energy of 4.00 Ry). To supplement those calculations we calculated three-state Born-II-approximation reactance-matrix elements for intermediate L ($7 \leqslant L \leqslant 13$ at 1.44 Ry, $7 \leqslant L \leqslant 16$ at 2.25 Ry, and $8 \leqslant L \leqslant 22$ at 4.00 Ry) and Born-I-approximation[18] reactance-matrix elements for high L ($14 \leqslant L \leqslant 19$

TABLE I. Integral cross sections $Q(E)$ for $1s$-$2p$ transition as a function of impact energy E.

				$Q(a_0^2)$ Theory											
E (Ry)	L_1[a]	L_2[a]	Expt.	Present	low L[b]	$1s$-$2s$-$2p$ c.c.[c]	B	O[d]	VPS[d]	CM	DW	$1s$-$2p$ c.c.	$1s$-$2s$-$2p$ c.c. XE	BW	FM
0.81	7	13	0.87	0.996	0.996	1.101	1.81	1.68	0.85	0.90		1.53	3.83		
1.00	7	13	1.9	1.383	1.382	2.514	3.26	2.80	1.56	0.60	4.58	2.36	4.73		0.68
1.21	5	13	2.4	2.463	2.384	3.464	3.86	3.27	1.97	0.71	4.39	3.09	4.76	1.77	
1.44	6	13	2.6	3.710	3.590		4.11	3.53	2.27	0.94	4.26	3.31	4.53	2.04	1.94
2.25	6	16	3.2	3.442	2.741		4.02	3.65	2.68	1.58	3.82	3.21	3.97	2.76	2.52
4.00	7	22	3.1	2.867	1.593		3.27	3.12	2.57	2.03	3.03	2.88	3.15	2.56	2.64

[a] For the present calculations close-coupling or correlation-method reactance-matrix elements were used for $0 \leq L \leq L_1$, Born II reactance-matrix elements were used for $(L_1+1) \leq L \leq L_2$, and, at the three higher energies, Born I reactance-matrix elements were used for $(L_2+1) \leq L$.

[b] This is the contribution to the present calculation which comes from $0 \leq L \leq L_1$.

[c] Three-state close coupling for $0 \leq L \leq 5$ and Born II for $6 \leq L \leq 13$.

[d] D. G. Truhlar (unpublished calculations).

at 1.44 Ry, $17 \leqslant L \leqslant 40$ at 2.25 Ry, and $23 \leqslant L \leqslant 75$ at 4.00 Ry). In each case the highest-L value for which Born II or Born I calculations were carried out was chosen to meet the convergence criteria for the differential cross section discussed below. The L values at which we switched to Born II and Born I reactance matrices are summarized in Table I.

For $L \geqslant 23$ the Born I reactance-matrix elements were calculated using only the long-range $c_i r^{-2}$ terms in the static (nonexchange) $1s$-$2p$ potential matrix elements. (These terms are given by Omidvar.[20]) This asymptotic approximation is valid at high-enough L and it was used to bypass the computational problems encountered when using the program described in Ref. 19 for $L > 23$. The formulas used for the asymptotic Born I reactance-matrix elements are[21]

$$R^L_{1sL2pL+1} = \frac{64\pi}{243}\left(\frac{2(L+1)}{2L+1}\right)^{1/2}\left(\frac{k_2}{k_1}\right)^{L+3/2}\frac{\Gamma(L+1)}{\Gamma(\frac{1}{2})\Gamma(L+\frac{5}{2})}\left[1+\sum_{n=1}^{\infty}\left(\frac{k_2}{k_1}\right)^{2n}\prod_{i=1}^{n}\frac{(L+i)(i-\frac{1}{2})}{i(L+\frac{3}{2}+i)}\right], \tag{1}$$

$$R^L_{1sL2pL-1} = -\frac{64\pi}{243}\left(\frac{2L}{2L+1}\right)^{1/2}\left(\frac{k_2}{k_1}\right)^{L-1/2}\frac{\Gamma(L)}{\Gamma(\frac{3}{2})\Gamma(L+\frac{1}{2})}\left[1+\sum_{n=1}^{\infty}\left(\frac{k_2}{k_1}\right)^{2n}\prod_{i=1}^{n}\frac{(L+i-1)(i-\frac{3}{2})}{(L+i-\frac{1}{2})i}\right]. \tag{2}$$

The sums in (1) and (2) were evaluated including only terms larger than 10^{-14}.

By $L = 23$ the results using (1) or (2) agreed with the results obtained using Ref. 19 to better than 0.1% and the agreement between the complete and asymptotic Born I calculations was getting better as L approached 23 from below. Thus the use of the asymptotic approximation caused essentially no error in the present calculations.

The differential cross sections were calculated from the reactance matrices using a program and methods described elsewhere.[12,13] Unlike the procedure used in Ref. 13 however, the Born reactance-matrix elements used in this work were *not* truncated to the nearest ten thousandth but were accurate to four significant figures. In Eq. (9) of Ref. 13, $\lambda_{\max} = 26$ at the lowest three energies, $\lambda_{\max} = 38$ at 1.44 Ry, $\lambda_{\max} = 80$ at 2.25 Ry, and $\lambda_{\max} = 150$ at 4.00 Ry. The calculation at 4.00 Ry required 444 677 Z coefficients.

The integral cross sections presented in Table I are not as sensitive to the large-L reactance-matrix elements as are the differential cross sections. The integral cross sections obtained including only the low-L contributions, where calculations more accurate than the Born II approximation were used, are given in Table I for completeness.

Table I also includes for comparison the results obtained in some other calculations where only the integral cross sections were calculated (or at least only the integral cross sections were published): the Born I approximation neglecting exchange[20,22] (B), the Ochkur approximation[23] (O), the Vainshtein-Presnyakov-Sobelman approximation[24] (VPS), the modified VPS approximation of Crothers and McCarroll[25] (CM), the distorted-wave approxition neglecting exchange[26] (DW), the two-state close-coupling calculations[20] ($1s$-$2p$ c.c.), the three-state close-coupling calculations neglecting exchange[20] ($1s$-$2s$-$2p$ c.c. XE), the three-state plus two-pseudostate close-coupling calculations of Burke and Webb[27] (BW), and the multistate eikonal treatment of Flannery and McCann[28] (FM). Many other approximation methods have been applied to this problem and they can be compared to the results in Table I. In general the integral cross sections calculated from the reactance matrices used for the present differential cross-section calculations and the integral cross sections of Burke and Webb are in better agreement with experimental integral cross sections than are the results of the various simpler approximation schemes, although in some cases less complete treatments are more accurate due to cancellation of errors for the integral cross section. Such cancellation of errors is less likely for differential cross sections. Thus it appears to be worthwhile to employ elaborate calculations.

The differential cross sections as a function of scattering angle Θ are presented in Table II. In addition to the calculations presented in Table II several convergence checks were carried out. At the lower three energies, the $1s$-$2p$ differential cross sections were recalculated through $\lambda_{\max} = 26$ but with the close-coupling reactance matrix for L equals L_1 (defined in Table I) at each energy replaced by a Born II reactance matrix. The results agreed to within 0.1% at 0.81 Ry, 4% at 1.00 Ry, and 13% at 1.21 Ry with the calculations presented in Table II at all angles. At the higher three energies a check was carried out to ascertain the effect of changing from Born II to Born I reactance-matrix elements. The $1s$-$2p$ differential cross sections calculated using the reactance-matrix elements described above were compared to the differential cross sections calculated after replacing the Born II reactance matrices for the highest two L values at each energy (12 and 13 at 1.44 Ry, 15 and 16 at

TABLE II. Differential cross sections $I(E,\Theta)$ for $1s$-$2p$ transition as a function of impact energy E and scattering angle Θ. Numbers in parentheses denote multiplicative powers of ten.

Θ	$I(a_0^2/\mathrm{sr})$ E (Ry)					
(deg)	0.81	1.00	1.21	1.44	2.25	4.00
0	2.47(−1)	9.67(−1)	2.95	6.01	15.79	41.18
5	2.43(−1)	9.30(−1)	2.78	5.57	12.69	21.89
10	2.32(−1)	8.31(−1)	2.36	4.50	7.52	7.59
15	2.14(−1)	6.93(−1)	1.82	3.28	4.10	2.87
20	1.92(−1)	5.43(−1)	1.31	2.28	2.27	1.19
30	1.44(−1)	2.92(−1)	6.23(−1)	1.04	7.38(−1)	2.19(−1)
40	1.04(−1)	1.47(−1)	3.06(−1)	4.56(−1)	2.39(−1)	4.36(−2)
50	7.90(−2)	8.71(−2)	1.74(−1)	2.26(−1)	9.50(−2)	1.96(−2)
60	6.93(−2)	7.19(−2)	1.22(−1)	1.45(−1)	6.00(−2)	1.29(−2)
70	6.84(−2)	7,22(−2)	9.64(−2)	1.07(−1)	4.11(−2)	6.42(−3)
80	6.92(−2)	7.48(−2)	7.97(−2)	8.17(−2)	2.61(−2)	3.68(−3)
90	6.63(−2)	7.50(−2)	7.01(−2)	6.92(−2)	1.91(−2)	3.54(−3)
100	5.78(−2)	7.19(−2)	6.92(−2)	6.77(−2)	1.78(−2)	3.25(−3)
110	4.60(−2)	6.64(−2)	6.94(−2)	7.18(−2)	1.65(−2)	2.31(−3)
120	3.75(−2)	6.05(−2)	6.72(−2)	7.59(−2)	1.40(−2)	1.41(−3)
130	4.08(−2)	5.65(−2)	6.36(−2)	7.73(−2)	1.06(−2)	1.11(−3)
140	6.24(−2)	5.60(−2)	6.37(−2)	7.51(−2)	7.42(−3)	1.43(−3)
150	1.02(−1)	5.96(−2)	7.04(−2)	6.87(−2)	5.27(−3)	1.82(−3)
160	1.48(−1)	6.62(−2)	7.89(−2)	6.07(−2)	4.88(−3)	1.37(−3)
170	1.86(−1)	7.28(−2)	8.66(−2)	5.32(−2)	5.91(−3)	5.91(−4)
180	2.01(−1)	7.56(−2)	9.00(−2)	5.12(−2)	6.64(−3)	3.28(−4)

2.25 Ry, and 21 and 22 at 4.0 Ry) with Born I reactance-matrix elements. The two calculations agreed to within 1% at 1.44 and 2.25 Ry and 4% at 4.00 Ry at all angles. Since the sum over L in the differential cross section converges more slowly as the energy is increased, checks were carried out at the four highest energies to ascertain the effect of removing the reactance-matrix elements for the highest L (13 at 1.21 Ry, 19 at 1.44 Ry, 40 at 2.25 Ry, and 75 at 4.00 Ry) from the calculations presented in Table II. The differential cross section converges most slowly at very large Θ where it oscillates as L is increased. Upon eliminating the term with highest L, the differential cross section remained unchanged within 1% for all Θ less than 150° at 1.21 Ry, 176° at 1.44 Ry, 178° at 2.25 Ry, and 177° at 4.00 Ry. Even at $\Theta = 180°$ it remained the same within 4% at all energies.

The effect of the large-L contributions on the differential cross sections at the four highest energies is shown in Fig. 1 where the present differential cross sections are compared to cross sections computed by setting all reactance matrices with $L > L_1$ (defined in Table I) equal to zero. The figures show that inclusion of higher-L values generally raises the ratio $I(E,0)/(E,180)$ as expected.

The contributions from the Born I reactance matrices are most important at the two highest energies. If these reactance matrices are left out the differential cross section oscillates about the present results with "wavelengths" (angular spacing between successive minima or maxima in the oscillation pattern) of about 20°. Figure 1 also shows for comparison the Born I approximation neglecting exchange. The Born-I-approximation differential cross sections are monotonically decreasing functions of scattering angle. At small scattering angles they agree in angular dependence but overestimate the magnitude as compared to the more accurate results. At larger scattering angles they underestimate the cross section. These conclusions agree with those obtained previously[29,30] by comparing the Born approximation to experimental differential cross sections for the $1\,^1S$-$2\,^1P$ excitation of He in a comparable energy range (impact energies $1\frac{1}{2}$–4 times threshold energy). This gives further support to the expectation that this should be the general result for optically allowed excitations at intermediate energy.

The authors are grateful to Dr. Richard L. Smith for assistance with some of the calculations.

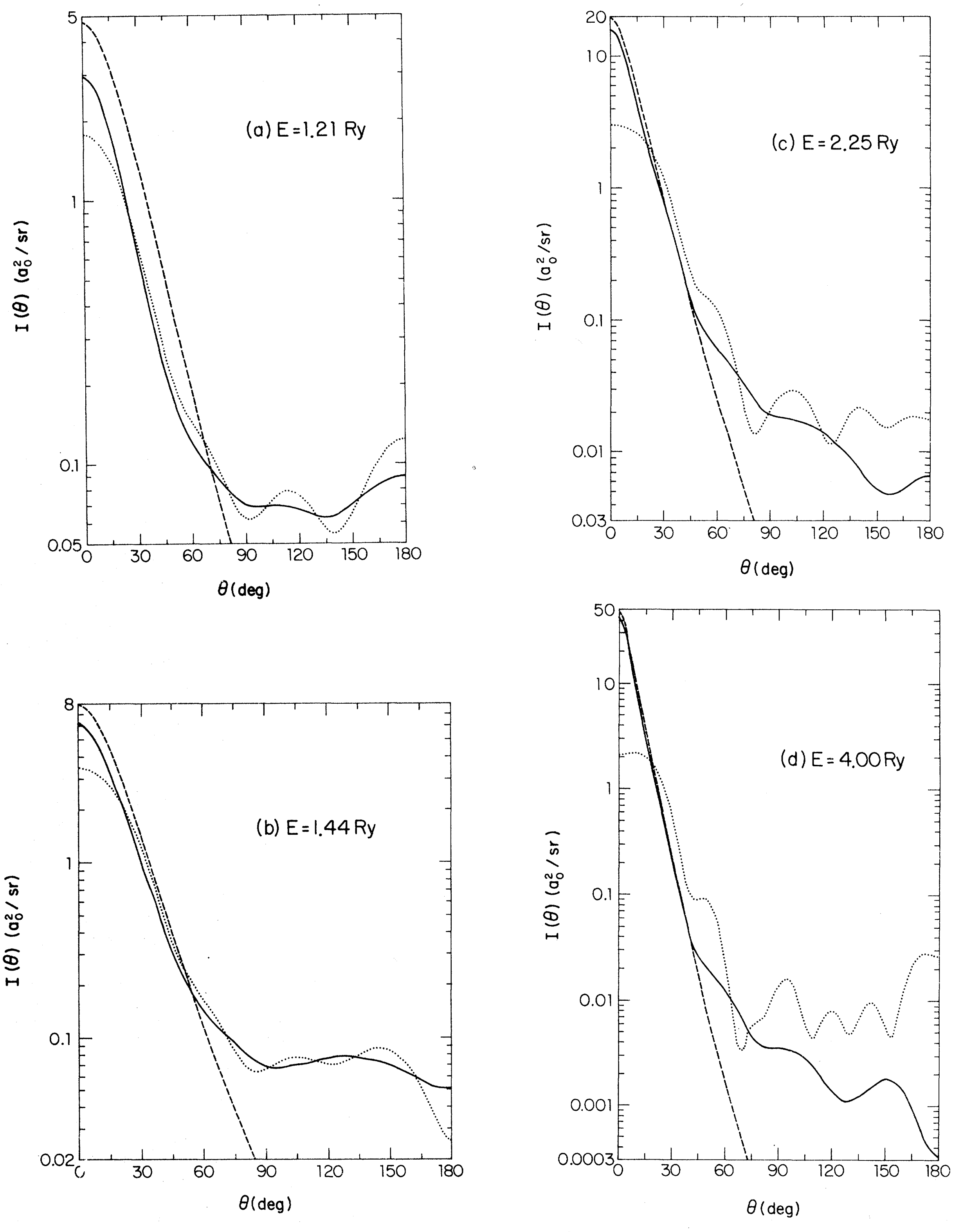

FIG. 1. Differential cross sections as functions of scattering angle for the $1s$-$2p$ excitation of the hydrogen atom at electron impact energies (a) 1.21 Ry, (b) 1.44 Ry, (c) 2.25 Ry, and (d) 4.00 Ry. Solid lines, complete present calculations; dotted lines, low-L results, i.e., the results if only the contributions from $0 \le L \le L_1$ (where L_1 is defined in Table I) are included; dashed line, first Born approximation.

TABLE AI. Born-approximation reactance-matrix elements for $L = 14$ at total energy 1.25 Ry (impact energy 2.25 Ry in the 1s initial channel and 1.50 Ry in other channels). Numbers in parentheses denote multiplicative powers of ten.

	$1sk_1L$	$2sk_2L$	$2pk_2L+1$	$2pk_2L-1$
$1sk_1L$	2.152(−8)	−1.156(−6)	5.508(−4)	−9.555(−3)
$2sk_2L$		6.739(−4)	−1.434(−1)	1.482(−1)
$2pk_2L+1$			−1.665(−2)	1.709(−2)
$2pk_2L-1$				−1.723(−2)

APPENDIX

Reactance-matrix elements for $L > 13$ or reactance-matrix elements smaller than 10^{-4} have apparently never been published before. Some accurate values of these may be useful as test cases for other workers and for this purpose some of our Born-approximation results are presented in Table AI.

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*Research supported in part by the NSF under research Grant No. GP-28684 and by the University Computer Center and the Graduate School of the University of Minnesota.

†Alfred P. Sloan Research Fellow.

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