Spin-dependent electron-impact excitation of sodium

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The spin-dependent cross sections for electron excitation of the $3p$ level of sodium at energies just above threshold are studied using the R-matrix approach. In addition to the total cross section for excitation, we present results for the excitation cross sections as a function of the change in the spin and orbital angular-momentum projection of the target. Calculations were made in the four- $(3s,3p,4s,3d)$, five- $(+4p)$, and six-state $(+4p,4d)$ approximations, for electron energies ranging from threshold to 4.0 eV. The results are compared with other calculations and with the recent experimental results of Han, Schinn, and Gallagher [Phys. Rev. A **38**, 535 (1988)].

I. INTRODUCTION

The alkali metals, with a single electron outside a closed shell, are particularly simple systems to describe theoretically. Since they are also easy to handle experimentally, they have been the subject of a great many studies as theorists and experimentalists seek to test and improve their techniques.

Sodium in particular has been one of the most popular targets for electron scattering over the years. This is due in part to the relative ease in preparation. More recently, the coincidence that the sodium D lines are at a wavelength that is appropriate for rhodamine dye has motivated a number of studies which give more information than the usual differential and total cross section experiments. The use of laser light to prepare atoms in pure states, in conjunction with the use of spin-polarized electron beams, opens the possibility of a "complete" measurement[1] of the scattering amplitudes that characterize the collision. While there have been no "complete" experiments to date, there have been a number of experiments[2] that have made partial measurements of the complete set of observables that characterize the reaction.

One of the more recent of these experiments was done by Han, Schinn, and Gallagher (HSG).[3] The angle-integrated partial cross sections for exciting the $3p$ state were measured as a function of the change in both the spin (Δm_S) and orbital angular-momentum (Δm_L) projections of the target electron.

In most respects the results of this experiment were consistent with those of a four-state $(3s,3p,4s,3d)$ close-coupling calculation by Moores and Norcross.[4] This is not surprising since one intuitively expects a close-coupling calculation to provide a reasonable description of the dynamics of an electron-sodium collision at low energies. There are, however, substantial discrepancies between the calculation of Moores and Norcross (MN) and the experimental data. Their calculations predict, for example, for Q_1^1 (the cross section for simultaneously changing the spin and orbital projections by 1) a value roughly half of that observed. One purpose of this paper is to discuss this discrepancy.

We compare theoretical results obtained using the R-matrix method with the measurements of HSG, and with the original calculation of MN. Compared with this early work, the descriptions of both the target and the interaction potential have been improved; in addition, we have improved the convergence of the close-coupling expansion with respect to target states.

II. DETAILS OF THE CALCULATION

The accuracy of electron collisional cross section depends first on the target wave function, and second on a reasonable description of the scattering event. We describe the target using properly symmetrized products of single-particle wave functions. Unlike the early MN calculation, in which a statistical model potential was used to describe the interaction of valence and scattered electrons with the core, each target state wave function in the present work included the $1s^2 2s^2 2p^6$ neonlike core explicitly. A Hartree-Fock calculation of the $3s$ ground state was used to define the core wave functions. The wave functions of the valence electron were calculated using a semiempirical approach. Rather than allowing excitations out of the core in order to describe its polarization, we included in the Hamiltonian a polarization potential of the form described by Norcross and Seaton,[5] i.e., (in atomic units),

$$V_{\rm pol}=\frac{-\alpha_d}{2r^4}[1-\exp(-r/\rho_l)^6]\ . \tag{1}$$

The static dipole polarizability α_d of the neonlike core is taken[6] to be $0.99a_0^3$, and the cutoff parameters ρ_l are adjusted to get correct binding energies. The values of ρ_l so determined are 1.439, 1.65, and 1.66 a.u., respectively, for

TABLE I. Calculated binding energies (in a.u.) for some valence states of Na, and dipole length f_L and dipole velocity f_V oscillator strengths. The experimental binding energies (Ref. 7) are averaged over the spin-orbit splitting. The modified dipole operator used the core-polarization correction (Refs. 8 and 9) $r_{\text{eff}}=r-(\alpha_d/r^2)[1-\exp(-r/\rho)^3]$, where the cutoff parameter ρ used in this expression was taken to be the average of ρ for the initial and final states.

	Binding energies			Oscillator strengths			
State	Theory	Experiment	Transition	f_L	f_v	f_L modified	Expt. and others
3*s*	−0.188 862	−0.188 857	3*s*-3*p*	0.9974	0.9626	0.9709	0.964[a]
3*p*	−0.111 551	−0.111 547	3*s*-4*p*	0.0141	0.0135	0.0122	0.0142[b]
4*s*	−0.071 547	−0.071 577	3*p*-4*s*	0.1703	0.1684	0.1711	0.163[b]
3*d*	−0.055 941	−0.055 936	4*s*-4*p*	1.4546	1.4453	1.4506	1.35[b]
4*p*	−0.050 933	−0.050 934	3*p*-3*d*	0.8678	0.8651	0.8615	0.83[b]
4*d*	−0.031 444	−0.031 442	3*d*-4*p*	0.1178	0.1231	0.1178	0.117[b]
			3*p*-4*d*	0.0994	0.0995	0.0979	0.106[b]
			4*p*-4*d*	0.9275	0.9598	0.9265	0.91[b]

[a]Reference 10.
[b]Reference 11.

$l=0$, 1, and 2. The use of the polarization potential results in binding energies that are much improved over Hartree-Fock results. This can be expected to affect the scattering calculation, especially near threshold. The calculated binding energies for some valence states, and oscillator strengths for some dipole allowed transitions are given in Table I.

For the four-state close-coupling approximation (4CC) we included, in addition to the $1s^2 2s^2 2p^6$ core, the 3*s*, 3*p*, 4*s*, and 3*d* valence states. For the five-and six-state calculations (5CC and 6CC), we added the 4*p* then the 4*d* states, respectively. We adopted the *R*-matrix approach to solving the close-coupling equations, working with the nonrelativistic version of the well-known RMATRX package.[12] The present calculation is by no means the largest calculation done using this code in terms of number of coupled states. It did, however, require perhaps the largest *R*-matrix box used to date—a boundary radius of $39a_0$ was required in order to contain the most diffuse (4*d*) orbital, and 28 basic functions per channel were required in order to achieve adequate convergence in the smallest cross section.

To be consistent with the target state description, a one-body polarization potential [of the form given by Eq. (1)] and two-body dielectronic potential[5] of the form

$$V_{\text{di}}(r_1,r_2)=-\alpha_d\frac{\hat{\mathbf{r}}_1\cdot\hat{\mathbf{r}}_2}{r_1^2 r_2^2}\{[1-\exp(-r_1/\rho)^6]\times[1-\exp(-r_2/\rho)^6]\}^{1/2} \quad (2)$$

were added to the $(N+1)$-electron scattering Hamiltonian in the *R*-matrix package. In Eq. (2) the values adopted for the cutoff parameter were the same for all partial waves and fixed at an average value of ρ_l, i.e., $\rho=1.583a_0$.

III. COMPARISON WITH EXPERIMENT

The observables measured in the HSG experiment are the partial cross sections for excitation with respect to a change in the spin and orbital angular momentum of the target electron. These cross sections are denoted by $Q^{\Delta m_S}_{\Delta m_L}$, where Δm_S and Δm_L denote the change in the atom's spin and orbital quantum numbers, respectively. Since the strength of the spin-orbit interaction is very weak in sodium (as illustrated by the fine-structure splitting) one can use symmetry arguments to show that the partial cross sections are independent of the sign of Δm_S and Δm_L. As a consequence we need only report values of $Q^{|\Delta m_S|}_{|\Delta m_L|}$. In their experiment HSG measured relative cross sections, which were put on an absolute scale by summing and normalizing at each energy point to previously measured values of the total cross section,[13] using the relationship

$$Q_{\text{tot}}=Q^0_0+Q^1_0+2Q^0_1+2Q^1_1 \ . \quad (3)$$

The present 6CC results for these cross sections are given in Table II, and total cross sections for excitation of the 3*d*, 4*s*, and 4*p* states in Table III.

In Fig. 1 we compare the results of two different 4CC calculations of the total 3*s*-3*p* cross section. Although MN used a relatively simple model, the two 4CC calculations are in excellent agreement below the 4*s* threshold (3.19 eV). Also in Fig. 1 we show results of the 6CC calculation, and the experimental results of HSG. The three calculations are practically indistinguishable for energies below the 4*s* threshold, and theory and experiment are in good agreement in this same range. One reason for the difference between theory and experiment at higher energy is an artifact of the measuring technique, which uses a laser to probe the population of the $3p_{3/2}(m_J)$ level resulting from the collision. This method detects, in addition to direct excitation to the 3*p* state, the cascade into this level from higher states, such as the 4*s* and 3*d*. If we include the cascade effect for the total cross section, the theoretical results are in good agreement with the measurement over the entire energy range.

Calculated (4CC) and measured results for Q^1_1, the smallest and most sensitive of the four partial cross sections, are shown in Fig. 2. MN did no calculation between 3.0 and 4.0 eV so we could not compare the structure in this energy range. Agreement of two calculated cross sections is still good, but significant discrepancy be-

TABLE II. 6CC results for partial and total cross sections (units of πa_0^2) for electron-impact excitation of the $3p$ state of sodium.

E (eV)	Q_{tot}	Q_0^0	Q_0^1	Q_1^0	Q_1^1
2.20	12.65	5.31	5.34	0.55	0.445
2.30	14.73	7.20	5.70	0.63	0.284
2.40	17.57	9.36	5.51	1.08	0.276
2.50	21.38	11.97	5.52	1.67	0.278
2.60	24.57	14.09	5.66	2.15	0.262
2.70	26.60	15.42	5.77	2.46	0.239
2.80	27.83	16.18	5.83	2.69	0.219
2.90	28.61	16.61	5.82	2.88	0.206
3.00	29.20	16.87	5.76	3.08	0.202
3.10	29.89	17.07	5.69	3.36	0.206
3.25	30.15	16.04	5.07	4.18	0.336
3.40	29.91	16.91	4.69	3.89	0.266
3.55	30.58	17.59	4.46	4.01	0.258
3.70	30.61	17.32	3.95	4.51	0.158
3.85	30.92	17.40	3.59	4.81	0.153
4.00	31.02	17.35	3.34	5.03	0.141

tween the calculated and measured results is evident.

In Fig. 3 we compare the 5CC and 6CC R-matrix results for Q_1^1 with those of HSG. The 5CC and 6CC calculations are in excellent agreement with the 4CC results below 3.0 eV, and are almost identical to each other between 2.2 and 4 eV. This means that coupling to the $4d$ state is weak in this low-energy region. The peak near the $4s$ threshold may be due to resonances caused by the $4p$ dipole potential tail. In general, all the close-coupling calculations yield results that are significantly smaller than the experimental results.

Part of this discrepancy is caused by the radiative cascade effects described above. If we ignore effects due to fine and hyperfine structure that will mix spin states before radiative decay, there is a simple way to correct the partial cross section $Q_{|\Delta m_L|}^{|\Delta m_S|}$ for these cascade effects. For the $4s$ level, we suppose that electrons occupying this level decay to $m_L=-1,0,1$ states with equal probability, so that the contribution from $4s$ will be $\frac{1}{3}Q_{4s}^{|\Delta m_S|}$, where the $Q_{4s}^{|\Delta m_S|}$ are the partial cross sections for excitation of the $4s$ level involving a change in the spin $|\Delta m_S|$ of the target electron. For the $3d$ cascade correction we apply the selection rule $|\Delta m_L|=0$ or 1 for the radiative decay, and thus the corrections for the $3d$ state are

$$^{\text{corr}}Q_1^{|\Delta m_S|}=Q_{(3d)2}^{|\Delta m_S|}+\tfrac{1}{2}Q_{(3d)1}^{|\Delta m_S|}+\tfrac{1}{3}Q_{(3d)0}^{|\Delta m_S|} \tag{4a}$$

and

TABLE III. 6CC results for total cross sections (units of πa_0^2) for excitation of the $4s$, $3d$, and $4p$ states of sodium.

E (eV)	$Q(3s\rightarrow 4s)$	$Q(3s\rightarrow 3d)$	$Q(3s\rightarrow 4p)$
3.25	1.25		
3.40	2.45		
3.55	3.72		
3.70	1.55	3.11	
3.85	1.70	3.51	0.23
4.00	1.88	3.95	0.61

$$^{\text{corr}}Q_0^{|\Delta m_S|}=\tfrac{1}{3}Q_{(3d)0}^{|\Delta m_S|}+Q_{(3d)1}^{|\Delta m_S|}, \tag{4b}$$

where $Q_{(3d)2,1,0}^{|\Delta m_S|}$ are the corresponding partial cross sections for impact excitation from the ground state to the $3d$ level.

Also shown in Fig. 3 are the 6CC results for Q_1^1, including the cascade correction from the $4s$ state alone, and from the $4s$ and $3d$ states together. If we include the $4s$ cascade, the cross section is 74% larger at 3.25 eV, and

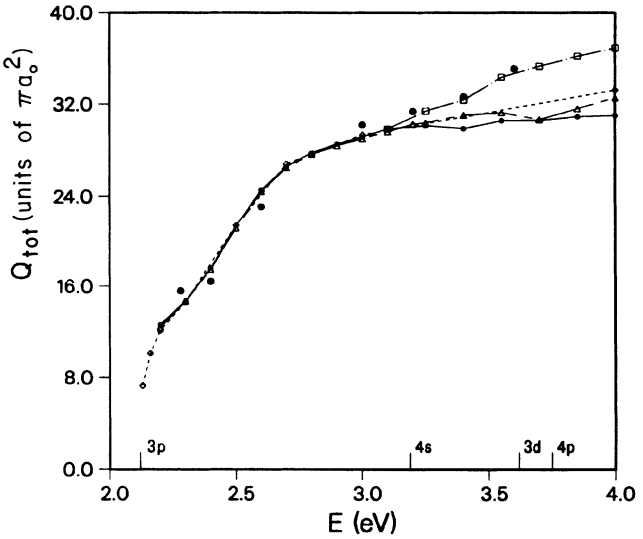

FIG. 1. Comparison of calculated and experimental values for the $3s$-$3p$ total cross sections as a function of the incident energy. Present 6CC, –○–; present 6CC including $4s$ and $3d$ cascade, –·–□–·–; present 4CC, – – –△– – –; MN (Ref. 4), – –◇– –; HSG (Ref. 3), ●. The lines linking the points are merely a guide to aid the reader and should not be interpreted as representing the results of an actual calculation. Thresholds for exciting higher states are indicated.

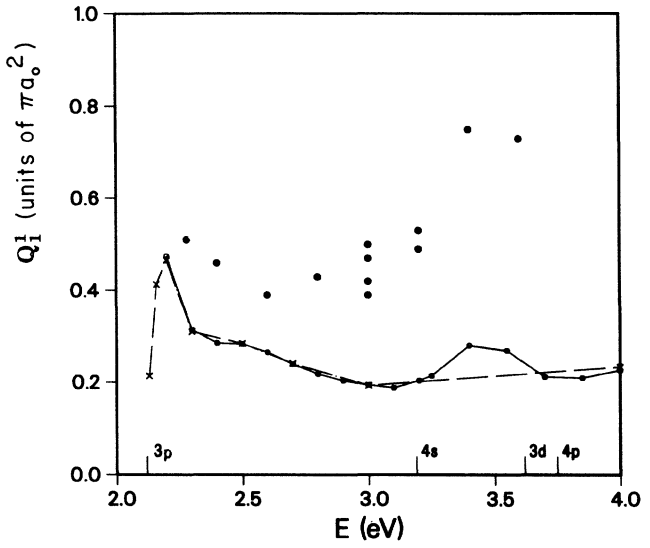

FIG. 2. Comparison of two 4CC calculations and experimental values for the Q_1^1 partial cross sections. Present 4CC, $-\circ-$; MN (Ref. 4), $-\cdot-\times-\cdot-$; HSG (Ref. 3), ●.

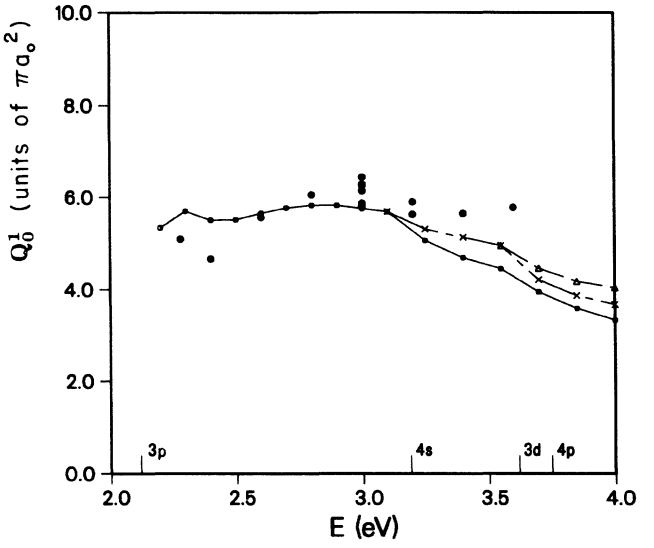

FIG. 4. As Fig. 3 for the Q_0^1 partial cross section. Present 6CC, $-\circ-$; present 6CC including 4*s* cascade, $---\times---$; present 6CC including 4*s* and 3*d* cascade, $--\Diamond--$; HSG (Ref. 3), ●.

three times as large at 3.55 eV. And if we include the 3*d* cascade above 3.62 eV as well, the cross section Q_1^1 increases by a factor of 5. All the calculations for Q_1^1 are still significantly smaller than the experiment below the 4*s* threshold. At 3.0 eV the electron beam of full width at half maximum (FWHM) 0.3 eV would overlap the sharply rising cascade contribution from the 4*s* state, partly accounting for the measured cross section.

In Figs. 4–6 we show 6CC results with and without cascade for the other three partial cross sections. For Q_1^0 (Fig. 5), as for Q_1^1, correcting for cascade improves the agreement with experiment above 3.0 eV. However, for Q_0^1 (Fig. 4) the calculations all decrease at higher energies, a trend not seen in the experiment. The results for Q_0^0, the largest partial cross section (Fig. 6), are in good agreement (on a fractional basis) at all energies. It should be noted, however, that the absolute difference between the measured and calculated values is largest for Q_0^0 and smallest for Q_1^1.

HSG noted that their results might have been affected by electron spiraling, which would increase the

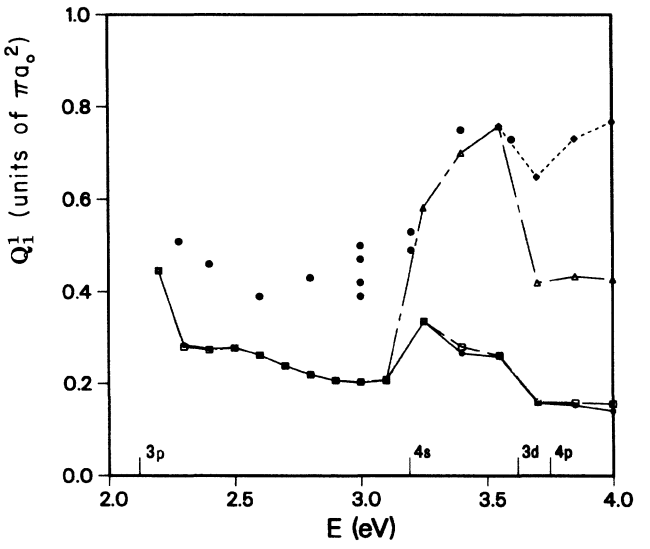

FIG. 3. Comparison of *R*-matrix calculations with and without cascade corrections, and with experimental values for the Q_1^1 partial cross section. Present 6CC, $-\circ-$; present 5CC, $-\cdot-\square-\cdot-$; present 6CC including 4*s* cascade, $---\triangle---$; present 6CC including 4*s* and 3*d* cascade, $--\Diamond--$; HSG (Ref. 3), ●.

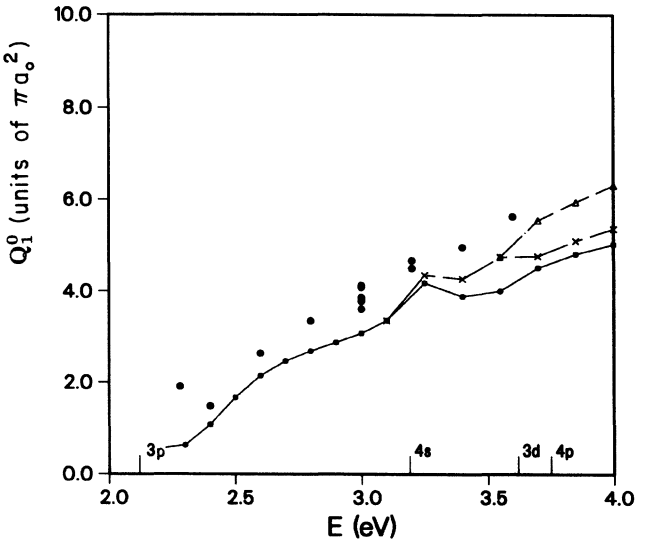

FIG. 5. As Fig. 3 for the Q_1^0 partial cross section.

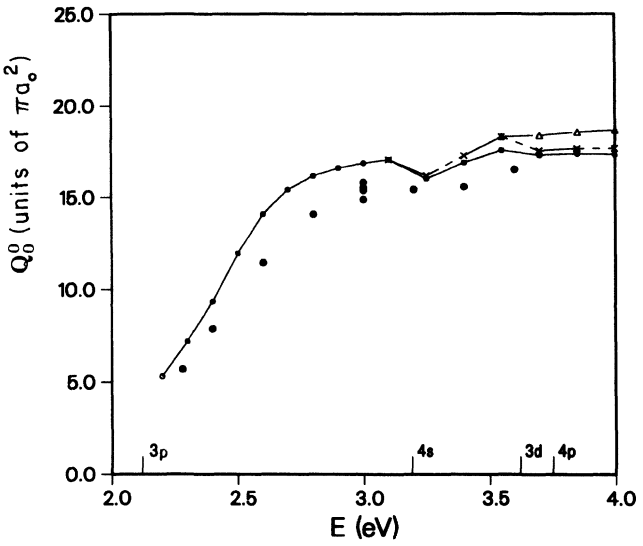

FIG. 6. As Fig. 3 for the Q_0^0 partial cross section.

$|\Delta m_L|=1$ results relative to those for $|\Delta m_L|=0$, particularly in the first 0.5 eV above threshold. Their results were not corrected for this possible effect, but they suggested that it could account for as much as 30% of the measured Q_1^1 and 8% of Q_1^0 at 2.55 eV. A correction of this magnitude would improve agreement between observed and calculated results for both Q_1^1 and Q_1^0 in the energy range where the greatest discrepancies remain. Further, if a correction of this magnitude were applied to Q_1^0 and Q_1^1, and the total cross section renormalized, the experimental results for Q_0^0 and Q_0^1 would increase somewhat near threshold, again improving the agreement with the present calculations.

The present calculations can also be compared with measured results[14] for the polarization of the radiation emitted by the decaying $3p$ state. This is expressed in terms of $Q_{|\Delta m_L|}^{|\Delta m_S|}$ by

$$P=\frac{3(9\alpha-2)(Q_0-Q_1)}{12Q_0+24Q_1+(9\alpha-2)(Q_0-Q_1)}\ , \tag{5}$$

where $\alpha=0.288$ and

$$Q_{|\Delta m_L|}=\sum_{|\Delta m_S|=0}^{1} Q_{|\Delta m_L|}^{|\Delta m_S|}\ . \tag{6}$$

We have already noted the excellent agreement between the present 4CC and 6CC calculations and those of MN for the total $3s$-$3p$ cross section. Agreement between the calculated values of P is also excellent. The calculated and measured[14] polarizations are also in good agreement. It should also be noted that the $Q_{|\Delta m_L|}^{|\Delta m_S|}$ measured by HSG can be used in Eqs. (5) and (6) to *deduce* the polarization, which turns out to be in reasonable agreement with the previous measurement[14] except in the ~ 0.5 eV above threshold where electron spiraling was suspected.

IV. CONCLUSIONS

We have done calculations of the $3s$-$3p$ electron-impact excitation of sodium using the R-matrix method to describe the scattering process, and three different descriptions of the target states. All theoretical results are in good agreement with each other, and with the original calculation of MN. This provides welcome confirmation that the much simpler representation of the interaction potential used in that earlier work is useful. It also suggests that the quantities calculated here for the excitation of the $3p$ state are unlikely to be much affected by either further improvements in the interaction potential or further extensions of the close-coupling expansion.

Some troubling discrepancies between theory and experiment remain. Most seriously, the Q_1^1 cross section below the $4s$ threshold is measured to be almost twice as large as the theoretical predictions. This very small cross section is $\sim 1\%$ of the total cross section and is difficult to measure; the four points at $E=3.0$ eV give some idea of the scatter in the data. Electron spiraling in the experiment could account for some of this difference in the energy region 2.1–2.5 eV. We can at the moment point to no particular aspects of either the calculations or measurements that might account for the rest.

Future work will involve extension of these calculations to differential cross sections, to more detailed analysis of resonance and threshold structure associated with the $n=4$ states,[15] and to superelastic[16] and elastic[17] scattering by laser-excited $3p$ states. Comparison with the superelastic scattering measurements, in particular, may contribute to an explanation of the discrepancy in excitation cross-section results, since these two processes are described by exactly the same scattering amplitudes.

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[1] B. Bederson, Comments At. Mol. Phys. **1**, 65 (1969).

[2] Some examples are J. J. McClelland, M. H. Kelley, and R. J.

Celotta, Phys. Rev. Lett. **58**, 2198 (1987); H. W. Hermann, I. V. Hertel, and M. H. Kelley, J. Phys. B **13**, 3465 (1980); J. L. Riley, P. J. O. Teubner, and M. J. Brunger, Phys. Rev. A **31**, 1959 (1985).

[3]X. L. Han, G. W. Schinn, and A. Gallagher, Phys. Rev. A **38**, 535 (1988). Improved measurements yielded the data presented here, which differ somewhat from that published [G. W. Schinn (personal communication)].

[4]D. L. Moores and D. W. Norcross, J. Phys. B **5**, 1482 (1972).

[5]D. W. Norcross and M. J. Seaton, J. Phys. B **9**, 2983 (1976).

[6]W. Müller, J. Flesch, and W. Meyer, J. Chem. Phys. **80**, 3297 (1984).

[7]W. C. Martin and R. Zalubas, J. Phys. Chem. Ref. Data **10**, 153 (1981).

[8]P. Hafner and W. H. E. Schwarz, J. Phys. B **11**, 2975 (1978).

[9]T. C. Caves and A. Dalgarno, J. Quant. Spectrosc. Radiat. Transfer **12**, 1539 (1972).

[10]T. Andersen, O. H. Madesen, and G. Sorenson, Phys. Scr. **6**, 125 (1972). This value includes the cascading correction to the earlier value of 0.903 [Andersen *et al.*, J. Opt. Soc. Am. **60**, 1199 (1970)].

[11]W. L. Wiese, M. W. Smith and B. M. Miles, *Atomic Transition Probabilities,* Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 22 (U.S. GPO, Washington, D.C., 1966), Vol. 2.

[12]K. A. Berrington, P. G. Burke, M. Le. Dourneuf, W. D. Robb, K. T. Taylor, and Vo Ky Lan, Comput. Phys. Commun. **14**, 367 (1977).

[13]B. Stumpf and A. Gallagher, Phys. Rev. A **32**, 3344 (1985). Those measurements were, in turn, normalized to the first-order Born approximation at high energy.

[14]W. Jitschin, S. Osimitsch, H. Reihl, H. Kleinpoppen, and H. O. Lutz, J. Phys. B **17**, 1899 (1984), and references therein.

[15]A. R. Johnston, Ph.D. thesis, University of Nebraska, 1983.

[16]J. J. McClelland, M. H. Kelley, and R. J. Celotta, Phys. Rev. A **40**, 2321 (1989).

[17]M. Zuo, T. Y. Jiang, L. Vušković, and B. Bederson, Phys. Rev. A **41**, 2489 (1990).