Electron-impact ionization of laser-excited sodium atom

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We report the experimental results of the total electron-impact ionization cross sections of laser-excited $3P$ sodium in the energy range from threshold to 30 eV. These cross sections were measured simultaneously for both ground-state and $3P$ excited-state atoms as a function of electron-impact energy. Excited-state data were calibrated with respect to the ground-state data that were normalized to the recent Johnston and Burrow measurements [Phys. Rev. A **51**, R1735 (1995)]. We find good general agreement with theory. However, the position of the maximum is almost the same as that predicted by the calculations employing generalized oscillator strengths based on the Born approximation, and is at a higher energy than that which the convergent close-coupling predicts. [S1050-2947(96)50611-6]

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We present measurements of the electron-impact ionization cross section of excited sodium prepared in the $3P$ state by the absorption of laser light. These data, serving as a guideline for existing calculations, may lead to a deeper understanding of the collision process and induce the development of improved models or techniques. Accurate and reliable values of electron-impact ionization cross sections are also required in many areas of application; e.g., modeling of the upper atomosphere, the aurora, the interstellar medium, and electrical discharges under various conditions of pressure, power, and frequency, such as for semiconductor processing [1,2]. Although total ionization cross sections have been measured for a large number of ground-state atomic species and for some ground-state ions, electron-impact ionization of atoms and ions in excited states is still an unexplored area. The main technical difficulty in producing excited atoms in a controlled fashion for this measurement was overcome with the availability of single-mode dye lasers. Thus, measurements of ionization cross sections are possible for a limited number of atomic excited states. The sodium atom in the present experiment, like the other alkali-metal targets, was easy to produce and detect. Furthermore, it can be considered as an almost ideal two-level system, with the transition frequency conveniently located within the tuning range of the dye laser.

The total ionization cross sections of ground-state and excited-state sodium were first calculated by McGuire [3], employing generalized oscillator strengths (GOSs) based on the Born approximation. The calculations were done in the one-electron common-central potential, unrelaxed-core approximation. Bray and Stelbovics [14] developed a convergent close-coupling (CCC) approximation and used it to calculate the total ionization cross section of ground-state atomic hydrogen [5], achieving a complete quantitative agreement with measurements over almost the entire region except the threshold region. To treat a more complex system, Bray [6] used a generalized CCC approach by including a model of a single electron above a frozen Hartree-Fock core and calculated the total ionization cross sections of ground-state and excited-state sodium.

On the experimental side, measurements of ionization cross sections for the sodium ground state were performed in the 1960s by Brink [7], McFarland and Kinney [8], and Zapesochnyi and Aleksakhin [9]. All obtained similar results, which are much larger than the predictions of both theories [3,6]. Recent experimental data by Johnston and Burrow [10], normalized to the $3P$ excitation cross section, are a factor of 2 lower than those found in the previous experiments [7–9], and are in agreement with theoretical prediction [3,6]. To the best of our knowledge no measurements of the ionization cross section for any excited state in sodium have been reported. The only electron-impact ionization experiment involving a laser-excited, short-lived system was performed by Trajmar, Nickel, and Antoni [11] on barium.

In this paper we present electron-impact ionization experiments on sodium prepared by laser excitation of the $3\,^2P_{3/2}$ ($M_L=1$ or $M_L=-1$) state and obtained ionization cross sections from threshold to 30 eV. Actually, electron-impact ionizations for both ground-state and $3P$ excited-state sodium have been studied simultaneously in the same energy range by measuring the ion current with laser off and laser on as a function of energy and other experimental parameters. Preliminary data [12] motivated an expansion of the CCC approximation [6] to calculate electron-impact parameters, including the total ionization cross section of $3P$ excited sodium.

In order to describe the principles of the present experiment [13], let us first consider ground-state ionization collisions. When an electron beam of speed v_e intersects an atomic beam of speed v_a perpendicularly, the total ionization cross section σ_g at a given relative collision energy is related [14] to the electron beam current (I_e) and the ion current (I_i) by

$$\frac{I_i}{I_e}=SF\sigma_g\frac{\sqrt{v_e^2+v_a^2}}{v_e v_a}, \tag{1}$$

where S is the total number of atoms per second entering the interaction region and F includes the electron atom beams' overlapping factor and the detection efficiency of collisionally produced ions. Because $v_e \gg v_a$ the relative collision energy is practically equal to the electron energy E, and the above equation can be approximated by

$$\frac{I_i}{I_e} = SF\sigma_g \frac{1}{v_a}. \tag{2}$$

In the threshold region the cross section is very sensitive to the projectile energy. Thus, the electron energy distribution has to be considered. Taking into account that the electron beam has a Gaussian energy distribution, Eq. (2) should be modified to

$$\frac{I_i}{I_e} = c\int_{E_1}^{E_2} \sigma_g e^{-a^2(E-E_0)^2} dE, \tag{3}$$

where a is related to the width of Gaussian distribution, E_0 is the nominal electron energy, E_1 and E_2 are the energy limits, and the constant

$$c = \frac{SF}{v_a N}, \tag{4}$$

where N is the normalization coefficient for the electron-energy distribution. We performed preliminary measurements to determine the nominal electron-energy and electron-energy distribution using methods described in Ref. [15] and found the electron-energy distribution width (full width at half maximum) to be about 0.2 eV.

When the $3P$ excited state was studied, a cw laser beam was used to prepare the initial state. The major advantage of the crossed-beam arrangement in the ionization experiment is that the fraction f of the excited-state atoms in the interaction region can be measured. Based on Eq. (3), the total ionization cross section for excited-state scattering (σ_e) can be related to the measured quantities and σ_g by

$$\frac{I_i}{I_e} = c(1-f)\int_{E_1}^{E_2} \sigma_g e^{-a^2(E-E_0)^2} dE + cf\int_{E_1}^{E_2} \sigma_e e^{-a^2(E-E_0)^2} dE. \tag{5}$$

Equations (3) and (5) are the basic equations used to determine the total ionization cross section of excited $3P$ sodium.

The experimental setup, consisting of the electron beam, the sodium atom beam, and the laser beam, was described in previous publications [15,16]. The beams are mutually perpendicular, overlapping with an interaction volume of $1.2 \times 20 \times 1$ mm^3. The ion signal is measured by using an ion detector [17] placed next to the interaction region. It includes an extractor grid, cylindrical shielding body, a repeller, and a collector. The grid with negative voltage was put on the front end of the cylinder to extract ions from the interaction region. The repeller and collector are separated from the cylinder body by porcelain insulation. Since the current measured during the experiment was small (on the order of 10^{-13} A), very high insulation resistance was required.

The experimental arrangement was identical for the ground-state and excited-state ionization measurements. $3P$ sodium atoms were prepared with a traveling-wave laser-field configuration [18]. The average f can be found from the atomic beam displacement at the detector plane [19]. The displacement is proportional to the average force acting on the atom during its propagation through the interaction region. In the traveling-wave laser-field configuration the fraction of excited atoms varies slightly along the distance the atoms travel through the interaction region. In addition, the extraction efficiency of the produced Na^+ is distance dependent. Thus, the effective f was different from the measured average fraction of excited atoms. The effective f in the experiment depends on the laser frequency detuning, laser polarization, and power density. These parameters were varied and the effects on the measured f were checked during the experiment. In addition, a computer simulation [20,21] was performed in order to estimate the correction to f including the effects of spatial inhomogeneities, as well as the geometry of the interaction and ion detection region. The results from this estimate were used to check the correction to f and the uncertainty of its determination.

The absolute cross sections σ_g and σ_e were obtained from measured quantities using Eqs. (3) and (5), respectively. First, ground-state relative cross sections were normalized to the experimental results of Johnston and Burrow [10]. Thus, the constant c was determined. Then, the total ionization cross sections of $3P$ sodium (σ_e) were obtained by calibration with respect to the ground-state data (σ_g) employing Eq. (5). The total error in σ_e, including calibration with respect to σ_g and its normalization procedure, is estimated to be 25%. The ratio σ_e/σ_g is independent of the normalization procedure. The total error in the ratio, estimated to be 20%, is mainly due to statistical errors in intensity measurements and the error in the excited-state fraction determination.

The theoretical ratio can therefore be compared directly with the experimental value. The experimental ratio as a function of the electron-impact energy is plotted in Fig. 1 together with the calculated results of McGuire [3] and Bray [6]. The figure indicates that the experimental ratio of σ_e/σ_g lies between the theoretical predictions. Both calculations are within the experimental errors. However, GOS [3] is always higher than the data, while CCC [6] is always below the experiment.

The absolute cross sections of σ_g and σ_e are given in Table I and shown in Fig. 2 together with calculations [3,6] and other [9,10] data. Our experimental results for the ground-state cross sections are consistent in shape with the data from Ref. [10], which were used for normalization. We like to point out an interesting fact regarding Ref. [10]. The CCC calculations [6] are below the data in Ref. [10], but are just within the claimed experimental errors at the maximum. However, the GOS calculations [3], which were not included in [10], are identical in both shape and magnitude to the data. Thus, we used the Johnston and Burrow data to normalize our results because they are the only recent experimental results, and they are in excellent agreement with theory. The

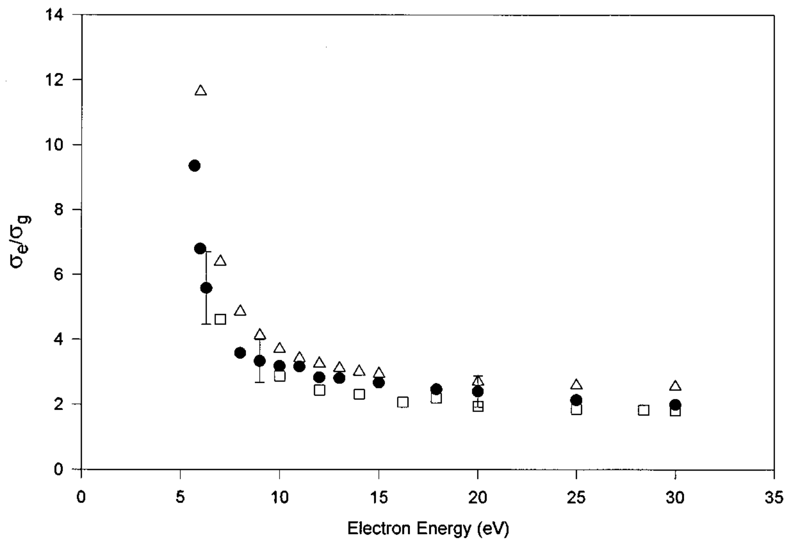

FIG. 1. Ratio of the $3P$ excited-state (σ_e) to the ground-state (σ_g) total ionization cross section for electron-sodium collisions as a function of energy. Experimental data: ●, present. Calculated results: △, McGuire [3]; □, Bray [6].

GOS calculation for σ_e is about 50% higher than the CCC calculation in the region of the maximum. Present experimental results for σ_e show that the position of the maximum is at an energy close to the one predicted by the GOS calculation, while the CCC calculation underpredicts the maximum by 3 eV. After normalizing to σ_g from Ref. [10], our σ_e results are in good agreement with CCC in the threshold region. At the energies around the maximum and above, our data fall between the two calculations, slightly closer to the GOS results. The difference is such that GOS is just inside and CCC just outside our 1σ experimental errors.

It is worth noting here that the laser-prepared initial state in the experiment was a single, polarized state of Na ($3\,^2P_{3/2}$, $M_L=1$ or $M_L=-1$). Ionization cross-section (σ_e) data are compared with calculated cross sections for an unpolarized excited target. With our experimental uncer-

TABLE I. Total ionization cross sections for electron scattering from the sodium ground state (σ_g) and the $3P$ excited state (σ_e) in units of 10^{-20} m^2. Experiment: our data, Zapesochnyi and Aleksakhin [9], and Johnston and Burrow [10]. Theory: McGuire [3] and Bray [6].

E_0 (eV)	σ_g (Present work)	σ_e (Present work)	σ_g Ref. [9]	σ_g Ref. [10]	σ_g Ref. [6]	σ_e Ref. [6]	σ_g Ref. [3]	σ_e Ref. [3]
3.3		1.24						
3.6		2.77			6.62			
3.9		4.62						
4.2		6.23						
4.6		7.86						
5.0		9.24				10.61		10.9
5.3	0.41	10.5			0.54			
5.7	1.24	11.6			1.32			
6.0	1.75	11.9	2.0	1.26			1.22	14.2
6.3	2.22	12.4						
7.0			4.0	2.47	2.6	12.00	2.47	15.8
8.0	3.8	13.6	5.5	3.42		12.07	3.42	16.6
9.0	4.2	14.0	6.2	4.04			4.10	16.9
10.0	4.4	14.0	6.5	4.43	3.85	11.04	4.54	16.8
11.0	4.33	13.7	6.6	4.62			4.83	16.5
12.0	4.74	13.4	6.7	4.74	4.00	9.72	4.96	16.1
13.0	4.63	13.0	6.8	4.82			5.04	15.7
14.0			6.9	4.86	3.85	8.90	5.05	15.2
15.0	4.57	12.2	6.8	4.88			5.03	14.8
16.0			6.8	4.87	3.69	8.03		
17.9	4.47	11.0	6.7	4.8	3.53	7.44		
20.0	4.25	10.2	6.5	4.70	3.42	6.65	4.63	12.5
25.0	3.93	8.39	6.1	4.44	3.02	5.58	4.14	10.7
30.0	3.68	7.32	5.9	4.16	2.66	4.82	3.69	9.4

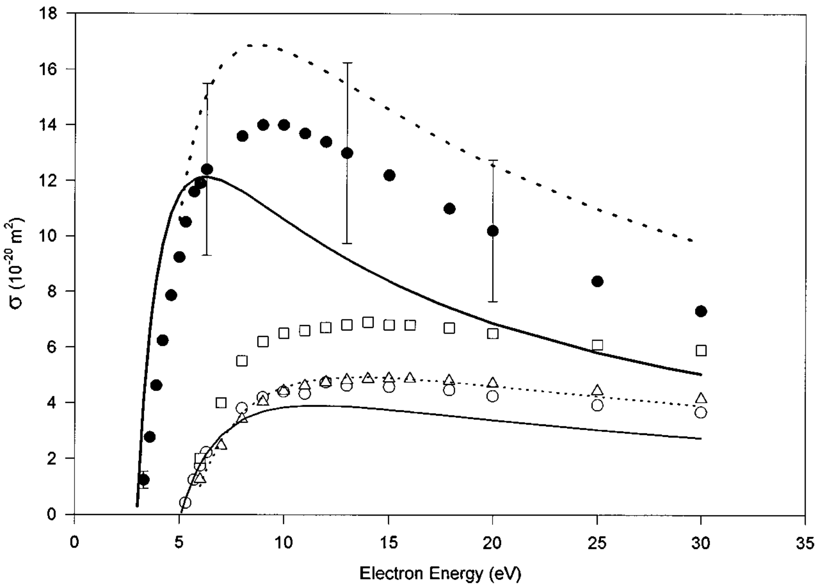

FIG. 2. Total ionization cross sections of the $3P$ excited state (σ_e) and ground state (σ_g) for electron-sodium collisions as a function of energy in units of 10^{-20} m^2. Experimental data: ○, present (σ_g); ●, present (σ_e); □, Zapesochnyi and Aleksakhin (σ_g) [9]; △, Johnston and Burrow (σ_g) [10]. Calculated results: thin dashed line, McGuire (σ_g) [3]; thick dashed line, McGuire (σ_e) [3]; thin full line, Bray (σ_g)[6]; thick full line, Bray (σ_e) [6].

tainty we do not expect to observe a dependence on magnetic sublevels of the ionization cross section. It would be inter--esting to see a theoretical prediction for the M_L dependence, even though electron excitation from $3P$ would be more sensitive to this dependence [3].

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