Absolute Electron-Impact Ionization Cross Section Measurements Using a Magneto-Optical Trap

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We present a new method for measuring absolute total electron-impact ionization cross sections. The technique measures fractional loss rates from a magneto-optical trap due to electron-impact ionization. The method requires only relative measurements of the number of target atoms and therefore eliminates a major source of difficulty in previous experiments. We report total ionization cross sections of Rb for electron energies from 50 to 500 eV. [S0031-9007(96)00306-7]

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We have developed a novel method to determine absolute total electron-impact ionization cross sections by passing an electron beam through a trapped-atom target. The magneto-optical trap (MOT) is operated such that the ions produced by electron collisions escape from the trap while excited and elastically scattered atoms are retained. As compared to standard crossed-beam experiments, this technique greatly simplifies the experimental procedure as only the electron beam current density and the fractional loss rate of the trapped atoms need be measured to obtain the ionization cross section. We avoid measurements of the absolute number of the target atoms and the overlap of the atomic and electron beams, eliminating major sources of error in the crossed-beam method [1].

Recent progress in the theory of electron-impact ionization, in particular the convergent close-coupling (CCC) method developed by Bray and Stelbovics [2], has stimulated much interest. The total cross sections for electron-impact ionization of H atoms so calculated are in excellent agreement with experiment up to 500 eV [3]. However, subsequent application of the CCC method to Na [4] yielded total ionization cross sections much lower than the experiment of McFarland and Kinney [5]. More recently, Johnston and Burrow measured the ratio of the peak ionization cross section to the $3^2S \rightarrow 3^2P$ excitation cross section near threshold for Na [6], and found good agreement with the CCC calculations. In the case of He [7], the CCC total ionization cross sections for the ground state agree well with experiment but the total ionization cross sections from the He(2^3S) metastable level are almost a factor of 2 lower than experiment [8]. These new developments underscore the need for improved methods for measuring ionization cross sections of ground-level atoms as well as for experiments in the much less explored area of ionization of atoms out of excited levels.

We demonstrated the technique using rubidum atoms, which we have used for other atom trapping experiments in our laboratory. Other atoms may also be used, including other alkalis, metastable rare gases, and alkaline earths. Our MOT [9] uses three pairs of intersecting, orthogonal, retroreflected laser beams tuned slightly to the red of the $5S_{1/2} \rightarrow 5P_{3/2}$ transition of Rb, with a magnetic quadrupole field to provide a magnetic-field zero and gradient at the center of the intersection region. Forces from the lasers capture Rb atoms from a room-temperature vapor [10], then cool and confine them near $B = 0$ where a ball (cloud) of trapped atoms forms. We superpose a repetitively pulsed electron beam on this cloud and measure the ionization cross section as follows. We turn off the trapping fields (magnetic and laser) for a short time preceding each electron pulse, leaving the atoms in the ground state. Then the electron pulse ionizes some of the atoms, and we restore the trapping fields. Ionized atoms are unaffected by the lasers so they leave the trap volume, but nonionized atoms are recooled and returned to the center of the trap. The rate at which atoms are ejected from the trap, divided by the electron beam flux, gives the total cross section for producing Rb^+, Rb^{2+}, ..., which we refer to as the total ionization cross section $\sum_n \sigma^{n+}$. A distinct but related quantity is $\sum_n n\sigma^{n+}$ which is directly proportional to the total ion current resulting from electron-impact ionization.

Figure 1 shows a schematic diagram of the apparatus. the trapping or cooling light is provided by an external-cavity-stabilized diode laser locked 12 MHz to the red of the $^{85}\mathrm{Rb}\ 5S_{1/2}(F = 3) \rightarrow 5P_{3/2}(F' = 4)$ transition [9]. The diode laser is modulated at 2.91 GHz [11] to produce sidebands at the $5S_{1/2}(F = 2) \rightarrow 5P_{3/2}(F' = 3)$ transition needed to keep the atoms pumped into the $F = 3$ ground level. The laser beams are 1.1 cm in diameter with a total power of about 5 mW. Coils on the outside of the chamber provide the magnetic field gradient (28 G/cm) needed for the trap. The trap produces a roughly spherical cloud less than 0.5 mm in diameter with 10^6 atoms at about 100 μK temperature. The number of trapped atoms is proportional to the fluorescence intensity of the cloud, which we image onto a photodiode and record the current on a digital oscilloscope. An electrostatically focused electron gun [12] produces a repetitively pulsed electron beam of 2.5 to 9 mm in diameter (FWHM), with total current of 200 to 650 μA, and with pulse widths of 0.16—2.0 ms.

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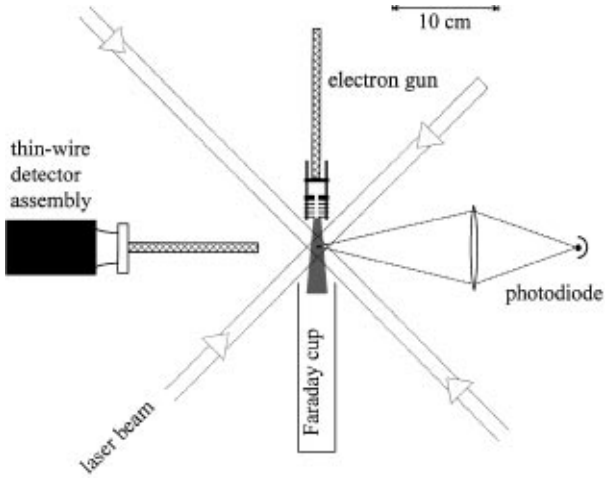

FIG. 1. Scale drawing of the trapping and collision region. Not shown are two of the laser beams, the magnetic field coils, and the diode laser with its associated stabilization and modulation equipment.

A gimbal mount allows the beam to be precisely aimed. The electron beam energy and its spread (~0.5 eV) were determined using a retarded-potential difference method.

Figure 2 shows the time sequence for one trap cycle. At $t = 0$, the current to the magnetic field coils is switched off and the field decays with a 0.4 ms time constant. The 2.91 GHz modulation is concurrently shifted by 200 MHz which moves the laser sideband out of resonance with the $F = 2 \rightarrow F' = 3$ transition. Spontaneous Raman scattering quickly (0.3 ms) pumps the atoms into the $F = 2$ ground level which is unaffected by the trapping or cooling laser, thus turning off the cooling action of the laser. By $t = 1$ ms the magnetic field is gone and the atoms are no longer excited by the laser. Negligible expansion of the cloud occurs during this time. At $t = 1$ ms a short electron beam pulse impacts the atom cloud. Within 1 μs of the electron beam cutoff the trap is turned on again. The remaining neutral atoms are recaptured, returned to the center of the trap, the fluorescence is recorded, and the sequence is repeated.

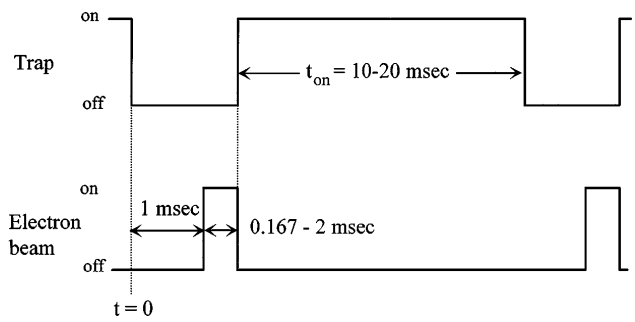

FIG. 2. Timing diagram for one electron-beam pulse cycle. First the trap is turned off by shutting off the magnetic field and changing the laser modulation frequency. Then the electron gun produces a short pulse, following which the trap is turned back on to recapture the remaining neutral atoms.

Since each electron pulse produces a small decrease in the population of the trap, we determine the loss rate by observing slow changes in the number of atoms N in the cloud. The time dependence of N is governed by the rate L at which atoms are captured into the trap from the room temperature background Rb vapor, the loss rate Γ_0, due to collisions with background atoms, and the loss rate $\Gamma_e f$, due to electron-atom collisions, where f is the duty cycle of the electron beam

$$\frac{dN}{dt} = L - (\Gamma_0 + \Gamma_e f)N . \tag{1}$$

Beginning with no atoms in the trap we record the fluorescence signal at the end of each trapping cycle as the trap captures Rb atoms from the background vapor and comes to equilibrium. By fitting two such transients, for the electron beam on and off, by solutions of Eq. (1), we obtain Γ_e. From the measured current density J we find the cross section from $\sigma = e\Gamma_e/J$, where e is the electron charge. The simplicity of this relation between the two measured parameters (J, Γ_e) and σ_{ion} makes the technique especially appealing. We emphasize that no absolute measurement of the target density is required.

Our optical trap has a depth of about 3×10^{-5} eV, so a recoiling (nonionized) atom of sufficiently high velocity (> 8 m/s) may escape the trapping volume. Since the recoil velocity increases with increasing scattering angle, large-angle scattering of nonionizing collisions contributes to the loss rate and is a potential source of error in the ionization measurement. At the energies of this experiment, forward scattering is dominant so corrections due to escape of nonionized atoms from the trap should be small. Using Born approximation calculations for the nonionizing differential cross sections, normalized to experiment, and combined with a model of the trap depth similar to Ref. [13], we estimate such corrections to the ionization cross section to be less than 2% for an infinitely short electron beam pulse. For a pulse of finite duration, the maximum velocity of the atoms that can be retrapped is reduced since the recoiling atoms move freely toward the outer edge of the trapping region until the laser beams are turned back on. To experimentally account for these lost, nonionized atoms we measure the loss rate using different electron-beam pulse widths and extrapolate the results to zero pulse width. The results are shown in Fig. 3. At high energies (250 and 500 eV) the slope is quite shallow, since the differential cross sections are sharply peaked at small scattering angles, where the recoil is small. At low energies the large-angle scattering becomes more prominent, giving a bigger slope to the pulse width dependence. However, the low energy electrons have less energy to impart, so even atoms recoiling with larger scattering angles can be recaptured. As a check, we used the Born approximation differential cross sections and the trap model to calculate the expected slopes of the data in Fig. 3. At 50 eV, where the extrapolation is

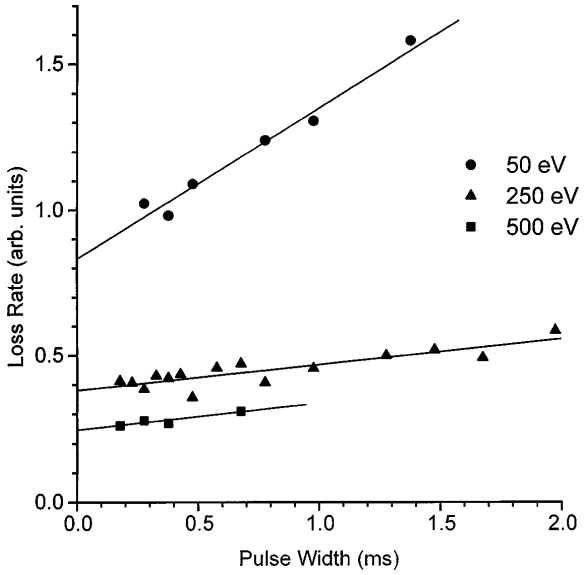

FIG. 3. Measured loss rates as a function of electron beam pulse length.

most important, the calculated slopes agree with the measurements to better than 30%. At all energies reported here, we estimate the contribution from nonionizing processes to the observed cross section at zero pulse width to be a few percent or less.

We determine J by measuring the current on a long, thin tungsten wire translated perpendicular to the electron beam. An Abel transform of this profile, together with the total electron beam current and the cylindrical symmetry of the electron beam, gives the current density at the target. To test this procedure, we simulated one-dimensional scan profiles from a variety of slightly asymmetric functions (which were more asymmetric than our data) for which the peak value was known. In each case the Abel transformation returned peak values that agreed to within 2%. When combined with the uncertainty in the total electron current, we estimate a 7% uncertainty in the current density. We estimate our measurement uncertainty for Γ_e to be 6%, based on our systematic checks and fits of the solution of Eq. (1) to our data. The extrapolation to zero electron beam pulse width contributes a larger uncertainty at lower energies, decreasing from 9% at 50 eV to 5% at 500 eV. Combining the various uncertainties we find that the total uncertainty in our measurements ranges from 13% at 50 eV to 9% at 250 and 500 eV.

In early ionization measurements an electron beam ionized a static alkali vapor target gas, the ions were collected on surrounding plates, then analyzed by a mass spectrometer. Other measurements of ionization cross sections [5,14] used the crossed-beam method [15], in which an electron beam intersects an atomic beam and the resulting ions are extracted and detected. Reference [1] points out that the uncertainty in the determination of the number of target particles may cause uncertainties as large as 30% in the ionization cross sections.

In Ref. [14] Brink reported the absolute cross sections for Rb^+ by measuring the total ion current from all Rb^{n+} ions. Tate and Smith [16] gave the relative values of the positive ion current due to the production of Rb^+, Rb^{2+}, and Rb^{3+} by electron-impact ionization. Using Brink's absolute cross sections for Rb^+ at 200, 300, and 500 eV, we obtain the cross sections of Rb^+ at lower energies and the cross section for Rb^{2+} and Rb^{3+} at energies up to 500 eV from Fig. 5 of Ref. [16] allowing that each Rb^{n+} ion generates n times the positive-ion current of each Rb^+ ion. Adding the Rb^+, Rb^{2+}, and Rb^{3+} cross sections together gives a set of total ionization cross sections ($\sum_n \sigma^{n+}$) shown in Fig. 4 as "Expt. a" which may be compared with our data. McFarland and Kinney [5] also obtained the absolute cross sections for Rb^+ by measuring the total ion current. We combine McFarland's [17] reanalysis of these data with the Rb^+ cross sections of Ref. [16] to obtain another set of total ionization cross sections which correspond to "Expt. b" in Fig. 4. Our cross sections are larger than both set a and set b at low energies but lie between these two sets at higher energies. The difference between our Rb ionization cross section and "Expt. b" is no more than 15% in contrast to a much larger difference in the Na ionization cross section between the Johnston-Burrow and the McFarland-Kinney results as shown in Fig. 1 of Ref. [6]. Nygaard and Hahn [18] reported total electron impact ionization cross sections in the form of $\sum_n n\sigma^{n+}$ rather than $\sum_n \sigma^{n+}$.

Our technique for measuring ionization cross sections differs radically from previous methods. It takes advantage of the unique properties of the MOT to determine the

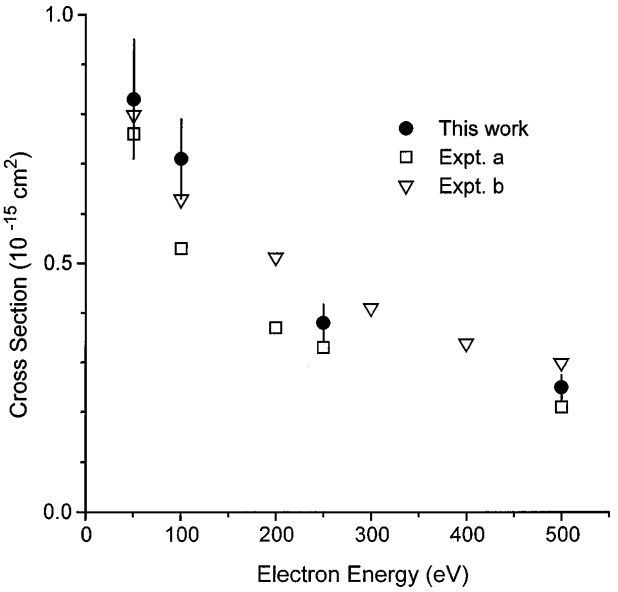

FIG. 4. Total ionization cross section ($\sum_n \sigma^{n+}$) for Rb: this work (•), Expt. a (□), Expt. b (▽). See text for explanation of Expts. a and b.

ionization cross section relying on observation of the nonionized atoms to determine the ionization cross section. To obtain absolute cross sections by crossed-beam measurements it is necessary to determine the target atoms density distribution, the electron beam current, the spatial overlap of the atomic and electron beams, and the ion current resulting from electron-impact ionization. Even for ground state atoms, it is generally difficult to determine the absolute number accurately. The required measurement of the spatial distribution of the target atoms makes the determination of the spatial overlap of the atomic and electron beams difficult. On the other hand, in our new method only relative target atom numbers are needed, along with the electron beam current density at the trapped atom cloud. This avoids measuring the absolute number of target atoms and the overlaps of the atomic and electron beams.

We previously used the MOT to measure the total electron scattering cross section [19]. In that experiment, the trap remained off after the end of the electron beam for a long enough "waiting" time to allow slowly recoiling atoms to escape the trap, so the electron-induced loss rate of the trapped atoms gave the total scattering cross section. In that work, the main advantage of the MOT target as compared to the atomic beam recoil technique [20] is to provide a determination of the contribution of small-angle scattering to the total cross section which is difficult with cross-beam experiments. In the present work, we run the trap cycle with zero waiting time so as to recapture all the scattered nonionized atoms. That the ionized atoms can be sorted out from the others by changing the modulation cycle shows the versatility of the trapped-atom target. In a related application, Dinneen *et al.* [21] measured photoionization cross sections with a MOT.

A direct extension of this new method would measure electron-impact ionization cross sections for atoms in excited states. During the off cycle of our present experiment (Fig. 2), we turn off the magnetic field and move the laser frequency out of resonance with the pumping transition. If instead the laser is unaltered, the significant fraction of the atoms in the $5p$ level will make it possible to measure the ionization cross section of the Rb($5p$) atoms. Such studies are of current interest. For electron-impact ionization of He(2^3S) atoms, fair agreement was found between experimental measurements [8] and theoretical calculations based on binary-encounter and Born-type approximations [22,23], but the newly developed CCC method gives cross sections [7] much smaller than the experimental values. Measurements for other excited atoms should be especially interesting.

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