Absolute measurement of the photoionization cross section for ground-state cesium atoms*

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Measurements of the absolute cross section for the photoionization of cesium atoms near threshold are reported. The results agree well with recent model potential calculations and are approximately a factor of 2 smaller than earlier experimental results.

I. INTRODUCTION

The importance of the alkali metals in a variety of laboratory and astrophysical processes[1] has stimulated many investigations of their properties. A convenient method for theoretical study of such species is to approximate the distribution of core electrons with a model potential, thereby reducing the many-electron problem to a more tractable form. In two recent papers, Weisheit[2] and Norcross[3] have reported such calculations for photoionization of ground-state cesium, which is the most difficult alkali metal to treat theoretically because of the importance of both spin-orbit and core-polarization effects.

While the two calculated cross sections agree closely, they nevertheless disagree significantly with the results of previous experiments.[4-6] The most recent experimental data due to Marr and Creek[6] are, for example, approximately a factor of 2 larger than the calculated values. In an effort to resolve this discrepancy, a new measurement of this cross section has been performed using a crossed-beams apparatus which eliminates many of the potential difficulties in the previous experiments, all of which were performed in vapor cells.

II. EXPERIMENTAL

The apparatus is shown schematically in Fig. 1. Briefly, a cesium atom beam is crossed by a beam from a frequency doubled dye laser. A small fraction of the illuminated atoms are ionized and detected by a calibrated particle multiplier. The photoionization cross section, σ, is then given by the usual small signal expression:

$$\sigma = S/knlI\,, \tag{1}$$

where S is the number of ions counted per second by a multiplier of efficiency k. The number density of target atoms is n, the target beam thickness is l, and the number of photons passing through the alkali beam per second is I. The measurement of each of the quantities on the right side of Eq. (1) is now discussed together with a more detailed description of the apparatus.

The cesium-atom beam is formed by effusion from a stainless steel and copper oven which is typically operated at temperatures of 50–80 °C. Owing to the large photoionization cross section for dimers, Cs_2, relative to atoms,[7] it is important that the dimer content of the beam be small. At the maximum oven temperature used in the experiment, the relative dimer concentration is estimated to be $\lesssim 0.07\%$.[8]

A flag located immediately in front of the oven orifice may be used to block the beam, while a series of large apertures provides collimation to the beam and reduces the gas load on the reaction chamber. Final collimation is provided by an aperture mounted on the ion-collector assembly. The width of this aperture together with knowledge of the geometry of the system is used to calculate the beam width l at the point of intersection with the laser beam. After passing through the interaction region, the alkali beam is deposited on a cooled copper surface. Immediately prior to striking this surface, the atom flux may be sampled by either of two tungsten hot-wire ionizers mounted at right angles to each other on a movable scanner. By moving each of the wires successively through the cesium beam and recording the resulting ion currents the profile of the beam may be determined. This procedure is used to ensure that the beam apertures are properly aligned and are not clogged by cesium deposition. From these measurements it is observed that the beam is highly uniform and in consequence the surface ionization current i is related to the number density n of atoms at the interaction region by an equation of the form

$$n = ig\langle 1/v\rangle/e\,\delta\,, \tag{2}$$

where g is a geometrical factor related to the dimensions of the beam and size of the hot-wire ionizer, e is the electron charge, and δ is the

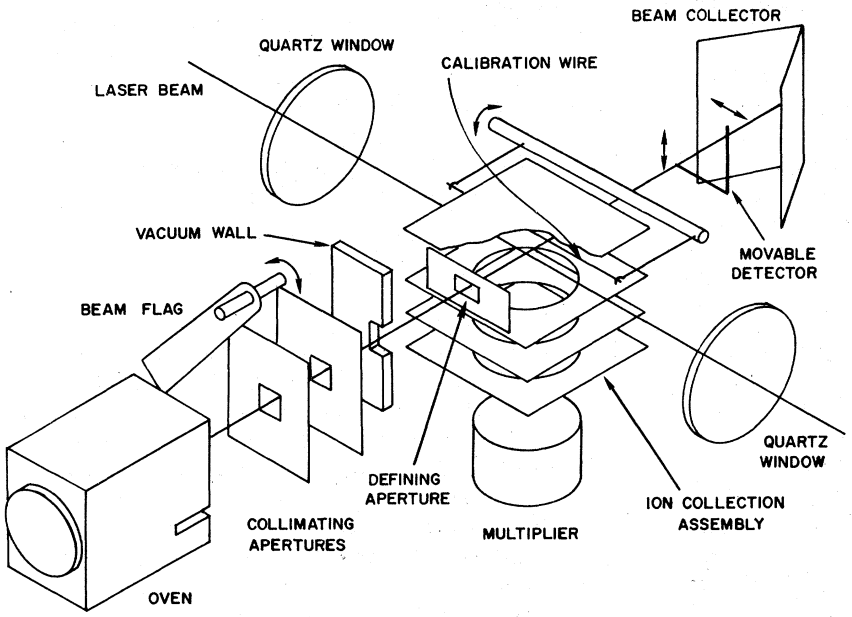

FIG. 1. Schematic of the apparatus.

surface ionization efficiency of cesium atoms on tungsten (assumed to be unity).[9] The mean inverse velocity $\langle 1/v \rangle$ is calculated assuming a velocity distribution appropriate to effusive flow from an orifice of known temperature. Although deviations from such a distribution, due to the scattering of slow atoms from the beam, have been shown to be significant in certain experiments,[10] such effects are not important here,[11] as the present cesium beam is short, has a large cross-sectional area and a low number density, typically $\sim 10^6$ cm^{-3}.

Radiation in the near uv is required to photoionize ground-state cesium atoms. This radiation is provided by frequency doubling the output of a nitrogen pumped dye laser. This laser system[12,13] is operated at a repetition rate of 10 pulses per second and yields output powers of several kilowatts and a pulse width of ~5 nsec. The photon beam enters the machine through a fused quartz window, intersects the alkali beam at the interaction region and exits the machine through a second fused quartz window. The photon flux, which is typically $\gtrsim 10^{13}$ photons sec^{-1}, is determined from measurement of the average laser power by use of an Eppley thermopile which was absolutely calibrated by the manufacturer prior to delivery. In normal use the thermopile is positioned to intercept the laser beam immediately in front of the entrance window. Allowance for the reflection losses at the entrance window then permits the photon flux at the interaction region to be determined. Measurement of the photon flux emerging from the apparatus shows that no transmission losses occur other than those resulting from reflection at the windows.

Ions formed in the interaction region are extracted by a field of 50 V/cm and pass through a series of grids where they are accelerated to 5 keV prior to detection by a Johnston particle multiplier. The photoions S are distinguished from ions due to other sources by chopping the alkali beam, and by appropriately gating the counting electronics relative to the laser pulses. Two sources of extraneous ions are the photoionization of residual gas, particularly cesium, and the photodesorption of surface ions. In order to reduce the count rate due to the first of these, a liquid-nitrogen-cooled trap is located in the reaction chamber to reduce the density of background cesium atoms. The count rate due to liberation of surface ions is observed to be small provided the surfaces of the ion collector assembly are not directly illuminated by the laser beam.

As mentioned previously the output pulse width of the present laser system is only ~5 nsec and, as a result, if two or more photoions are produced during a single laser pulse, they are not resolved by the ion detection system. The experimental conditions are therefore chosen such that the photoion count rate is $\lesssim 0.1$ per laser pulse, and the probability of the simultaneous production of two or more photoions is then negligibly small.

An important facet of the experiment is the determination of the absolute counting efficiency, k, of the ion-detection system. To accomplish this an additional hot-wire ionizer, located in the ion-collector assembly, is moved into the cesium beam at the region normally traversed by the

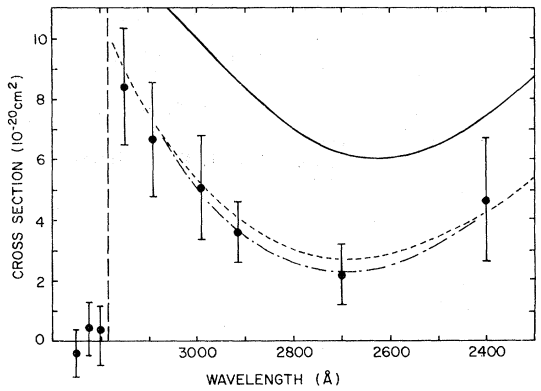

FIG. 2. Summary of atomic cesium photoionization work. ———, experiment of Marr and Creek (Ref. 6); — — —, calculation of Weisheit (Ref. 2); — . —, calculations of Norcross (Ref. 3); ⊕, this work.

laser beam. Cesium atoms which strike the wire are surface ionized and these ions are extracted from the interaction region using the same bias conditions as in the photoionization measurement. They are then accelerated to 5 keV before impacting on the multiplier, the first element of which is operated at ground potential. The production rate of these cesium ions is then chosen so that they may be measured either as a current to the first dynode or, when the multiplier high voltage is applied, as a count rate. The ratio of these two quantities gives the counting efficiency, k, of the detection system for cesium ions of given energy:

$$k = \frac{R}{j/e}, \tag{3}$$

where R is the dead time corrected count rate, j is the measured current, and e is the electron charge. In the measurement of R, the multiplier gain and the external amplification are increased until the count rate saturates indicating that all ions liberating secondary electrons at the first dynode of the multiplier are being counted. These gain and amplification conditions are also used in the photoionization experiment. As expected, the value of k is found to be independent of the ion-count rate. k was determined at frequent intervals during the course of these measurements and was observed to decrease slowly over a period of weeks, presumably as a result of dynode contamination, from an initial value $k = 0.62$.

III. RESULTS AND CONCLUSIONS

The present results are presented in Fig. 2. The points shown represent an average of several measurements at each wavelength. The error bars include both the standard deviation of the mean, and the rms systematic error. The principal source of systematic error in these measurements is associated with the determination of the cesium atom beam density, n, and results from uncertainties in the ionization efficiency of the hot-wire detector, the diameter of the wire, the cross-sectional area of the beam, and in the quantity $\langle 1/v \rangle$. These combine to produce a resultant rms uncertainty of ±10%. Lesser sources of systematic error include that present in the determination of k, estimated to be ±5%, which results primarily from the dead time correction of R, and in the determination of I, estimated to be ±5%.

Several cross-section determinations were made at photon energies below the atomic threshold. The cross section for photoionization of molecules is known to be a maximum in this region,[7] and the small magnitude of the effective cross section at these energies therefore indicates that dimers are present in negligible amounts. This result is consistent with estimates of the dimer concentration inferred from the oven conditions.[8]

Also shown in the figure are the results of the previous measurements due to Marr and Creek, and the calculations of Weisheit and of Norcross. While the earlier measurements differ significantly from the calculations, the present results appear to substantiate the theoretical values.

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