# Absolute measurement of the photoionization cross section of the excited 7S state of cesium

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We report the first measurement of the absolute cross section for photoionization of the 7S state of cesium. The measurement employed a new technique in which the density of the excited-state atoms was determined by the amount of fluorescence. The cross section for photoionization by 540-nm light is $1.14(10)\times10^{-19}$ cm$^2$. We also propose a second new technique for the absolute measurement of photoionization cross sections which is based on modulated fluorescence.

## I. INTRODUCTION

The photoionization behavior of alkali-metal atoms has been a subject of considerable theoretical and experimental study. Cesium has been of particular interest lately because the effects of spin-orbit interaction and core polarization are more pronounced than in the lighter alkali metals. This has been quite significant because the spin-orbit interaction gives rise to the Fano effect which is used for producing polarized electrons. However, these effects make the theoretical calculation of the photoionization cross section more difficult, particularly for $S$ states. Simple quantum-defect theories which are adequate for light alkali metals give rather poor agreement with experimental results for cesium. Weisheit[1] and Norcross[2] have carried out semiempirical calculations in which they included both core polarization and the spin-orbit interaction, while *ab initio* calculations by Chang and Kelly,[3] Johnson and Soff,[4] and Huang and Starace[5] treated the spin-orbit interaction but neglected core polarization. Lahiri and Manson[6] have done a simple Hartree-Slater calculation for a number of low-lying states. Attempts to check the accuracy of these theoretical approaches have been limited by the lack of accurate and consistent experimental data on absolute cross sections. In modern times there have been only three absolute measurements of photoionization cross sections for $S$ states of cesium, all of which were for the 6S ground state. Marr and Creek[7] obtained results for the ultraviolet region of the spectrum which were about a factor of 2 larger than the values calculated by either Weisheit or Norcross. However, Cook *et al.*[8] repeated this measurement and obtained values in agreement with those calculations but with uncertainties of $\pm30\%$. Grattan *et al.*[9] have also measured this cross section at a wavelength in the vacuum ultraviolet with similar results.

We report here the absolute measurement of the cross section for the 7S state accurate to $\pm9\%$. This measurement used the new technique of "fluorescence normalization" in which the excited-state atomic density was determined from the amount of fluorescence. This technique avoids the uncertainties in determining the molecular background and the ground-state atomic density which have limited these previous absolute measurements.

We shall conclude with the discussion of another technique for the absolute measurement of photoionization cross sections. The technique proposed is an extension of fluorescence normalization but under some conditions has substantial advantages over fluorescence normalization and other techniques which have previously been used.

## II. EXPERIMENTAL METHOD AND APPARATUS

For this measurement a beam of cesium atoms in an electric field was excited to the 7S state by a cw dye laser. This normally forbidden transition is allowed due to the small (5 parts in $10^5$) mixing of $S$ and $P$ states by the static electric field. The number density of 7S atoms was determined from the amount of fluorescence emitted as the atoms spontaneously decayed. The laser radiation also caused a small fraction of the 7S atoms to be photoionized, and the resulting ion current was measured. Strictly speaking, the process observed is two-photon photoionization with a resonant intermediate state. It is an extremely good approximation to treat this as two single-photon processes, however, because both the $6S\rightarrow7S$ transition and the $7S\rightarrow$continuum transition were far from saturation. The density of excited atoms was $10^{-4}$ times the density of ground-state atoms, and only 1 part in $10^4$ of the excited atoms was photoionized. This allows the 7S photoionization cross section to be determined from the fluorescence signal, the laser power, and the photoionization current.

The apparatus is shown schematically in Fig. 1. A highly collimated beam of atomic cesium intersected a standing-wave laser field at right angles in a region of static-electric field. The cesium beam was produced by a two-stage oven to reduce the dimer fraction. The output nozzle was a microchannel plate which produced a beam with a cross section $0.5\times2.5$ cm$^2$ and a half-angle divergence of $\sim0.05$ rad. This passed through a multislit collimator which reduced the divergence in the direction of the laser beam to 0.013 rad. A final 2-cm-wide aperture just before the intersection with the laser beam provided a precisely defined intersection geometry 2.0 cm long with a diameter equal to that of the laser beam (0.05 cm). The cesium beam density ($\sim5\times10^9$/cm$^3$) was uniform to

 

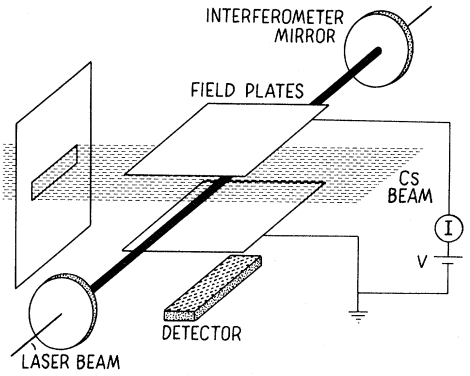


FIG. 1. Schematic of apparatus.

within a few percent over the intersection region because the total distance from nozzle to intersection (6 cm) was much shorter than the distance scale for beam-density redistribution along the direction of the laser (100 cm). This uniformity was confirmed using a hot wire detector.

The standing-wave laser field was produced by an amplitude- and frequency-stabilized cw ring dye laser mode-matched into an electronically tunable, semiconfocal Fabry-Perot interferometer buildup cavity. The laser had a typical output of 250 mW and a short-term frequency jitter of 0.5 MHz peak to peak. With the resonant frequency of the interferometer locked to the laser frequency, the electromagnetic field strength inside the interferometer corresponded to two linearly polarized traveling waves, each with 116 times the power incident on the buildup cavity.

A calibrated photodiode measured the light transmitted through the cavity. The intensity profile for the laser beam in the cavity is that of the lowest-order eigenmode of a semiconfocal cavity with a 25-cm focal-length curved mirror.[10] This is a Gaussian profile with a spot-size radius of 0.021 cm at the input mirror and 0.029 cm at the output mirror. Less than 1% of the power was contained in higher-order modes.

The $7S$-state population was monitored by observing the 850- and 890-nm light that was emitted in the $6P$-$6S$ step in the $7S$-$6P$-$6S$ cascade decay. This light was detected by a cooled silicon photodiode $0.5\times5.5$ cm$^2$ that sat 9 mm below the interaction region. Glass color filters not shown in Fig. 1 prevented scattered 540-nm laser light from reaching the detector.

The static-electric field was produced by applying voltage to the top field plate and grounding the lower. The laser and cesium beams intersected in the middle of the $5\times7.5$ cm$^2$ electric field region. The top field plate was coated with evaporated gold and the lower field plate was coated with an electrically conducting optically transparent (84%) coating. The photoionization current was measured using an ammeter in the line providing voltage to the top field plate.

Data were obtained by scanning the laser over the $6S_{F=4}\rightarrow7S_{F=4}$ Stark-induced transition and simultaneously recording the fluorescence signal and the photoionization current. Typical data are shown in Fig. 2. Scans

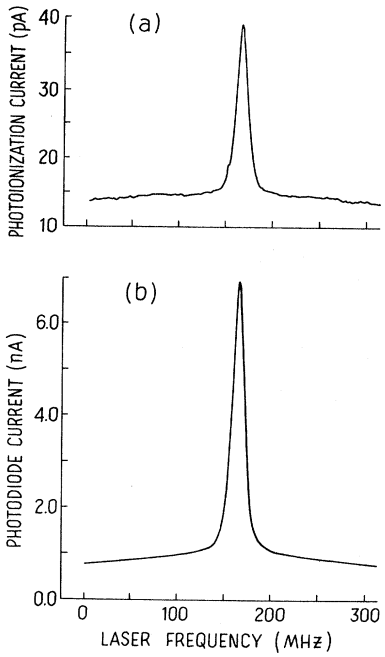


FIG. 2. (a) Photoionization current vs laser frequency. (b) Fluorescence detector current vs laser frequency. Zero of the laser frequency scale was chosen arbitrarily.

were made at several voltages between 1500 and 3000 V/cm and with both positive and negative voltage. The fluorescence line shape is composed of a narrow (14 MHz) resonance peak superimposed on a low broad (575 MHz) pedestal. The pedestal arises from a diffuse background vapor of cesium in the interaction region and is discussed in more detail in Ref. 11. The photoionization current has an identical line shape but it has an additional background component which is independent of laser frequency.

The $7S$ number density was calculated from the $7S$-state lifetime and the total emitted fluorescence. The determination of the total fluorescence is inherently the least accurate and aesthetically the most unpleasant part of the experiment, as it involves finding the detector quantum efficiency and the detection solid angle. The detector size, uniformity, and angular dependence of the response were measured. The quantum efficiency was given as 0.80(4) by the manufacturer.[12] The detection solid angle was calculated by numerical integration using the geometry of the apparatus and the measured transmission of the lower field plate and filters at various angles of incidence. This included the light which was reflected off the upper field plate, the reflectivity of which was measured. There was a negligible contribution due to reflection off other surfaces in the apparatus since these were all far from the interaction region and painted flat black. The laser power in the buildup cavity was determined by dividing the transmitted power by the transmission coefficient for the output mirror. The power measurement was made using a photodiode calibrated against a Coherent Inc. power meter recently calibrated to a NBS standard.

## III. RESULTS

The total photoionization current is given by

$$I = e \int_{\vec{r}} S(\vec{r}) n(\vec{r}) \sigma \, d\vec{r} \,, \tag{1}$$

where $S(\vec{r})$ is the laser photon flux, $n(\vec{r})$ is the density of $7S$ atoms, and $\sigma$ is the photoionization cross section of interest. The fluorescence signal is

$$F = \int_{\vec{r}} n(\vec{r})(1/\tau)\xi \, d\vec{r} \,, \tag{2}$$

where $\xi$ is the detection efficiency and $\tau$ the $7S$ lifetime.[13] As mentioned previously, the $6S \rightarrow 7S$ excitation rate and $7S$ photoionization rate are much smaller than $1/\tau$. This allows us to assume that $n(\vec{r})$ has the same Gaussian form as $S(\vec{r})$. With this substitution, Eqs. (1) and (2) can easily be solved to yield

$$\sigma = \frac{2I\xi\pi w^2}{FS_0\tau} \,, \tag{3}$$

where $S_0$ is the total number of laser photons per second and $w$ is the Gaussian beam radius at the interaction region. Our measurements give the cross section in $cm^2$ as

$$\sigma = 1.14(10) \times 10^{-19} \,.$$

The significant contributions to the 9% uncertainty are 5% for detector quantum efficiency, 6% for laser power, 3% in measurement of photoionization current, 3% for solid angle, and 2% for the $7S$ lifetime.

There are several possible sources of additional systematic error which we have considered. First, there was a background current which we attribute primarily to photoemission from surfaces; however, this background was eliminated in the analysis by using only the 14 MHz-wide components of the measured currents shown in Fig. 2. A second possible source of error was multiplication of the photoionization products through collisions with neutral atoms or secondary surface emission. This can be ruled out by the observation that the measured photoionization cross section was independent of the strength of the applied field. At the highest field there was a substantial increase in the noise on the photoionization current, however, suggesting some multiplication or arcing phenomena. Therefore only the values obtained at the lowest electric field (1500 V/cm) were used in obtaining $\sigma$. Another possible source of systematic error is anisotropic radiation trapping. Calculations indicate this should be quite small. However, we also checked this empirically by measuring the ratio of fluorescence signal to beam density as a function of beam density. When the density was increased from 5 to 40 times $10^9$ atoms/$cm^3$ this ratio decreased by less than 20%. This indicates that radiation trapping is not a significant effect for the density ($5 \times 10^9/cm^3$) at which the photoionization measurement was made.

The cross section we obtain can be compared with the theoretical value obtained by Lahiri and Manson.[6] Their calculation predicts a value about one half of what we measure. However, 540 nm is in a region where their calculated cross section has a very strong dependence on energy (wavelength) so that an error of only 0.001 eV in their value for the Cooper minimum would explain the discrepancy. They believe the uncertainty in the calculation of this minimum is considerably larger than 0.001 eV.

## IV. EXTENSIONS OF PRESENT WORK

The present work has provided the first measurement of a photoionization cross section for an excited $S$ state of cesium. This provides a good test of the different theoretical treatments of photoionization of cesium, a better test than has been possible using the less accurate and disparate values measured for the ground state. However, it is obviously desirable to measure the dependence of this cross section on wavelength. This could be done using the fluorescence normalization technique if a second laser was used to do the photoionization. The obvious choice for a second laser would be a relatively high-power pulsed laser. However, such a laser would allow the use of a new technique for measuring the cross section which is something of a hybrid between fluorescence normalization and the popular saturation technique,[14] but can have significant advantages over both approaches.

This technique, henceforth called modulated fluorescence, could be used quite generally for determining excited-state photoionization cross sections. In the modulated fluorescence technique one would excite the atoms to the state of interest with one laser and monitor the change in the fluorescence when the photoionizing laser (or lamp) is pulsed. The photoionization rate would then be simply the fractional change in the fluorescence signal divided by the lifetime of the state. The only quantities which must be determined absolutely are the lifetime of the state and the photoionizing laser intensity. In this respect it is similar to the saturation technique and superior to fluorescence normalization. However, it has a significant advantage over the saturation approach in that it requires considerably lower photoionization rates and hence lower laser power. In the saturation method it is necessary to achieve ionization rates which are at least comparable and preferably several times larger than the spontaneous decay rate. However, the modulated fluorescence technique will work with photoionization rates which are a small fraction of this. The necessary fraction is ultimately limited by the signal-to-noise ratio of the fluorescence measurement, but this ratio is characteristically quite high. There are many cases where the necessary photoionization rate, $R_{PI}$ could be $10^{-2}$–$10^{-3}$ of the decay rate.

The three techniques mentioned are thus complementary to each other since each has a range of experimental conditions where it is generally superior. If unlimited laser power is available ($R_{PI} > 1/\tau$) the saturation technique would usually be best because it is usually easier to measure photoionization current than fluorescence. For an intermediate rate [$10^{-3}(1/\tau) < R_{PI} < 1/\tau$] the modulated fluorescence technique would be superior. Finally, for even lower photoionization rates, such as in the experiment reported, the problem of obtaining a sufficiently

high signal-to-noise ratio to observe the modulated fluorescence becomes worse than the additional difficulty of determining the absolute fluorescence detection efficiency. In this case the fluorescence normalization technique would be the best choice.

## ACKNOWLEDGMENTS

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