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Photoionization of Helium Metastable Atoms near Threshold*

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We have experimentally determined the absolute cross sections for the photoionization of He(2^1S) and He(2^3S) metastable atoms from threshold to 2400 Å. The results are compared with recent theoretical calculations.

Several calculations of the photoionization cross sections for He(2^1S) and He(2^3S) atoms have been reported.[1-4] The recent calculations are in substantial agreement, although they have slightly different wavelength dependence and threshold values. The present experiment, which provides the first measurements of photoionization from a rare-gas metastable state, was undertaken in order to establish a technique which can be used for other more complicated systems, and to assess the relative merits of the various theories.

The apparatus used for these measurements has been described in detail elsewhere[5] and only a brief description is included here. A beam of 2^1S and 2^3S metastable atoms is produced by electron impact and may then be irradiated with the light from a helium discharge lamp which causes the He(2^1S) metastables to be quenched via transitions of the type $2^1S \rightarrow n^1P \rightarrow 1^1S$. The He($2^3S$) atoms are not quenched since the 2^3S state is the lowest state in the triplet system. Data appropriate to each metastable species may then be derived from observations with the helium lamp alternately on and off. After removal of charged particles the beam is irradiated by the output beam of a pulsed laser. Ions formed are extracted by an electric field and detected with a Johnston particle multiplier. To discriminate against ions resulting from collisions of the metastable atoms with the background gas the output from the multiplier is fed to two scalers. One of these is gated so as to count those ions produced during the laser pulse, while the other is gated for an equal time interval when no laser pulse is present and therefore counts only ions formed from background gas. The difference in the two scaler count rates is then due solely to photoions. The metastable atom flux is determined by measuring the current of secondary electrons ejected from a surface whose absolute secondary-electron ejection coefficient γ is determined *in situ* using a technique described elsewhere.[6]

The ultraviolet radiation is obtained by frequency doubling the output of a tunable dye laser operating at 10 pulses/sec in the visible. This dye laser is pumped by a nitrogen laser and, with a suitable choice of dye or dye mixture,[7,8] output powers in excess of 50 kW at a linewidth of 1.5 Å are attained throughout the visible spectrum. Frequency doubling is achieved in a nonlinear crystal of either ammonium dihydrogen phosphate[9] or lithium formate monohydride.[10] The resultant ultraviolet beam has a linewidth of 0.5 Å, a pulse power of several kilowatts, a pulse width of 5 nsec, and a cross-sectional area at the interaction region of 4 mm^2. A thermopile is used to measure the mean power and hence mean photon flux in the laser beam. In practice two thermopiles are utilized; one is used to monitor the ultraviolet flux while the second, which was absolutely calibrated by the manufacturer before de-

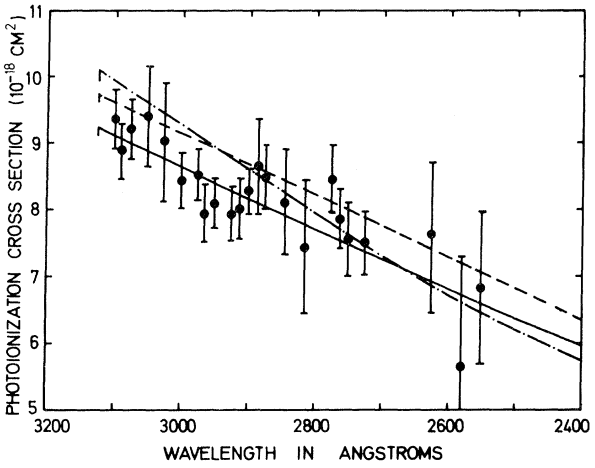

FIG. 1. Photoionization cross section for He(2^1S) atoms as a function of wavelength. Closed circles, present results; solid line, Jacobs's results; dash-dotted line, Norcross's results; dashed line, Burgess and Seaton's results.

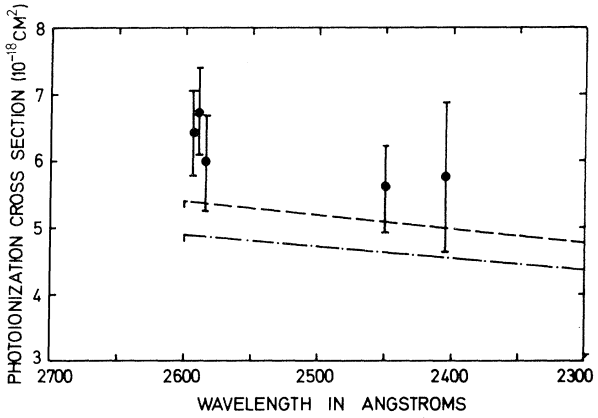

FIG. 2. Photoionization cross section for He(2^3S) atoms as a function of wavelength. Closed circles, present results; dash-dotted line, Norcross's results; dashed line, Burgess and Seaton's results.

livery, is employed as a standard against which the first is periodically referenced using a low-power cw laser. The sensitivity of the monitor thermopile remained constant during the course of this study indicating that it is not damaged by the high-power pulsed radiation. The linearity of the thermopile was checked by placing neutral density filters in the beam These tests, together with the observation that, at a fixed wavelength, the photoion production rate is directly proportional to the measured photon flux, show the detector response to be linear over the range of input powers used in the present experiment.

The cross section Q for photoionization is given by

$$Q = S/(FNlk),$$

where S is the mean photoion count rate, typically 0.1 to 2.0 sec^{-1}, F is the mean photon flux, and l is the path length of the photons through the metastable beam. N, the metastable atom density, is of the order of 10^3 cm^{-3} and is determined from measurements of the velocity distribution and total flux of the metastable atom beam. The efficiency k with which the signal ions are detected is not measured, although the value $k = 0.95 \pm 0.05$ may be inferred from the data of Rundel, Aitken, and Harrison.[11] The uncertainties in the velocity distribution of the metastable atoms, in γ, and in the absolute calibration of the Eppley thermopile lead to an rms systematic error in the cross sections of $\pm 14\%$.

The experimental results are shown in Figs. 1 and 2, which also include the theoretical results. The error bars represent 1 standard deviation of the mean in the observed count rates, with no systematic error included.

The results for He(2^1S) atoms are seen to be in excellent agreement with those obtained theoretically. The uncertainties are such that the results cannot be said to strongly favor any particular theory. However, the variation of the cross section with wavelength is seen to be in considerably better agreement with that calculated by Burgess and Seaton,[2] and Jacobs[4] than with that of Norcross.[3]

The experimental results for He(2^3S) are somewhat greater than those predicted theoretically, although the inclusion of a $\pm 14\%$ systematic error would cause the error bars on the data to overlap the theoretical values of Burgess and Seaton. It should be noted, however, that the experimentally determined ratio of the singlet to triplet photoionization cross sections at wavelengths above the triplet threshold should be much less susceptible to systematic error than either of the separate cross sections. This ratio is not in agreement with that predicted theoretically and it is felt therefore that there may indeed be a discrepancy which warrants further theoretical study of these processes. It is interesting to note that the measured He(2^3S) photoionization cross section is nearly equal to that of He(2^1S) at the same wavelength.

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Hyperfine-Structure Studies of Highly Excited D and F Levels in Alkali Atoms Using a cw Tunable Dye Laser*

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Highly excited S, D, and F levels in cesium and rubidium have been populated utilizing a two-step optical excitation method. In the first step an rf lamp is used to transfer atoms into the first P states, and the subsequent excitation is performed with the tunable radiation of a cw dye laser. We give results for the hyperfine structure of several D and F states in Cs^{133} and Rb^{87}, obtained in level-crossing and optical double resonance experiments.

Until recently, accurate hyperfine structure studies of excited states in alkali atoms have been confined to the sequences of ^2P levels, which are readily available for observation through direct excitation with resonance radiation. Detailed studies of P levels have revealed several effects which cannot be explained by a simple, one-electron picture of the alkali atoms. These include magnetic core polarization, electric-quadrupole shielding, and possible deviations from expected Landé g_J factors.[1] Clearly, a corresponding knowledge of the properties of non-P states is of great interest for the understanding of the various atomic interactions.

It is well known that the D-state fine-structure intervals are often anomalously small and even inverted, and recent experimental work has shown that similar, large anomalies occur in the D-state hyperfine structure. Data on D-state hyperfine structure are still quite limited, and F-state data are nonexistent. The scarcity of data can be traced to inadequacies of existing experimental methods. Archambault *et al.*[2] have made rough estimates of the hyperfine structure of the $5^2D_{5/2}$ state in Na^{23} and the $9^2D_{5/2}$ state in Cs^{133}, which were produced by electron bombardment. With the recently introduced cascade decoupling[3] and cascade rf spectroscopy methods[4] the Columbia group has investigated the hyperfine structure of several S and D levels in the alkali atoms.[5] In these experiments the states are produced by cascading from a P level, excited by a conventional rf lamp. However, for reasons of intensity, this method is limited to comparatively low-lying S and D levels as the absorption oscillator strengths in the S-P transition sequence decrease very rapidly with increasing P-state energy, and conventional lamps are not very efficient sources for the higher resonance lines. To get access to more highly excited non-P levels we have utilized a two-step excitation method,[6] taking advantage of recent advances in the technology of tunable lasers. In the first step the strong D_1 and D_2 lines from a powerful rf lamp are used to transfer atoms from the ground S state to the first excited P states. In the second step, the intense tunable radiation from a cw dye laser, operating with rhodamine 6G, is used to excite atoms from a P level to a highly excited S or D level. Absorption of light by atoms in the $5^2P_{3/2}$ level of