## Photoionization of the 7<sup>2</sup>P Excited States of Cesium

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Angular distribution and spin polarization of photoelectrons produced by linearly polarized light from the  $7^2P$  excited states of Cs have been measured. Combination with earlier cross-section measurements of other authors allows a complete determination of the transition matrix elements describing the photoionization process for the  $7^2P_{1/2}$  state.

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A common feature of recent research in different areas of atomic physics is the effort toward "complete" or "perfect" experiments.<sup>1,2</sup> One is no longer satisfied with measurements yielding just the moduli of the transition amplitudes that describe the processes being studied, but one also wants to know their relative phases in order to gain all information which is attainable. Measurements of spin polarization,<sup>3</sup> angular distributions,<sup>4</sup> angular correlations,<sup>5</sup> and experiments with oriented targets<sup>6</sup> have been made to this end. One of the processes being studied in this context by our group is the photoionization of atoms and first results have been reported<sup>7</sup> for atoms in their ground states.

Recently a theoretical analysis of one-electron systems<sup>8</sup> has shown that complete sets of experiments are generally possible for photoionization of the excited atomic states. As an example the  $7^2P_{1/2}$  excited state of Cs will be discussed in the following and our measurements of the angular distributions of the photoelectrons and of their spin polarization will be reported for the  $7^2P_{1/2}$  and  $7^2P_{3/2}$  states.

In the experiments, the  $7^2P_{1/2}$  state has been excited and subsequently ionized by the light of a single dye laser. According to the selection rules of electric dipole radiation, the photoionization process leads to the  $\epsilon^2 S_{1/2}$  and  $\epsilon^2 D_{3/2}$  continuum states. The process

 $Cs(7^2P_{1/2}) + \hbar \omega \rightarrow Cs^+ + e^-(1.5 \text{ eV})$ 

is therefore described by only two complex transition amplitudes which we call  $M_s$  and  $M_p$ . They are defined by Jacobs<sup>9</sup> and are essentially the reduced dipole transition matrix elements.

A complete experimental determination of the transition amplitudes yielding results for  $|M_{s}|$ ,  $|M_{D}|$ , and the relative phase  $\varphi_{s} - \varphi_{D}$  is possible with the following set of four measurements:

(1) The total photoionization cross section for  $\pi$  light:  $\sigma_{tot}(\pi)$ .

(2) The ratio of total cross sections for circularly and linearly polarized light:  $R = \sigma_{tot}(\sigma) / \sigma_{tot}(\pi)$ .

(3) The angular distribution of photoelectrons  $\sigma(\theta)$  for  $\pi$  light.

(4) The spin polarization  $\vec{\mathbf{P}}_{\perp}(\theta)$  of photoelectrons perpendicular to the symmetry plane for  $\pi$  light. The total cross section

The total cross section

$$\sigma_{\rm tot}(\pi) = \frac{1}{6} \left( |M_{\rm s}|^2 + |M_{\rm p}|^2 \right)$$

and the ratio

$$R = \frac{\sigma_{\text{tot}}(\sigma)}{\sigma_{\text{tot}}(\pi)} = \frac{\frac{1}{4} |M_{\boldsymbol{p}}|^2}{\frac{1}{\pi} (|M_{\boldsymbol{s}}|^2 + |M_{\boldsymbol{p}}|^2)}$$

determine the moduli  $|M_s|$  and  $|M_p|$  [c.f., Eqs. (25) and (26) of Ref. 8]. The experimental results of  $\sigma_{tot}^{expt}(\pi) = (6.2 \pm 0.5) \times 10^{-18} \text{ cm}^2$  (Ref. 10) and  $R^{expt} = (1.30 \pm 0.03)$  (Ref. 11) yield  $|M_p^{expt}|$ =  $(1.07 \pm 0.04)a_0$  and  $|M_s^{expt}| = (0.42 \pm 0.02)a_0$ , where  $a_0$  is the Bohr radius. Theoretical values have been obtained using the quantum defect method<sup>12</sup> (QDM) with a modification for cesium.<sup>13</sup> They are  $|M_p^{\text{theor}}| = 1.06 a_0$ , in very good agreement with experiment and  $|M_s^{\text{theor}}| = 0.34 a_0$ . For  $|M_s|$ the agreement is not so good, which is in line with the general observation that the QDM is less reliable for s states.

For a determination of the missing phase  $\varphi_s - \varphi_D$  we have measured the angular distribution  $\sigma(\theta)$  and the spin polarization  $\vec{P}_{\perp}(\theta)$  using linearly polarized light. Taking the direction of the electric vector as quantization (z) axis, theory predicts

$$\sigma(\theta) = [\sigma_{\text{tot}}(\pi)/4\pi] [1 + b_2 P_2(\cos\theta)], \qquad (1)$$

where  $P_2(\cos\theta)$  is the Legendre polynomial.<sup>14</sup> For  $\pi$  light, spin polarization is only possible perpendicular to the symmetry plane spanned by the unit vector in z direction  $\bar{e}_z$  and the wave vector  $\bar{k}$  of the ejected electron:  $\bar{P}_{\perp}=P_{\perp}\bar{e}_{\perp}$ , where VOLUME 45, NUMBER 14

$$\vec{e}_{\perp} = \vec{e}_{\perp} \times \vec{k} / |\vec{e}_{\perp} \times \vec{k}|$$
 and

$$P_{\perp}(\theta) = \left[ \left( \sigma_{\text{tot}} / 4\pi \right) \sigma(\theta) \right] d_2^{\perp} P_2^{(1)}(\cos\theta), \qquad (2)$$

where  $P_2^{(1)}(\cos\theta)$  is an associate Legendre function.

The symmetry parameters  $b_2$  and  $d_2^{\perp}$  are related to the ratio of the moduli  $|V| = |M_s| / |M_p|$  and

to their relative phase  $\varphi_{s} - \varphi_{D}$  according to

$$b_2 = [1.0 - 2.83V \cos(\varphi_s - \varphi_D)] / (1 + |V|^2)$$
(3)

and

$$d_{2}^{\perp} = [1.4|V|\sin(\varphi_{s} - \varphi_{p})]/(1 + |V|^{2}).$$
(4)

Since the relative phase appears either in the cosine or in the sine function, both parameters must be measured to uniquely determine  $\varphi_s - \varphi_D$ , once |V| is known.

The apparatus used for the determination of  $\sigma(\theta)$  and  $P_{\perp}(\theta)$  is shown in Fig. 1. An atomic beam of Cs is crossed with the light from a flashlamp pumped dye laser (Zeiss FL8A). The laser produces pulses of about 0.3 mJ and 400 ns duration, focused into 1 mm<sup>2</sup>. Only electrons ejected into a small angular range pass a 2-mm aperture and are detected. The light is linearly polarized and the polarization vector can be rotated to vary the angle  $\theta$  between the *z* axis and the direction of observation.<sup>15</sup> For the spin experiments Mott scattering at 90 keV by a  $170-\mu g/cm^2$  gold foil is used to analyze the spin polarization. The scattered electrons are detected by a combination of scintillator, Perspex light guide, and photomultiplier (PM) during the laser pulse.



FIG. 1. Experimental setup. Cs, cesium beam; POL, polarizer for the laser light; Z, quantization axis in the direction of the electric field vector;  $\theta$ , angle between quantization axis and direction of observation.

In most of the angular distribution measurements the Mott detector was replaced by a Channeltron. A set of experimental points, normalized to equal light intensity, is shown in Fig. 2 with the statistical errors (single standard deviation). The dotted curve represents a least-squares fit to the experimental points which yields a value for  $b_2$ . The fit allows for a shift of the  $\theta = 0^\circ$  position due to misalignment. The asymmetry parameters  $b_2$  have been measured for various experimental conditions. In particular, we have applied a retarding electric field across the interaction volume, so that because of the spreading of the electron beam different angular ranges  $\Delta \theta$ were accepted by the aperture. This field also allows one to discriminate against photoelectrons from lower atomic states (e.g.,  $7^2S_{1/2}$ ) which might be populated from the  $7^2P$  states. The results for  $b_2$  with their experimental uncertainty, which includes an estimate of the systematic errors, are given in Table I.

The experimental points for the spin polarization  $P_{\perp}(\theta)$  are shown in Fig. 3. The curve is obtained from Eq. (2) with use of the experimental asymmetry parameters. The results for  $d_2^{\perp}$  are given in Table I together with their statistical errors.

The results for the relative phase for the  $7^2P_{1/2}$  state are  $\varphi_s - \varphi_p = 127.4^\circ$  or 232.6° from Eq. (3) and  $\varphi_s - \varphi_p = 319^\circ$  or 221° from Eq. (4). Since we have investigated the systematic errors in some



FIG. 2. Angular distribution of photoelectrons from  $7^2P_{1/2}$  (upper curve, upper ordinate) and from  $7^2P_{3/2}$  (lower curve, lower ordinate).  $N_e$  is the number of photoelectrons collected, normalized to equal light intensity.  $\Delta\theta$  indicates the angular resolution.

TABLE I. Results for the asymmetry parameters  $b_2$  and  $d_2^{\perp}$  and the relative phase  $\varphi_{\rm S} - \varphi_D$ .

|               | ,                            |                    | $\varphi_{\rm S} - \varphi_{\rm D}$ |              |
|---------------|------------------------------|--------------------|-------------------------------------|--------------|
|               | <i>b</i> 2                   | $d_2^{\perp}$      | expt                                | theor        |
| $7^2 P_{1/2}$ | $1.45 \pm 0.10$              | $-0.318 \pm 0.028$ | $232.6^\circ\pm8.0^\circ$           | <b>23</b> 4° |
| $7^2 P_{3/2}$ | $\boldsymbol{1.35 \pm 0.10}$ | $-0.127 \pm 0.030$ | $245.0^\circ \pm 6.0^\circ$         |              |

detail for the  $\sigma(\theta)$  but not for the  $P_{\perp}(\theta)$  measurement, we use the latter only to decide between the two possible values obtained from Eq. (3). The result for  $(\varphi_s - \varphi_p)^{expt}$  is listed in Table I.

The theoretical value for the relative phase has been calculated according to

$$(\varphi_s - \varphi_p)^{\text{theor}} = (\sigma_s - \sigma_p) + (\mu_s - \mu_p)\pi + \pi.$$

 $\sigma_s - \sigma_D$  is the difference of the Coulomb phases of the *s* and *d* partial waves for electrons of 1.5 eV (our photoelectron energy)<sup>16</sup> and  $\mu_s - \mu_D$  is the difference of the quantum defects for *nS* and *nD* states extrapolated into the continuum. The additional  $\pi$  accounts for the different signs of the radial transition elements. For the  $7^2P_{1/2}$ state the theoretical value is  $(\varphi_s - \varphi_D)^{\text{theor}} = 234^{\circ}$ in good agreement with experiment.

As shown in Figs. 2 and 3 we have done the same measurements for the  $7^2P_{3/2}$  state. This case is more complicated in that three transition matrix elements are involved leading to the  $\epsilon^2 S_{1/2}$ ,  $\epsilon^2 D_{3/2}$ , and  $\epsilon^2 D_{5/2}$  continuum states. Presently there is a discrepancy: If we analyze the experimental results using the very good approximation that the transition amplitudes into  $\epsilon^2 D_{3/2}$ .



FIG. 3. Spin polarization  $P_{\perp}(\theta)$ , perpendicular to the reaction plane, of photoelectrons from  $7^2P_{1/2}$  (upper half) and from  $7^2P_{3/2}$  (lower half).

and  $\epsilon^2 D_{5/2}$  continuum states are the same<sup>17</sup> and treat the excitation step in the fine-structure scheme, the result  $\varphi_s - \varphi_p = 245^\circ \pm 6^\circ$  is in agreement with result of the  $7^2 P_{1/2}$  measurement. However, because of the long duration of the laser pulse and the low ionization probability the excitation step should be treated in the hyperfine coupling scheme.<sup>18</sup> Then an equation similar to Eq. (3) is obtained,<sup>8</sup> which for the experimental value for  $b_2$  yields  $\cos(\varphi_s - \varphi_p) = -1.13 \pm 0.12$ . Experiments are in progress to investigate this problem.

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## Spin-Orbit Coupling Effects in Parameters that Describe Electron-Photon Coincidence Experiments

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Parameters are introduced to characterize the electron-photon coincidence rate for atoms where spin-orbit interaction is present in the radiating target state. It is shown that two of these parameters (called  $\Delta$  and  $\epsilon$ ) obey rigorous selection rules which require that they go to  $\pi/2$  at 0° and 180° electron scattering angles in the presence of spin-orbit coupling while their limit in the LS-coupled case is 0. Numerical results are presented.

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In recent years, application of electron-photon coincidence technique<sup>1-6</sup> to the study of inelastic scattering of electrons by atoms has resulted in valuable new information about the collision physics. The angular correlation parameters which are extracted from these measurements provide a stringent test of the theoretical models used to describe electron-impact excitation processes.<sup>7</sup> However, most cases studied to date have been atoms that are well described by an LS-coupling scheme (such as  $He^{1-6}$ ) where two parameters,  $\lambda$  and  $\chi$ , have been used to characterize the coincidence rate. For heavier atoms the introduction of spin-orbit interaction breaks the planar symmetry in the scattering amplitudes valid in the LS-coupling scheme and introduces explicit spin summations through the spin dependence of the scattering amplitudes due to the unpolarized nature of the incident electrons. As a consequence new parameters have to be introduced to describe the coincidence rate. In this Letter we use the approach of Fano and Macek<sup>8</sup> (a generalization of the earlier treatment of Macek and Jaecks<sup>9</sup>) to show that the spin-orbit interaction in the target atom produces effects in these parameters that

can be readily observed experimentally at electron scattering angles  $(\vartheta_e$ , with respect to the incident beam) close to  $0^\circ$  and  $180^\circ$ , in the excitation of an electronic state with angular momentum J = 1 from a J = 0 ground state.

In general the presence of spin-orbit interaction in the excited state of the target prohibits the reduction of the four independent Fano-Macek source parameters to two parameters  $(\lambda, \chi)$ , as is possible in the LS-coupled case.<sup>1-6</sup> We propose a new four-parameter description of the source parameters which makes the spin-orbit effect in the target more transparent.<sup>10</sup> The advantage is that two of these parameters obey rigorous selection rules which require that they go to  $\pi/2$  at  $\vartheta_{e} = 0^{\circ}$  and  $180^{\circ}$  in the presence of spinorbit coupling, while their limit in the LS-coupled case is zero. This behavior has not yet been observed experimentally since the few available measurements for heavier atoms have assumed planar symmetry for the scattering amplitudes. We present below numerical results for excitation of Ar,<sup>11, 12</sup> to illustrate the detailed behavior of these parameters.

A brief derivation of the new parameters is as