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Measurements on photo-ionization of 3s3p $^1P_1$ magnesium atoms

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Abstract. This paper presents experimental studies of resonant two-photon ionization of Mg via the $3s^2\ ^1S_0 \rightarrow 3s3p\ ^1P_1$ transition using 285.2 nm CW light. Dimensionless ratios of the cross section for ionization into a continuum state of S character relative to ionization into a state of D character are extracted from data obtained in two independent experiments. We obtain the values 0.102 ± 0.003 and 0.096 ± 0.012, respectively, and compare these to current theoretical predictions.

1. Introduction

Tunable intense light sources developed over the past decades have dramatically increased the number of experimental investigations of excited-state photo-ionization processes. Experimental investigations, however, have mainly concentrated on alkaline atoms. In these quasi-one-electron atoms electron correlations are relatively unimportant and structures in the photo-ionization process are dominated by the magnitude of the dipole matrix element. Alkaline earth atoms, representing two-electron systems, are quite different in that respect. Here, electron correlations in ground and excited states play an important role in the photo-ionization process [1]. These features have been subject to recent theoretical [2–7] and experimental [8–13] studies. Investigations of the alkaline earth atoms have so far primarily focused on magnesium, calcium and barium atoms.

This paper addresses experimental studies based on CW resonant two-photon ionization of neutral magnesium through the transition $3s^2\ ^1S_0 \rightarrow 3s3p\ ^1P_1$ at 285 nm. We use the angular momentum of the light to obtain dimensionless relative cross section ratios which can put theoretical models to a sensitive state-selective test. The paper is based on results obtained in a collimated atomic Mg beam (in Copenhagen) and by capturing the produced $^{24}Mg^+$ ions in a linear Paul trap (in Aarhus).

Besides the fundamental aspects, resonant photo-ionization of alkaline earth elements has proven useful for pure and efficient loading of a Paul trap [14]. Furthermore, in laser cooling and trapping of magnesium atoms, using the $3s^2\ ^1S_0 \leftrightarrow 3s3p\ ^1P_1$ transition, the two-photon ionization is a severe loss channel [15]. Detailed understanding of the ionization process may help in increasing the number of trapped atoms in a magnesium MOT.

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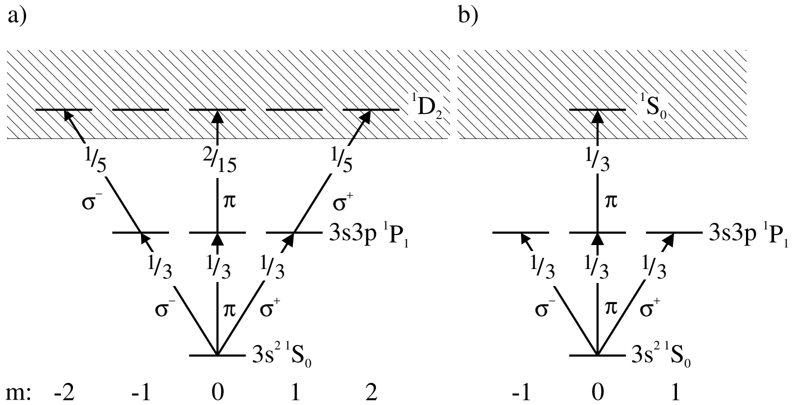

Figure 1. Possible channels from the $3s^2\ ^1S_0$ level to a continuum state of (*a*) $^1D_2$ and (*b*) $^1S_0$ character when light of pure circular ($\sigma^{\pm}$) or linear (π) polarization is applied. The numbers attached to the arrows are the square of the Wigner 3-j symbols for the transitions.

2. Theory of measurement

Magnesium has three stable isotopes: ^{24}Mg (79%), ^{25}Mg (10%) and ^{26}Mg (11%). ^{24}Mg seems to be the best candidate due to high abundance and absence of hyperfine structure, simplifying the theory of measurement.

The relevant terms for our photo-ionization studies of ^{24}Mg are presented in figure 1. The ionization energy of the Mg ground state $3s^2\ ^1S_0$ is 7.65 eV, i.e. by two-photon ionization ($\lambda = 285$ nm) we populate S and D states 1 eV into the continuum. In the experiments only one laser polarization is present at a time, so possible ionization paths reduce to those indicated by arrows. Due to the low power of the CW lasers involved in the experiments, the photo-ionization process can effectively be considered as a two-step process, the first step being the population of an intermediate $3s3p\ ^1P_1$ sublevel (all with numerically equal Wigner 3-j values) and the second the ionization via an electric dipole process. The ionization probability is assumed much smaller than the excitation probability so only the second step needs to be considered in order to obtain relative cross sections.

Letting J denote the total angular momentum, M the projection of J along the z axis and γ additional quantum numbers, the cross section of the photo-ionizing transition $|\gamma JM\rangle \rightarrow |\gamma' J' M'\rangle$ is, in general, given by [13, 16, 17]

$$\sigma_{\gamma' J' M', \gamma JM} = 4\pi^2 \alpha a_0^2 \hbar\omega \left| \hat{\epsilon} \cdot \left\langle \gamma' J' M' \middle| \sum_j \boldsymbol{r}_j \middle| \gamma JM \right\rangle \right|^2 \qquad (1)$$

where ω is the laser angular frequency, $\hat{\epsilon}$ is the laser polarization, α is the fine structure constant, a_0 is the Bohr radius and $\boldsymbol{r}_j$ is the position of the jth electron. Equation (1) can be rewritten in terms of first rank spherical tensors, $\hat{\epsilon}\cdot\boldsymbol{r}_j = \sum_q \epsilon_q^{\star} r_{jq}^{(1)}$, where ϵ_0, ϵ_{-1} and ϵ_1 refer to linear, left- and right-hand circular polarization components, respectively. Applying the Wigner–Eckart theorem

$$\left\langle \gamma' J' M' \middle| \sum_j \boldsymbol{r}_{j,q} \middle| \gamma JM \right\rangle = (-1)^{J'-M'} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix} \left\langle \gamma' J' \middle\| \sum_j r_j^{(1)} \middle\| \gamma J \right\rangle \qquad (2)$$

and summing over all possible final states $|\gamma' J' M'\rangle$, the total cross section for photo-ionization from a specific initial state $|\gamma JM\rangle$ is

$$\sigma_{\gamma JM} = 3(2J+1) \sum_{\gamma',J',M',q} \begin{pmatrix} J' & 1 & J \\ -M' & q & M \end{pmatrix}^2 \epsilon_q^2 \sigma_{\gamma' J',\gamma J} \tag{3}$$

where

$$\sigma_{\gamma' J',\gamma J} = \frac{4\pi^2 \alpha a_0^2 \hbar\omega}{3(2J+1)} \left| \left\langle \gamma' J' \left\| \sum_j r_j^{(1)} \right\| \gamma J \right\rangle \right|^2 \tag{4}$$

is the partial isotropic cross section, i.e. the cross section for transition to a specific J' state when all initial magnetic substates M are equally populated. Only the transitions sketched in figure 1 are possible. In the experiments we measure count rates for linear and right-hand circular polarization, $N_\parallel$ and N_+, that relate to the corresponding cross sections as

$$N_\parallel = A\sigma_{\gamma,1,0} \qquad N_+ = A\sigma_{\gamma,1,1} \tag{5}$$

where the count rate dependence on atom density, light intensity, detection efficiency and other experimental parameters is included in A. We assume that A in the two expressions are equal. Using equation (3) relations between the partial isotropic cross sections for $J' = 0$ and 2, represented by σ_S and σ_D, respectively, and the measured count rates are obtained. Linear polarization ($M = 0$ and $q = 0$) leads us to

$$N_\parallel = A\left(9\begin{pmatrix} 0 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 \sigma_S + 9\begin{pmatrix} 2 & 1 & 1 \\ 0 & 0 & 0 \end{pmatrix}^2 \sigma_D\right) = A(3\sigma_S + \tfrac{6}{5}\sigma_D) \tag{6}$$

whereas the case of right-hand circular polarization ($M = 1$ and $q = 1$) gives

$$N_+ = A\left(9\begin{pmatrix} 2 & 1 & 1 \\ -2 & 1 & 1 \end{pmatrix}^2 \sigma_D\right) = A\tfrac{9}{5}\sigma_D. \tag{7}$$

From equations (6) and (7) the ratio of σ_S to σ_D is expressed in terms of the ratio between the two measured ion yields:

$$\frac{\sigma_S}{\sigma_D} = \frac{3}{5}\frac{N_\parallel}{N_+} - \frac{2}{5}. \tag{8}$$

The ratio $\frac{\sigma_S}{\sigma_D}$ can be compared with theoretical model calculations.

3. Experimental setup

This section presents the experimental setup for measuring polarization-dependent photo-ionization rates. Two different experimental methods were employed: direct ion detection in a well collimated magnesium beam and capturing of the produced $^{24}Mg^+$ ions in a Paul trap, followed by CCD camera detection.

3.1. Magnesium beam setup

The beam experiment [15] is sketched in figure 2. A laser beam intersects a thermal magnesium beam at right angles and a channeltron detects the ion production. The atomic beam is generated by a 712 K oven with a 1 mm nozzle, yielding an output of about 10^{15} sr^{-1} s^{-1} atoms with a mean beam velocity of 930 m s^{-1}. At the intersection zone the beam diameter is 4 mm, corresponding to a Doppler width of 40 MHz, and the density is 5×10^4 atoms mm^{-3}.

The optical system consists of an argon ion laser (Coherent Innova 90-5) pumping a single-mode ring dye laser (Coherent 899-21) having a linewidth less than 500 kHz. The output at

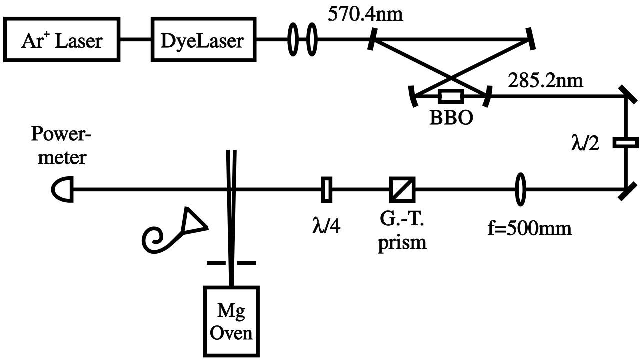

Figure 2. Schematic setup for the beam experiments with a well collimated magnesium beam and a channeltron for direct ion detection.

570.4 nm is frequency doubled in a ring cavity with a BBO crystal to obtain 285.2 nm. With an input power of 560 mW a typical output power is 30 mW. The laser frequency is tuned resonant to the 285.2 nm $3s^2\ ^1S_0 \rightarrow 3s3p\ ^1P_1$ transition in ^{24}Mg. A pinhole cleans the UV beam and a 500 mm lens focuses the beam to a diameter of 140 μm at the intersection zone. Before entering the chamber the beam passes through a Glan–Thompson prism to ensure linear polarization of high quality and a quarter-wave plate which is rotated to alternate between linear and circular polarization. The beam intensity is unaffected by the position of the wave plate. For each of the polarizations the degree of polarization after passage of the optical windows has been measured to be 99.9%. A channeltron is positioned 60 mm from the intersection zone in the plane of the two beams at an angle of 45° to each of them, see figure 2. For efficient ion collection a small grid biased by typically −25 V is placed in front of the channeltron. Finally a powermeter monitors the UV laser intensity during the experiment. Power fluctuations are below 5%.

3.2. Paul trap setup

Figure 3 shows a schematic drawing of the Paul trap experimental setup (the setup is almost identical to the one described in [14]). The laser light used for ionization of Mg is produced by frequency doubling the output of a CW tunable dye laser system in a BBO crystal. First, this light is linearly polarized by a polarization beam splitter (PBS). Subsequently, any polarization state can be prepared by means of a half-wave plate ($\lambda/2$) and a quarter-wave plate ($\lambda/4$). The ionizing laser intersects a Mg atomic beam in the centre of a linear Paul trap (the atomic beam effuses from an oven and is collimated by skimmers to a full divergence angle of 30 mrad). The laser beam and the atomic beam propagate in orthogonal directions to avoid first-order Doppler broadening of the involved transitions. Due to isotope shifts, the various isotopes in the atomic beam can be selectively ionized. In the experiment the laser frequency is tuned to be resonant with the $3s^2\ ^1S_0 \leftrightarrow 3s3p\ ^1P_1$ transition of ^{24}Mg. The ionization proceeds as described in section 2 and the ions produced are captured in the linear Paul trap. After passage through the trap, the power of the ionizing laser is monitored using a photodiode (PD). During trapping, the ions are laser cooled by driving the closed 280 nm transition $3s\ ^2S_{1/2} \leftrightarrow 3p\ ^2P_{3/2}$ of $^{24}Mg^+$ with a laser system almost identical to the one used for ionization. To avoid any contribution from the cooling laser in the final ionization step ($3s3p\ ^1P_1$ state → continuum), the two laser beams are never present at the same time. This is achieved by means of a mechanical chopper alternately blocking one beam and letting the other through at a frequency

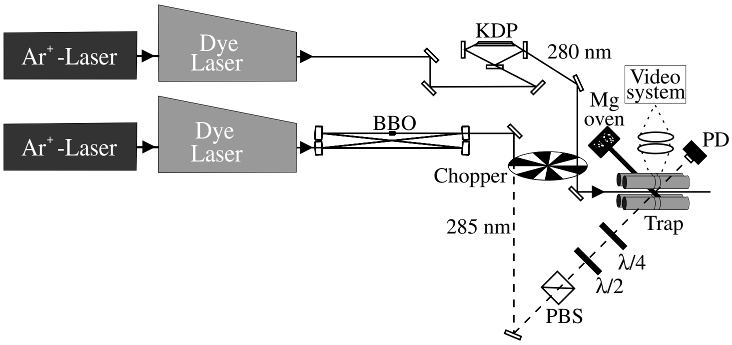

Figure 3. Schematic setup for the experiments using a linear Paul trap for capturing the ions produced by photo-ionization.

of 600 Hz. In the experiments discussed here ions are captured in the trap during a period of 20 s. Due to laser cooling the trapped ions form fluorescing Coulomb crystals [18] which are imaged and recorded with an image intensified digital camera system. After each image acquisition the trap is emptied by briefly switching off the trap potential. The polarization of the ionization laser light is altered and the experimental procedure is repeated. To improve statistics an extended series of measurements alternating between two different polarizations is performed.

4. Results and discussion

All photo-ionization experiments described below are carried out using the ^{24}Mg isotope but measurements with the two other isotopes have also been investigated. In the beam experiment the magnesium photo-ionization signal has been measured as a function of UV laser frequency. Figure 4 shows the data obtained. The three isotopes of Mg are clearly observed in the correct ratio. The FWHM of the ^{25}Mg peak is slightly larger compared with the other peaks because of hyperfine splitting induced by the nuclear spin. We could model the observed spectra by assuming a two-step model. The first step is excitation from $3s^2\ ^1S_0$ to $3s3p\ ^1P_1$ with a probability given by the steady state solution of the optical Bloch equations. The second step, from the $3s3p\ ^1P_1$ state into the continuum, we assumed proportional to the laser intensity. Integrating the ionization probability over the interaction volume, taking into account the spatial laser beam profile, yielded the fit in figure 4. No free parameters were used except the height of the ^{24}Mg peak. Such a simple model, we believe, is applicable since the ionization probability is much smaller than the first-step excitation probability.

In the atomic beam polarization measurements the ion production is counted for 30 s with the ionizing laser beam being circularly polarized. Then the polarization is changed to linear and the ion production is counted for 30 s. The relatively short acquisition time reduces effects from thermal drift. Counting statistics is improved by repeating the measurement several times. A series of measurements were performed at laser intensities of 40, 70 and 120 times the saturation intensity of 0.446 W cm^{-2} with average count rates of 2.2×10^4, 4.2×10^4 and 7.1×10^4 count s^{-1}, respectively. No power dependence was observed: the three series yielded the σ_S to σ_D ratios 0.101 ± 0.002, 0.101 ± 0.005 and 0.104 ± 0.002. The averaged result is stated in table 1.

The raw data from the Paul trap experiments consist of CCD images of ion crystals [18]. Figure 5 shows the image of a typical Coulomb crystal for the parameters used in the

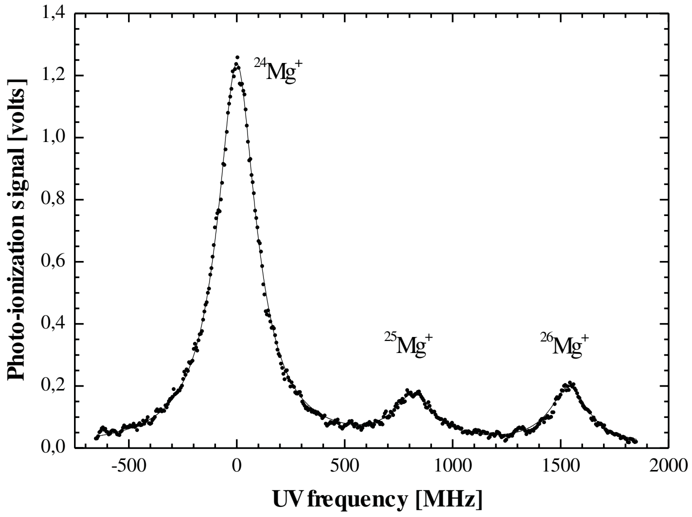

Figure 4. A typical spectrum of photo-ionization as a function of UV frequency showing the three stable isotopes and their relative abundances. The data were obtained with linear polarization. A fit based on a theoretical model is superimposed on the experimental curve.

Table 1. Experimental values from this paper and theoretical values from current calculations. The three values from the beam experiment are obtained at intensities 20, 35 and 60 W cm^{-2}. All experimental error bars are statistical.

	Beam experiment	Trap experiment	Theory
$\frac{N_+}{N_\parallel}$	1.196 ± 0.007	1.21 ± 0.03	
$\frac{\sigma_S}{\sigma_D}$	0.102 ± 0.003	0.096 ± 0.012	0.15^a, 0.13^b

[a] [4].
[b] The value is obtained by inspection of figure 1 in [5].

experiment. The Coulomb crystals formed in the linear Paul trap have an ellipsoidal shape with a uniform density distribution [19]. Hence, $N_\parallel/N_+$ in equation (8) can be determined by measuring the volumes of the crystals formed for linear and for right-hand circular polarizations of the ionizing laser light. From a series of measurements, where the polarization is alternated between linear and right-hand circular, we obtain the values stated in table 1.

The frequency shift of the $3s^2\ ^1S_0 \leftrightarrow 3s3p\ ^1P_1$ transition due to the Zeeman effect related to non-compensated B fields does not exceed 1 MHz and is negligible compared with the natural linewidth of $\Gamma(^1P_1) = 80$ MHz. As a consistency check the effects of alternating between horizontally and vertically polarized light and between circularly polarized light of opposite helicities have been investigated. The ratios of ion yields were found to be $N_\perp/N_\parallel = 1.01 \pm 0.11$ and $N_+/N_- = 0.99 \pm 0.06$, respectively. As expected, due to symmetry no effect is detected. The main uncertainty in the measurement of $N_+/N_\parallel$ is due to fluctuations in laser power ($\pm 4\%$) and the atomic beam flux as well as the uncertainty in the crystal size determination. As a consequence of the relatively small contribution from the $^1S_0$ channel, the deduced ratio σ_S/σ_D in equation (8) is very sensitive to uncertainty in the measured ratio $N_+/N_\parallel$. The experiment could be improved by actively stabilizing the laser power and by increasing the loading time.

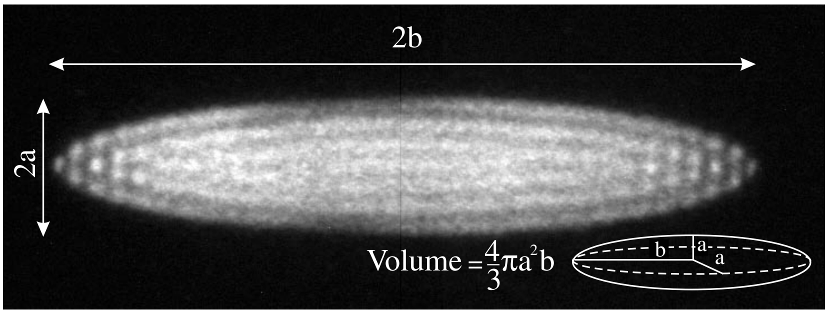

Figure 5. A typical $^{24}Mg^+$ Coulomb crystal produced by photo-ionization. The crystal has an ellipsoidal shape and a volume given by the lengths of the minor and major axes, $2a$ and $2b$, respectively. For the crystal shown here, $2b = 507\ \mu$m.

In table 1 we present a summary of the experimental results and compare them with current theoretical calculations [4, 5]. The two experimental values agree very well. The theoretical predictions are in reasonable agreement with measured values. However, we notice no agreement within the experimental error bars. Both theories use a finite L^2 basis set constructed from a set of one-particle frozen core Hartree–Fock orbitals. In the theoretical work [5] the authors extend the configuration-interaction (CI) model to include the resonant case by using a CI method for the continuum spectrum (CIC). The value obtained is somewhat lower compared with the value calculated by [4]. In a recent paper [6], the theory in [4] is refined beyond the frozen core approximation. The results, however, were found to be similar to those obtained in [4]. Further investigation of the influence of CIC and the basis set on the photo-ionization results might provide useful information on the discrepancy between theory and experiment.

5. Summary

This paper presents measurements on resonant CW two-photon ionization via the 285.2 nm transition $3s^2\ ^1S_0 \rightarrow 3s3p\ ^1P_1$ using two different experimental methods: direct ion detection in a well collimated magnesium beam and measurements based on counting the $^{24}Mg^+$ ions produced in a Paul trap. The polarization of the UV light is used to extract dimensionless cross section ratios. In the two experiments we obtain averaged S to D cross section ratios of 0.102 ± 0.003 and 0.096 ± 0.012, compared with theoretical predictions of 0.15 [4] and 0.13 [5].

Acknowledgments

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