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LETTER TO THE EDITOR

Measurement of photoionisation cross sections for the $7^2P_{3/2}$ and $6^2D_{3/2}$ excited states of caesium

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Abstract. Relative photoionisation cross sections for the $7^2P_{3/2}$ and $6^2D_{3/2}$ excited states of caesium have been measured in the visible spectral range of the ionising photon. The measurements have been performed using two pulsed dye lasers, pumped by the same excimer laser, for population of the excited states and for their photoionisation. The results for the $7^2P_{3/2}$ state show the theoretically expected smooth variation with wavelength, while the deep minimum predicted by theory for the $6^2D_{3/2}$ state has not been detected experimentally.

By the application of modern laser technology, photoionisation investigations have been extended to short-lived excited atomic states (Kaminski *et al* 1979, 1980, Hansen *et al* 1980, Grannemann *et al* 1977).

For excited states, structures in addition to the well known Cooper minima, which appear for ground-state atoms, may be expected in the photoionisation cross sections as a function of the energy of the ionising photon. Particularly striking minima have been predicted for the excited nD states of caesium, caused by a shape resonance in the εf continuum partial wave (Msezane and Manson 1975, 1982, Lahiri 1981). For the nP states of Cs on the other hand a smooth behaviour of the photoionisation cross section is predicted by all theories, e.g. Norcross and Stone (1966).

In our experiments we have measured for the first time the photoionisation cross sections over the visible spectral range of the ionising photon for both the $7^2P_{3/2}$ and the $6^2D_{3/2}$ excited states. Thus, a comparison can be made between the experimental results obtained for two states for which very different behaviours are expected theoretically.

The experiments have been performed using two pulsed dye lasers pumped by a Lambda Physics excimer laser. The wavelength of the first dye laser, laser 1, is tuned to resonance to excite either the $7^2P_{3/2}$ (455 nm) or the $6^2D_{3/2}$ (442 nm) state of atomic Cs. The wavelength of the second, the ionising dye laser, laser 2, is varied in the experiments. The laser energy ranged from ten to a few hundred microjoules.

A schematic diagram of the experimental set-up is shown in figure 1. Indicated are the exciting laser beam λ_1 and the ionising laser beam λ_2 which is delayed optically by about 4 ns to avoid errors due to different pulse shapes. The wedge-shaped beam splitter BS combined the two beams and directed the collinear beams to the diaphragm D, which limited the diameter of the beams. The diaphragm D was imaged by means of a telescope with a pinhole TP to the atomic Cs beam. After having

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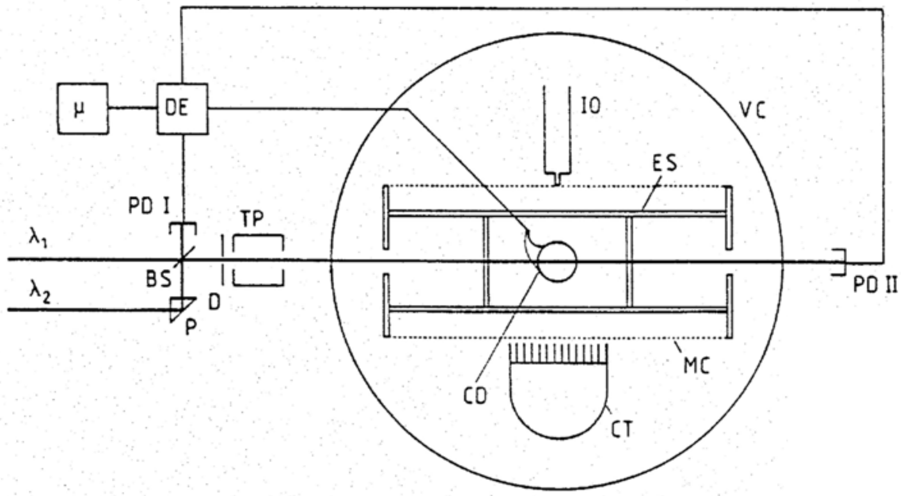

Figure 1. This figure shows the experimental set-up: exciting laser beam, λ_1; ionising laser beam, λ_2; prism, P; beam splitter, BS; diaphragm, D; telescope with pinhole, TP; vacuum chamber, VC; molybdenum mesh cylinder, MC; electrode system, ES; inner oven, IO; channeltron detector, CD; cooling trap, CT; photodiodes, PD I, PD II; detection electronics, DE; microcomputer, μ.

passed the interaction volume the beams hit the photodiode PD II, which allows the determination of the energy of the laser pulses. Its spectral sensitivity was determined against a calibrated thermopile.

The atomic beam is generated in a two-stage oven with a collimating nozzle. The inner-oven part IO was held at a high temperature of about 550 K to keep the Cs molecular contribution very small (Grannemann and van der Wiel 1975). The caesium reservoir was held between 370 and 410 K so that the density of Cs atoms in the interaction volume, which had been determined previously using a hot tantalum wire detector (Marton 1967), ranged from 10^8–10^{10} atoms/cm^3 depending on the particular measurements. After passing the interaction volume, the Cs beam hit a liquid-nitrogen cooling trap CT which reduced the background pressure to less than 2×10^{-6} mb.

Any static magnetic fields in the interaction volume have been compensated to below 8 A m^{-1} using Helmholtz coils in all three spatial directions. The interaction volume was also shielded against disturbing electric fields by a cylinder of molybdenum mesh MC. All photoelectrons generated in the interaction volume were accelerated by a special electrode system ES towards a channeltron detector CD in single pulse counting mode. The channeltron pulses had to be restricted to a maximum rate of 0.3 counts/laser pulse to avoid pile-up. A secondary beam from the beam splitter BS is detected by a photodiode PD I which is used to generate a trigger pulse for the electronic gates. The data were registered by an AIM 65 microcomputer μ which controls the experiment.

For the measurement of the photoionisation cross section, the following procedure was applied. The electron count rate and the laser energy were determined at each wavelength of the ionising light for laser 1 only, laser 2 only and for both lasers together. The background counts obtained when using the lasers separately were subtracted from the count rate obtained for both lasers together. Thus the photoelectrons created by resonant two-photon ionisation of caesium atoms (main contribution of laser 1) and possibly of residual molecules, as well as photoelectrons created by stray light at the caesium coated walls (main contribution of laser 2) could be eliminated. Also photoelectrons from molecules, excited by laser 1 and ionised by laser 2, could be excluded because a slight detuning of laser 1 off-resonance caused the net count rate to disappear (Grannemann *et al* 1976a, b). The background totalled between 10–20% of the signal for the $7^2P_{3/2}$ state and up to 60% in the case of the $6^2D_{3/2}$ state which required higher laser intensities, because of the quadrupole transition of the exciting step.

The net number of photoelectrons N_e created during a laser pulse in the interaction volume is

$$N_e = \sigma_{PI}(\lambda) \int_0^T j_2(t) n_a(t) \, dt$$

where $j_2(t)$ is the photon number per unit area and time, $n_a(t)$ is the number of excited atoms in the interaction volume and T is the laser pulse duration.

The ratio N_e/N_γ, where the relative photon number is

$$N_\gamma = \int_0^T j_2(t) \, dt$$

is a measure of the photoionisation cross section σ_{PI} as long as $n_a(t)$ is the same for all laser pulses and the pulse shape is stable enough.

In order to cover a wide spectral range of the ionising light, a number of dyes were used for laser 2. For each dye, data were taken in six groups of 90 laser pulses each at three to six different wavelengths within the most efficient range. The electron counts N_e and the laser energy integrated over the 90 pulses were stored by the microcomputer μ. The relative photon number N_γ was determined using the energy of the photons and the spectral sensitivity of the diode. The measurements of N_e/N_γ were repeated four to five times to check for small drifts of the experimental set-up.

The number of excited atoms available for ionisation was kept constant by controlling the temperature of the Cs oven to ensure a constant density of the ground-state atoms and by checking the background from laser 1 which is mainly due to photoionisation of the excited atoms by the resonant light.

With this set-up the results for the $7^2P_{3/2}$ excited state shown in figure 2 have been obtained. The experimental values have been normalised to the result of an earlier absolute cross section measurement at 455.5 nm (Heinzmann *et al* 1977). Also shown in figure 2 are the theoretical results for the photoionisation cross section computed using quantum defect theory (Burgess and Seaton 1960) shown as full circles, and using the modification by Norcross and Stone (1966) indicated by the two crosses. They show reasonable agreement with the experimental points, in particular concerning the smooth increase with wavelength.

For the $6^2D_{3/2}$ excited state a very similar experimental curve is obtained which is shown in figure 3. The experimental results, which are normalised to the theory at 588 nm, are in contrast to the theoretical calculations by Lahiri (1981) shown as dots and by Manson (1981) shown as crosses. From the comparison we conclude that the position of the minimum is not predicted correctly by theory, since we feel that no systematic error should be able to mask completely a minimum of that depth.

Sources of possible systematic errors are discussed below. One experimental problem is that different dyes require readjustments to the experimental set-up. However, any effect due to such adjustments would also show up in the $7^2P_{3/2}$ measurements. As can be seen from figure 2, the results obtained here for the cross sections with different dyes (Coumarin 47, 102 and 307) at the same wavelengths 485

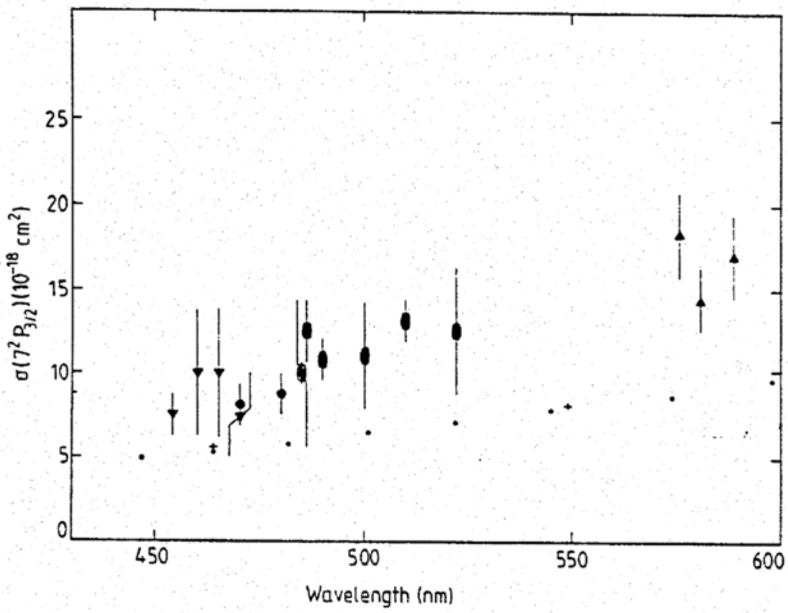

Figure 2. Photoionisation cross sections for the $7^2P_{3/2}$ state of atomic caesium. The dyes used for the ionising light are: Coumarin 47, ▼; 102, ●; 307, ⬮; and Rhodamine 6G, ▲. Theoretical results represented by dots and crosses.

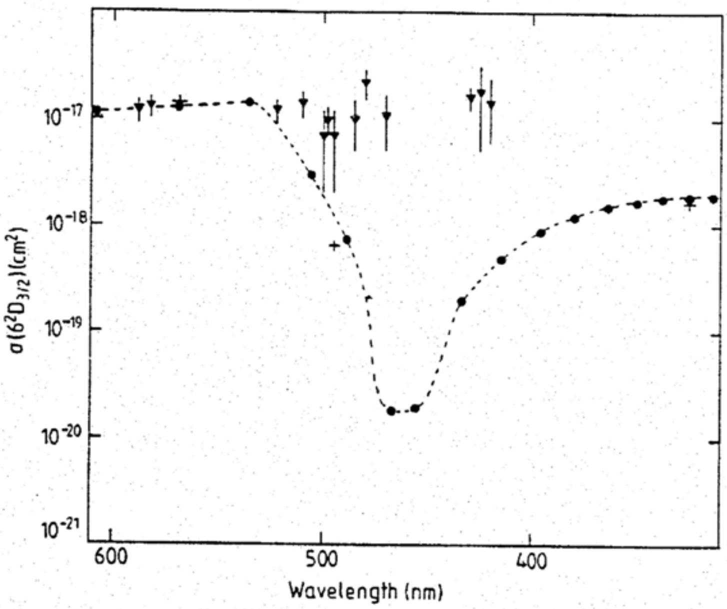

Figure 3. Photoionisation cross sections for the $6^2D_{3/2}$ state of atomic caesium: experimental, ▼, and theoretical, ● and +, results.

and 470 nm are in good agreement. Similarly the results for the $6^2D_{3/2}$ measurements, although without overlap, show a smooth continuation.

A further possible complication arises from the decay of the 6D state into the lower states, the 6P and 7P states in particular. Superfluorescent effects can be neglected because of the low excited-state density (Vrehen 1979, 1981). The spontaneous transitions should also produce only small effects, since no large population of the lower states can be built up within the duration of the laser pulse, which is small (10–15 ns) compared with the lifetime of the excited state, $\tau(6^2D_{3/2}) = 60 \pm 2.5$ ns (Marek 1977), while the cross sections of the three states are of the same order. Also the contributions of 7P and 6P states should increase with the wavelength, but we have a smooth decrease with wavelength.

We are led to the conclusion that a minimum in the visible spectral region of such amplitude and width as described by theory is not confirmed by the measurements. The gentle increase of the cross section with energy indicates that the minimum, if it exists, should be sought at lower wavelengths.

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