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LETTER TO THE EDITOR

Polarization effects in resonant two-photon ionization of caesium

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Abstract. An experiment is reported on two-step photoionization of Cs via the $6P_{3/2}$ intermediate state using circularly polarized radiation from a GaAs laser for the first step (8521 Å) and from an Ar-ion laser for the second step. We present values for the ratio of ion production rates for right- and left-handed circular polarization in the second step and for the spin polarization of the photoelectrons. The measurements are interpreted on the basis of a calculation which takes into account the hyperfine splitting of the levels involved in the first transition. A description in terms of fine-structure splitting of the 6P state is shown to be totally inadequate, even when allowing for possibly large spin–orbit effects in the continuum. The process is evaluated as a possible source of a spin-polarized electron beam.

Resonant two- and three-photon ionization of alkali atoms has recently been the subject of numerous investigations. Reports by Zeman (1974), Lambropoulos and Lambropoulos (1974), Weisheit (1972) and Nygaard *et al* (1975) are of immediate relevance for the present work. Measurements with circularly polarized light on the spin polarization of photoelectrons ejected in a two-step process in Cs and in a three-step process in Na were reported by Zeman (1974) and by Lambropoulos *et al* (1973), respectively. In both cases the measured polarizations differed markedly from those predicted using a fine-structure scheme with neglect of spin–orbit coupling in the continuum (Lambropoulos 1973). However, in both experiments low-repetition-rate dye lasers of very high intensity were used, which left open the possibility that saturation or optical pumping occurred or that a shift and broadening of the resonance level was involved. Moreover, a possible effect of spin–orbit coupling in the continuum can be invoked in order to reconcile experiment and theory (Lambropoulos and Lambropoulos 1974).

However, in both Cs and Na the hyperfine structure of the intermediate state, 7P and 3P respectively, is fully resolved into its F components. This implies that it is incorrect to ignore the hyperfine structure as long as the radiation of the second step is of sufficiently low intensity to consider the event as a two-step process. Very high intensities could lead to broadening of the intermediate levels and breakdown of the hyperfine-splitting scheme.

Since these processes have been proposed as efficient sources of spin-polarized electron beams (Lambropoulos *et al* 1973), it is worthwhile reinvestigating this prob-

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lem of choice of splitting scheme under conditions which permit the disturbing effects mentioned above to be ruled out. An additional feature of interest is that the spin polarization depends sensitively on the relative phase and magnitude of the various reduced matrix elements involved in the ionizing step, thus providing information on spin–orbit coupling in the continuum.

The process we have studied is two-step ionization of Cs via the $6P_{3/2}$ intermediate state using circularly polarized light at two different wavelengths for the two steps. We determined the ratio of ion production rates when using right-hand polarized light in the first step and right- or left-handed polarization in the second step. In addition, we report measurements of the spin polarization of the photoelectrons under the same two conditions of light polarization. Each of the three quantities measured

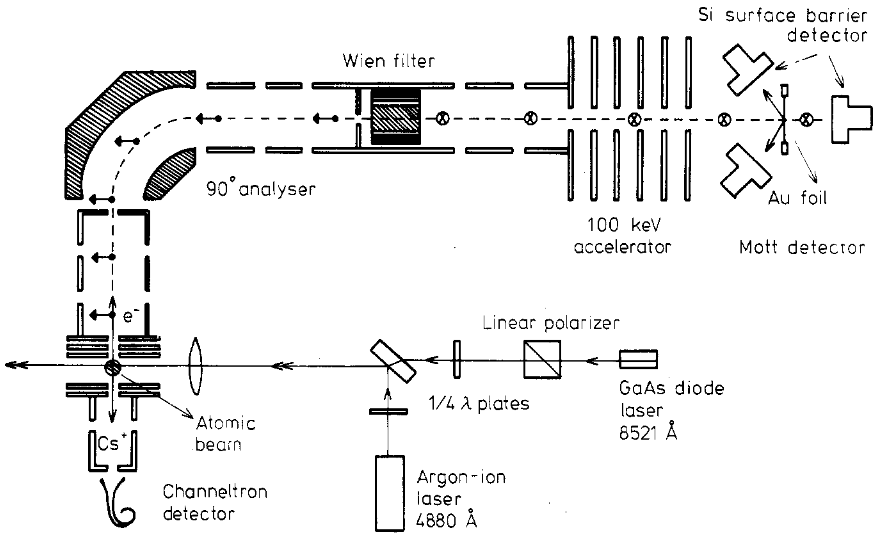

Figure 1. Experimental arrangement for measurements of ion count rate and electron spin polarization in the two-step photoionization of alkali atoms.

is a function of the relative magnitudes (two ratios) of the three matrix elements involved in the ionizing step: $6P_{3/2} \rightarrow \epsilon S_{1/2}$, $\epsilon D_{3/2}$ and $\epsilon D_{5/2}$. Therefore, a determination of one of the three observed quantities is not sufficient to decide between the fine- and hyperfine-structure scheme, if one allows the ratio $(6P_{3/2} \rightarrow \epsilon D_{5/2})/(6P_{3/2} \rightarrow \epsilon D_{3/2})$ to deviate from unity due to spin–orbit coupling in the continuum. The set of three measured values, however, overdetermines the two ratios and thus provides an internal consistency check for either of the two schemes of calculation.

The experimental arrangement shown in figure 1 will be discussed in detail in a forthcoming publication (Granneman *et al*). Here it suffices to mention that an atomic beam of Cs is crossed at right angles by two coincident light beams from two lasers. Photoions and photoelectrons are extracted in a direction perpendicular to the plane of the atomic and light beams. The ions are detected by a channeltron multiplier, while the electrons pass through a 90° electrostatic analyser and a Wien filter for 90° spin rotation and are detected in a Mott analyser.

Two lasers are used in this experiment. Excitation to the $6P_{3/2}$ level (8521 Å) is provided by an IBM developmental-type GaAs laser pulsed with 0·4–0·6 A, 1–2 μs pulses at a repetition rate of approximately 20 kHz. Coarse tuning of the wavelength is obtained by cooling the device to a temperature of about 100 K with an accuracy of 0·01 K. During the current pulse the laser medium heats up such that the wavelength scans rapidly across the two groups of hyperfine transitions from the ground state to the $6P_{3/2}$ state. The scan rate is a few tenths of a nanosecond per individual transition. The absorption can be observed as a corresponding attenuation of the light through the atomic beam. The IR light beam optics is comprised of a microscope objective for forming a low-divergence beam, a linear and circular polarizer and a variable attenuator. Significant attenuation of the IR beam is necessary for most measurements in order to remain in the intensity region of linear absorption and avoid the problem of saturation of the first transition. Moreover, optical pumping is not a problem since the duration of the excitation is very much shorter than the natural lifetime of the $6P_{3/2}$ state (35 ns).

The ionizing radiation (4880 Å) is produced by an Ar-ion laser (Spectra Physics 164 with cavity dumper) which is pulsed at the same frequency as the GaAs laser; the pulses are sufficiently long to overlap with the two absorption peaks of the $6S \rightarrow 6P$ hyperfine transitions. The blue light is properly polarized and mixed with the IR beam by reflection from an IR transparent mirror.

The results of the measurements are three values which represent averages over at least ten independent determinations under a variety of conditions. The ion 'asymmetry' $A_i = N_{rr}/N_{rl}$, i.e. the ratio of the ion count rates when using right- or left-handed circular polarization in the second step (the first step is right-hand polarized) amounts to $1{\cdot}338 \pm 0{\cdot}008$. The photoelectron spin polarization P_{rr} for the case of twice right-handed circularly polarized light equals $0{\cdot}44 \pm 0{\cdot}04$; the value of P_{rl} for the right–left combination amounts to $0{\cdot}085 \pm 0{\cdot}02$. The errors quoted include statistics and the uncertainty in the calibration of the Mott detector.

Our value of 0·44 for the spin polarization is in reasonable agreement with the less accurate results of comparable measurements on Cs (two-step ionization via the 7P state by Zeman (1974)) and on Na (three-step ionization via the 3P and 4D states by Lambropoulos *et al* (1973)). It has already been pointed out that some ambiguity remains in this earlier work due to the high light intensity; for instance, in the work of Lambropoulos *et al* (1973) the spin polarization was found to decrease with increasing light power and we suggest that this was at least partly due to saturation of the resonance transition and consequent reduction of the orientation of the intermediate state.

As regards the theory, one can go through the angular momentum algebra for the first transition in the fine-structure or hyperfine-structure scheme and obtain two different sets of populations of the $|J, M_j\rangle$ levels of the $6P_{3/2}$ state. After inclusion of the second step to the continuum, one arrives at expressions for the ion asymmetry and the spin polarization in terms of two quantities x and y, which are defined as follows:

$$x = \frac{R(\epsilon D_{5/2} \leftarrow 6P_{3/2})}{R(\epsilon D_{3/2} \leftarrow 6P_{3/2})}$$

and

$$y = \frac{R(\epsilon S_{1/2} \leftarrow 6P_{3/2})}{R(\epsilon D_{3/2} \leftarrow 6P_{3/2})}$$

where R is a radial overlap of excited and continuum states and the indices refer to the M_j quantum number. We have assumed that the asymptotic phase difference between the two D states is negligible. Since the fine-structure scheme has been used extensively in the recent literature (Lambropoulos and Lambropoulos 1974 and references therein), we have derived expressions for the ion asymmetry and the two polarizations in that scheme and also in the hyperfine scheme. Since A_i is the most accurate of the measured values, we have first used A_i to obtain a relationship between x and y. Using that relation, we plotted P_{rr} and P_{rl} as a function of x. Figure 2 shows two such plots for the fine and hyperfine schemes. From the figure we

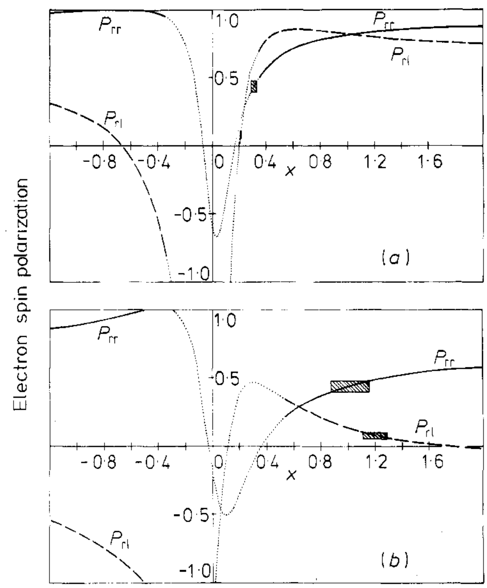

Figure 2. Electron spin polarization for the two-step process $6S \rightarrow 6P_{3/2} \rightarrow$ continuum in Cs as a function of x, i.e. the ratio of the $6P_{3/2} \rightarrow \epsilon D_{5/2}$ and $6P_{3/2} \rightarrow \epsilon D_{3/2}$ bound–free matrix elements. P_{rr} refers to right-hand circularly polarized light in both steps, P_{rl} to left-handed polarization in the second step. Calculations for (*a*) the fine-structure scheme; (*b*) the hyperfine-structure scheme. The calculation is based on the observed ion count rate 'asymmetry' (see text), which contains x and also y, i.e. the ratio of $6P_{3/2} \rightarrow \epsilon S_{1/2}$ and $6P_{3/2} \rightarrow \epsilon D_{3/2}$ matrix elements. The observed spin polarizations with their corresponding uncertainties are shown as shaded areas. The hyperfine scheme is compatible with the experiment and results in $0{\cdot}9 < x < 1{\cdot}3$ and $0{\cdot}45 < |y| < 0{\cdot}73$. The broken parts of the curves refer to non-physical regions of x, i.e. for which $y^2 < 0$.

can decide whether the two measured polarizations occur at a coincident value of x. The broken parts of the curves represent non-physical regions of x, i.e. for which $y^2 < 0$.

The conclusion from the experiment is obvious: the fine-structure calculation is totally inadequate, whereas the plot for the hyperfine scheme does contain a region of $0{\cdot}9 < x < 1{\cdot}3$ which can reasonably be reconciled with the two measured values of P_{rr} and P_{rl}. The corresponding value of y amounts to $0{\cdot}45 < |y| < 0{\cdot}73$.

In a forthcoming paper we shall give more details of the calculation and describe a further investigation of this problem involving transitions between particular hyperfine levels and the dependence of the observables on the wavelength of the Ar-ion laser. A final remark concerns the yield of photoelectrons. We are dealing here with a very efficient process for the production of a spin-polarized electron beam. The average current during the light pulses, under optimal conditions for the electron intensity, is of the order of 10^{-7} A. The energy spread amounts to 0·5 eV FWHM. We are aiming for CW operation of the GaAs laser, either the present IBM (see e.g. Picqué 1974) or a different commercially available one.

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