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LETTER TO THE EDITOR

One- and two-photon production of very highly excited states of caesium

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Abstract. Very highly excited p states of Cs with principal quantum numbers $30 \leqslant n \leqslant 85$ were produced by one-step photoexcitation with frequency-doubled light from a flash-lamp-pumped dye laser. Field ionization was employed for selective detection. Ionization occurs at field strengths $E \geqslant E_{min}$ with a sharp threshold. The relation $E_{min}(n^*) \propto (n^*)^{-4}$ was verified over a wide range. Two-photon excitation to a very high state via a virtual intermediate state leads to immediate ionization of most of the excited atoms; in this case the ion signal as a function of the light intensity I is not proportional to I^k, $k \geqslant 2$, whereas a direct two-photon transition into the continuum yields proportionality with I^2.

Highly excited atoms are being studied in many laboratories with various techniques and different scientific as well as technical goals (cf *Physics Today* November 1975). Very highly excited H atoms with quantum numbers n up to 69 have been produced by Bayfield and Koch (1974) in charge transfer to fast protons; optically excited sodium atoms in s and d states up to $n = 37$ were obtained by Ducas *et al* (1975). So far, photoproduction of highly excited atoms involved either one-photon absorption of already collisionally excited atoms (Latimer *et al* 1975, Koch and Bayfield 1975, Koch *et al* 1975) or two-photon absorption in a two-step process via a real intermediate state (Gallagher *et al* 1975, Ducas *et al* 1975, Fabre and Haroche 1975, Ambartsumyan *et al* 1975). Here we report the production and detection of very highly excited caesium atoms up to $n \simeq 85$ obtained with one-photon excitation from the ground state.

An atomic beam of caesium was crossed by a laser beam in a region between capacitor plates used for producing an ionizing electric field. The Cs^+ ions were extracted and detected by means of a channel electron multiplier.

Light from a flash-lamp-pumped dye laser, tuned by an intercavity interference filter and a Fabry–Perot etalon, was frequency-doubled by means of a KDP crystal. Since the wavelengths needed for one-step excitation of very highly excited caesium atoms are in the neighbourhood of 319 nm the laser was set at $\lambda \sim 638$ nm, which is favourable for the use of the dye rhodamin 6G dissolved in water. The laser operated with a peak power of about 1 kW after frequency-doubling. The light-pulse length was 0·3 μs and the line width about 0·3 cm^{-1}.

The principal quantum number n of the states formed was identified by measuring the wavelength of the dye-laser light with a monochromator (accuracy 0·01 nm), which had been calibrated with a He–Ne laser ($\lambda = 632{\cdot}8$ nm), and comparing the measured values with the very accurate spectroscopic λ(air) data of Kratz (1949). Given the

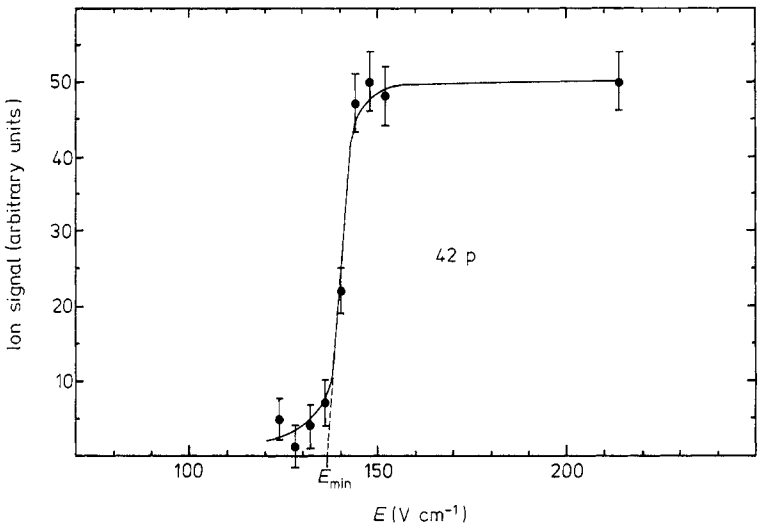

Figure 1. Ion signal plotted against electric-field strength for field ionization of caesium atoms in the 42 p state.

band width of the laser line and taking into account the frequency instability of the flash-lamp-pumped dye laser we estimate that above $n \sim 40$ two adjacent states can be excited with one laser setting and three states above $n \sim 70$.

Under our experimental conditions (Cs atom density in the beam $\sim 10^8$ cm^{-3}, interaction volume $\sim 0{\cdot}004$ cm^3) we observed typically five very highly excited atoms per laser shot. Background counts were negligible. The atomic-beam system is differentially pumped and equipped with liquid-nitrogen-cooled slits and traps; the pressure in the interaction region was about 3×10^{-9} Torr. The counting electronics are gated with an open time of 1 ms immediately after each shot; in this way multiplier dark pulses were sufficiently suppressed.

Figure 1 shows the ion signal as a function of the applied electric-field strength for field ionization of Cs(42 p) atoms. The very sharp threshold at $E = E_{min}$ is similar to the measurements of Ducas *et al* (1975) for field ionization of s states of sodium. On the other hand, Ducas *et al* found a much less pronounced threshold for field ionization of d states of sodium.

The measurements of $E_{min}(n^*)$ are shown in figure 2. The effective quantum number is given by $n^* = n - \Delta$, where $\Delta = 3{\cdot}6$ for the p states of caesium. Without the Stark effect, the theory gives $E_{min}(n^*) = 16(n^*)^{-4}$ au (Ducas *et al* 1975); this relation is shown as a broken line in figure 2. Because of the Stark effect the actual minimum field strength is larger.

Two-photon excitation via a virtual intermediate p state was studied by simply removing the frequency-doubling crystal. We found that without any ionizing electric field between the capacitor plates about 80% of the very highly excited atoms were ionized immediately in the electromagnetic field of the laser light (peak power 80 kW without frequency-doubling). Immediate ionization was observed for $29 \leqslant n \leqslant 85$. Measurements of the ion signal as a function of the light intensity I, obtained with a photon energy equal to half the excitation energy of the $n = 42$ state, are shown as open circles in figure 3. The signal does not increase proportional to I^2, as one would expect for a two-photon process. At high intensities the signal is proportional

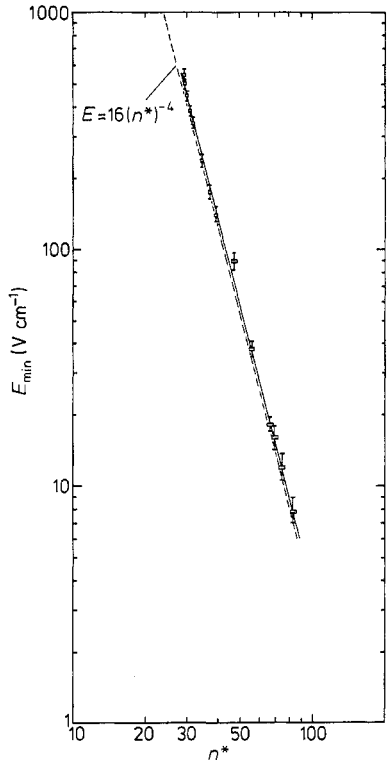

Figure 2. Minimum electric-field strength for field ionization of caesium p states plotted against their effective quantum number.

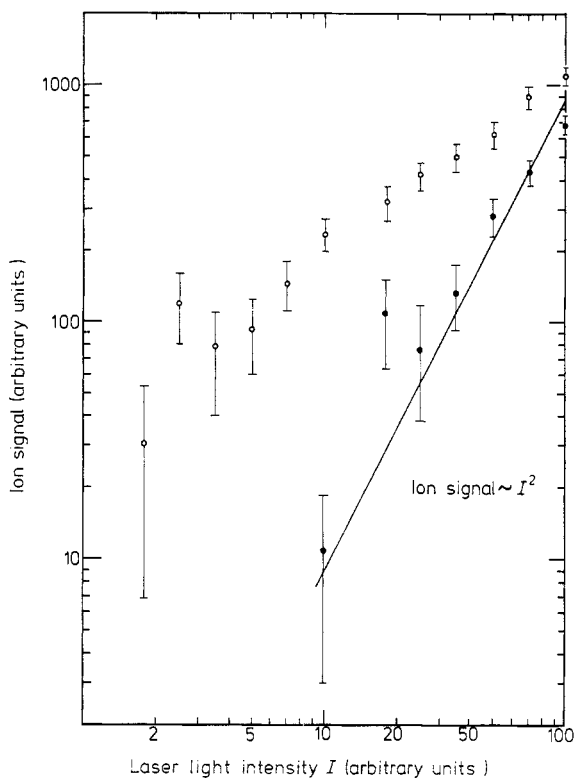

Figure 3. Ion signal plotted against light intensity obtained without electric field in the interaction region. Open circles: two-photon excitation of the 42 p state (9 meV below ionization threshold); full circles: two-photon excitation of a continuum p state (88 meV above threshold).

to I^k with $k \lesssim 1$; the measurements are not yet accurate enough to decide whether $k = 2$ is approached at low light intensities. For comparison we measured the ion signal obtained with laser light of slightly shorter wavelength for which two-photon excitation led to a continuum state. For this direct two-photon ionization the ion signal is proportional to I^2 (figure 3, full circles).

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