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Above-threshold ionization of cesium by 1.9-μm light

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Multiphoton ionization of cesium atoms has been studied with both linearly and circularly polarized light pulses of 1.9-μm wavelength. Photoelectron energy spectra show that the use of circular polarization leads to a strong reduction of above-threshold ionization (ATI), in comparison to linear polarization, and not to suppression of low-order ATI peaks. This behavior is different from that expected for hydrogen under similar circumstances. Calculations reproduce the behavior of cesium and indicate that it can be attributed to the large size of the ground state of cesium.

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The advent of high-intensity laser systems has made it possible to systematically study above-threshold ionization (ATI), the emission of an electron from an atom by the absorption of more photons than the minimum required for photoionization. Although the basic features of the process are fairly well understood [1–4], new and surprising aspects of ATI are still being discovered [3,5,6] as more sophisticated laser systems become available. Calculations of ATI are usually done with one-electron models, whereas most experiments are done on the rare gases, which have more than one electron in the outer shell. The fairly good agreement between such calculations and experiments indicates that the one-electron approximation is reasonable, and that the details of the atomic structure are not very important for the shape of the ATI spectrum. It remains unclear, however, if the remaining differences are to be attributed to the improper modeling of the atom or to an incomplete understanding of the ATI process (see, e.g., Refs. [5,6]). Relatively few experiments on ATI have been done using hydrogen [7,8] or alkali-metal atoms [9,10]. There is a clear need for more experiments on such (effectively) one-electron atoms, so that experiment and theory can be more closely compared under a wider range of circumstances.

The alkali-metal atoms have the disadvantage that they ionize rather easily, due to their small ionization potentials. This makes it hard to reach a sufficiently high intensity to observe an appreciable amount of ATI before the ground state of the atom is depleted. One way to circumvent this problem is to use very short light pulses, as was done by Nicklich *et al.* [10] using cesium atoms and 620-nm light pulses. Although only two photons were necessary for ionization in that case, a large number of small ATI peaks was observed due to the short pulse duration (40 fs) and the excellent sensitivity of their experimental setup. Another method to observe appreciable ATI, used in the present paper, is to decrease the photon energy, so that a large number of photons is required for ionization, even for an alkali-metal atom.

We present experiments on multiphoton ionization of cesium using 1.9-μm light. At least six photons are needed for ionization in this case, comparable to the number of visible photons required to ionize hydrogen or xenon, and up to seven ATI peaks are observed. The results can also be compared to recent results on excess-photon detachment from the negative chlorine ion [11], since the same wavelength was used there, and the ionization potential of Cs is very similar to the detachment potential of Cl^- (3.9 eV versus 3.6 eV). Our results show a strong dependence on the polarization of the light. When changing from linear to circular polarization, ATI is found to be strongly reduced in the latter case, and the average electron energy moves to lower values. This behavior is different from that observed for Cl^- [11] and that expected for hydrogen under similar circumstances [12], where using circular polarization results in more ATI. We have done calculations that reproduce the behavior of cesium and show that the difference with hydrogen can be attributed to the larger size of the cesium ground state.

In the experiment, a beam of atomic cesium is crossed with a $\lambda = 1908$ nm light beam in a magnetic-bottle electron spectrometer [13]. The energy of the light pulses entering the spectrometer is monitored by a photodiode. The electron signals are collected into several intensity bins on the basis of the single-shot photodiode signal. The atomic beam is created by heating bulk cesium in an oven and passing the vapor through a hot ($T \approx 500$ K) pipe, to decrease the amount of cesium dimers in the vapor [14]. A small exit hole in the pipe and a liquid-nitrogen-cooled skimmer define the atomic-beam direction. The 1908-nm radiation is generated by Raman shifting the 1064-nm output of a Nd:YAG (yttrium aluminum garnet) laser in a 2-m-long cell containing molecular hydrogen at a pressure of ≈ 12 bar [11]. The resulting first Stokes line ($\lambda = 1908$ nm) is separated out by a Pellin-Broca prism. A 4-ns or a 20-ps pulse of 1908-nm radiation could be generated by using a seeded nanosecond or a mode-locked picosecond Nd:YAG laser, respectively. Special care was taken to ensure that linearly polarized light entered the Raman cell, to suppress rotational Raman transitions. The 1908-nm light was sent through a polarizer and a Soleil Babinet compensator to allow the polarization to be varied and through a spatial filter to ensure a good spatial beam quality. The 1908-nm light was focused into the spectrometer by $f/6$ optics; the size of the focus was measured to be 20 μm (nearly diffraction limited) by scanning a slit across the focus. The intensity was varied by adjusting the energy of the

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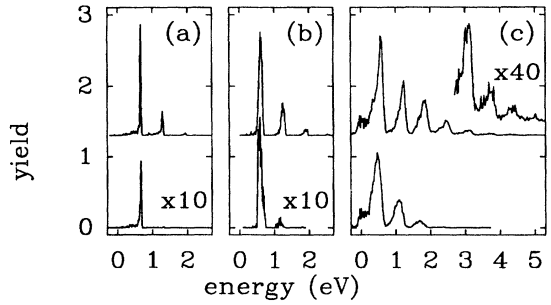

FIG. 1. Photoelectron energy spectra of cesium, irradiated by 1908-nm light. Lower traces are circular polarization; upper traces linear polarization (with an offset). (a) 4-ns pulse, $I=70$ GW/cm^2; (b) 20-ps pulse, $I=0.13$ TW/cm^2; (c) 20-ps pulse, $I=3$ TW/cm^2. In (a) and (b) the circular-polarization data have been magnified by a factor of 10 for visibility. In (c) part of the linear polarization data is also magnified, to show the extent of the ATI peaks.

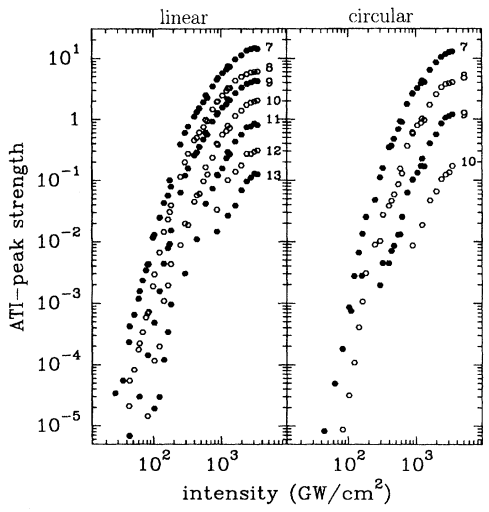

FIG. 2. Intensity dependence of the strengths of the different peaks in the ATI series visible in Fig. 1, for ps laser pulses, for both linear and circular polarization. The number of photons absorbed from the ground state for each peak is also indicated.

1064-nm light that entered the Raman cell. We verified experimentally that the 1-T magnetic field of the spectrometer had a negligible effect on the multiphoton ionization process by varying the direction of the linear polarization from parallel to orthogonal to the magnetic field.

The resulting electron spectra for both linearly and circularly polarized light for different pulse durations and intensities are shown in Fig. 1. A series of well-separated ATI peaks is observed. Up to seven ATI peaks are visible in the case of linear polarization. The absorption of six photons of 1908-nm light from the cesium $6s$ ground state would lead to electrons with a very small energy (4 meV). This peak is not observed in the experiment. A 10-V extraction voltage was applied in the interaction region to verify that the absence of the six-photon peak was not caused by a bad transmission of low-energy electrons in our spectrometer. We conclude that the lowest-order ionization process is suppressed by the increase in the ionization potential due to the ac Stark shift, the so-called channel closure. This is consistent with an estimate of the intensity in our laser focus and the calculated intensity dependence of the ionization potential. The broadening towards lower energies of the peaks is explained by the combined effect of the ac Stark shift of the ground state of Cs, the incomplete cancellation of the shift of the continuum threshold by ponderomotive acceleration (focusing conditions are such that we are in the "intermediate pulse" regime [4]), and space charge. Figure 2 shows the intensity dependence of the strengths of the ATI peaks for the ps pulses. From the intensity dependence we find a saturation intensity of $I=2.4\times10^{11}$ W/cm^2 and $I=4.4\times10^{11}$ W/cm^2 for linear and circular polarization, respectively [15]. As an aside we note here that these saturation intensities (using 1908-nm light) are higher than those found using 1064-nm light with a similar pulse duration [9]. The cesium atom thus does not show the unusual decrease of saturation intensity for the long wavelength, found for Cl^- [11] when comparing 1064- to 1908-nm light.

The most prominent feature of the data is that ATI is found to be suppressed when using circular polarization instead of linear polarization. Note that at peak intensities in the laser focus of $I=3\times10^{12}$ W/cm^2 [Fig. 1(c)] the circular-polarization spectra show less ATI than the linear polarization spectra, even though the atoms can survive up to higher intensities in the circular-polarization case [16].

In order to explain these results, we have performed a relatively simple model calculation. The observed saturation intensities are relatively low. Thus the ponderomotive potential U_p, which is the minimum kinetic energy of a free electron quivering in the light field, is always smaller than the photon energy, and smaller than the binding energy E_i of the initial state ($U_p/\hbar\omega<0.25$, $U_p/E_i<0.04$). This implies that we are in the multiphoton regime, and not in the tunneling regime. To describe ATI processes at such intensities, several approximate approaches have been put forward in the literature [17–19]. We have resorted to a procedure which we believe is the simplest possible while retaining the essential features [20]. In short, the photoionization process is split into two parts, the first part being the absorption of a sufficient number of photons (i.e., six) for the electron to become (nearly) free, the second part being the distribution of the electron over the various ATI channels. This second part is calculated by projecting the wave function resulting from the first part onto Volkov states, the plain-wave solutions for a free electron quivering in an electromagnetic field [18]. This is the same continuum dynamics as is used in the quasistatic model [21], but treated quantum mechanically. Since we are not in the tunneling regime, the first step has to be treated differently from the quasistatic model and is treated in standard lowest-order perturbation theory, taking the interaction of the valence electron with the light as the perturbation [17]. In the first part, the difference between linear and circular polarization manifests itself in the different selection rules for the magnetic quantum number m, $\Delta m=0$ and $\Delta m=+1$ per photon for linear and circular polarization, respectively. In the second part, it is manifest in the different quiver motions of the electron in the Volkov states for linear and circular polarization, respectively.

The calculation was done on cesium and, for comparison, on hydrogen. For cesium the atomic potential for the

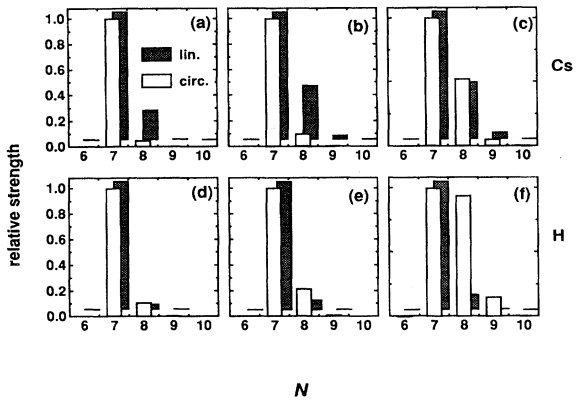

FIG. 3. Calculated ATI spectra for cesium and hydrogen, for both linear and circular polarization. Each spectrum is normalized to the strongest peak, N is the number of photons absorbed from the ground state. (a)–(c) Cesium: (a) $I=70$ GW/cm^2, (b) $I=0.13$ TW/cm^2, (c) $I=0.2$ TW/cm^2, and 0.4 TW/cm^2, for linear and circular polarization, respectively. The intensities were chosen to match the intensities of Fig. 1 [(c) corresponds to the experimental saturation intensities]. (d)–(f) Hydrogen: (d) $I=3$ TW/cm^2, (e) $I=6$ TW/cm^2, (f) $I=9$ TW/cm^2, and 17 TW/cm^2, for linear and circular polarization, respectively. The intensities in (d)–(f) have been chosen such that $U_p/\hbar\omega$ is the same as in (a)–(c).

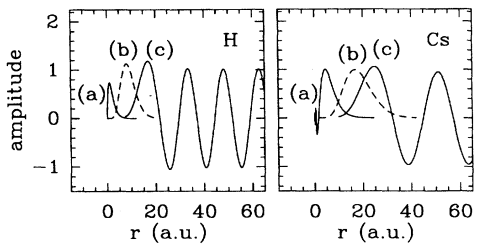

FIG. 4. Radial wave functions for hydrogen and cesium, in the case of circular polarization, for seven-photon absorption. (a) Ground-state wave functions; (b) ground-state wave functions multiplied by r^7; (c) continuum final state with $l=7$.

outer electron is approximated by a central field $V(r) = -1/r - \exp(-\alpha r)(Z-1)/r$, with Z and α adjusted to yield approximately the right energies for the s and p states of cesium ($Z=55$, $\alpha=2.4333$). The $6s$ state in this model potential is taken as the ground-state wave function for cesium. For the calculation on hydrogen $V(r)=-1/r$, $1s$ is the initial state. The wavelength ($\lambda=547$ nm) and the intensity of the light for the hydrogen calculation were chosen such that the ratios $E_i/\hbar\omega$ (the order of the ionization process) and U_p/E_i (the scaled intensity) were the same as in the cesium calculation. Note that $U_p/\hbar\omega$ is also conserved with this choice.

The results of the calculation are shown in Fig. 3. The calculation for cesium reproduces the polarization dependence observed in the experiment. For the intensities used here circular polarization shows less ATI than linear polarization. The amount of ATI observed in the experiment for circular polarization is also reproduced well by the calculation. For linear polarization the agreement is not as good, the experiment shows even more ATI than the calculation. This discrepancy may be due to the neglect of the atomic core in the final-state wave function in our calculation. With linear polarization this core might have important effects: low-angular-momentum states, which are deformed by penetration of the atomic core, are allowed as final states. Furthermore, the electron may be driven back by the light field after it has become free, and rescatter inelastically from the atomic core, leading to much higher electron energies [5,2,6]. This rescattering mechanism could be responsible for the additional ATI observed in the experiment, compared to our calculation for linear polarization.

The results for hydrogen are also shown in Fig. 3. In this case circular polarization yields more ATI then linear polarization. These results for hydrogen behave similarly to other calculations using a nearly identical wavelength [12,22]. They also compare favorably with experiments on hydrogen, using linear polarization [7]. Unfortunately we are not aware of any experimental results under similar conditions on hydrogen for circular polarization. From the calculation we also find saturation intensities for multiphoton ionization. These are 3×10^{11} W/cm^2 and 6×10^{11} W/cm^2 for linear and circular polarization, respectively, for cesium; and 1×10^{13} W/cm^2 and 7×10^{13} W/cm^2 for hydrogen. Note that the saturation intensities for cesium from this calculation agree reasonably well with the saturation intensities found in our experiment.

We have found a simple qualitative explanation for the difference in behavior of hydrogen and cesium; it is illustrated in Fig. 4. The wave functions for hydrogen show the mechanism by which circular polarization suppresses low-order ATI peaks, first proposed by Bucksbaum *et al.* [1]. For circular polarization, the perturbed ground-state wave function can, in the absence of resonances, be approximated by multiplying the ground-state wave function by r^N, where N is the number of photons absorbed. A high-order perturbation thus strongly emphasizes the large-r part of the initial state. The final-state wave function has a a high angular momentum l, due to the $\Delta m=+1$ selection rule for each photoabsorption. It is repelled from the atomic core by a strong centrifugal barrier $l(l+1)/2r^2$. For the first ATI peak ($N=l=7$), the perturbed wave function does not extend to such a large radius, and the spatial overlap with the continuum is poor. When additional photons are absorbed the perturbed wave function extends to larger radii, and the (higher-energy) continuum wave function can penetrate deeper into the core, improving the spatial overlap. The ATI spectrum is now determined by the competition between the improving spatial overlap as more photons are absorbed and the decreasing probability of higher-order ATI due to the weakness of the perturbation [1,23]. The result of this competition is highly dependent on the allowed angular momentum of the final state and on the energy of the outgoing electron. For Xe and $\lambda=1064$ nm, this can lead to the suppression of the four lowest-energy peaks [1]. For hydrogen and $\lambda=547$ nm, it leads to the relative suppression of the $N=7$ peak, as was visible in Fig. 3.

In the case of cesium, however, as is evident from Fig. 4, the size of the perturbed wave function is much larger. This

can be traced back to the large spatial extent of the ground-state wave function of cesium. It is clear that in this case the overlap with the final state is much better than for hydrogen. Furthermore, this overlap does not improve significantly after the absorption of additional photons (in fact it decreases slightly). This shows that the centrifugal-barrier mechanism for suppression of low-order ATI is not applicable in this case for cesium.

Note that the different polarization dependence of the ATI spectrum of cesium and hydrogen cannot be explained in terms of the continuum dynamics for a free electron alone, since $U_p/\hbar\omega$ is exactly the same in both cases. According to this continuum dynamics, the main peak for circular polarization is expected at an energy of $2U_p$ [21]. For Figs. 3(c) and (f), $2U_p/\hbar\omega=0.4$, well below the seven-photon peak. Thus the anomalously large eight-photon peak in hydrogen is a consequence of the relative suppression of the seven-photon peak by the initial ionization step.

We can now qualitatively understand why the Cl^- negative ion shows suppression of the low-order ATI peaks by circular polarization [11], in contrast to cesium under similar conditions. The ground-state wave function of Cl^- is much smaller due to the shorter range of the attractive potential for the outer electron, so that the centrifugal barrier plays a much more important role.

Another interesting comparison is between the present experiment and six-photon ionization of xenon by 532-nm light [1,24], which has similar $E_i/\hbar\omega$ and U_p/E_i, but a lower allowed final-state angular momentum ($l\geqslant 5$ versus $l\geqslant 7$ in cesium). This Xe experiment does not show suppression of the low-order ATI peaks for circular polarization, but an approximate calculation still does (the crosses in Fig. 3 of Ref. [1]). This discrepancy may be due to the approximations involved in the calculation. Note that the one-electron model calculations described above cannot be applied quantitatively to multielectron systems like Xe and Cl^-.

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