Observation of Resonant Excess-Photon Detachment via a Window Resonance in the Cesium Negative Ion

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Efficient excess-photon detachment to lowest nonvanishing order is observed with long ($\sim$0.7 ns) optical pulses and intensities of $\leq 6\times10^9$ W/cm^2. A Feshbach ("window") resonance in Cs^- suppresses single-photon detachment, while simultaneously enhancing the probability of absorption of one or more excess photons. The perspectives for future spectroscopic excess-photon detachment and studies of direct two-electron multiphoton detachment are briefly discussed.

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One of the more interesting effects observed when an atom is exposed to an intense laser field is the process of excess-photon absorption (EPA). In EPA the atom absorbs more photons than the minimum number required to reach the ionization limit. This phenomenon was first observed for neutral atoms in 1979 by Agostini *et al.* [1] and has since been a topic of intense investigation (see overviews of Refs. [2,3]). The similar process in negative ions, known as excess-photon detachment (EPD), was not observed until last year [4–6]. The study of EPD represents a new situation in strong-field atomic physics since the additional electron in a negative ion is bound in a short-range potential as opposed to the long-range Coulomb potential. Furthermore, the complex substructure in EPA spectra for neutral atoms due to transient resonances [7] is not expected to appear in multiphoton absorption in negative ions, because these systems in general lack excited bound states. These special features simplify theoretical descriptions and make calculations on strong-field phenomena in negative ions attractive [8–10].

Resonances in multiphoton detachment may occur, however, since there exist doubly excited states above the detachment limit [11,12]. In the present work, we show how EPD from the ground state of Cs^- is strongly enhanced when the first photon reaches an energy that coincides with the $Cs^-(6p_{1/2}7s)$ Feshbach state (Fig. 1). At this energy, the cross section for single-photon detachment from $Cs^-(6s^2)$ exhibits a dramatic minimum, which is caused by the interference between excitation of the $Cs^-(6p_{1/2}7s)$ state and the $6s\varepsilon l$ continuum [13–15]. The resulting Fano profile [16] exhibits a single-photon cross section which drops 2 orders of magnitude over an energy range of 20 cm^{-1}. The minimum should facilitate absorption of additional photons since depletion of the ground-state Cs^- ions during the rise time of the pulse is minimized. At the same time, the presence of the $6p_{1/2}7s$ state is expected to favor excess photon absorption compared to the similar process via a pure virtual state. Our experimental technique, which is based solely on detection of neutral atoms and positive ions, can be an efficient way of investigating EPD and related strong-field phenomena in negative ions as well as a sensitive spectroscopic tool. Moreover, future theoretical treatment should be feasible due to the low number of photons involved in the absorption processes.

The experimental setup is described only briefly since a more detailed description is given elsewhere [17]. A 25-keV Cs^- beam of typical ion current $\sim$5–10 nA enters an ultrahigh vacuum chamber where it is crossed at 90° with the linearly polarized, focused laser beam. Following the interaction region, the ion beam is charge-state analyzed and Cs and Cs^+ are detected by two electron multipliers. The Nd:YAG (yttrium aluminum garnet) pumped ns dye laser is used in the visible range between 14800 and 15600 cm^{-1}. It is focused into the chamber by a 32-cm-focal-length lens giving a maximum intensity

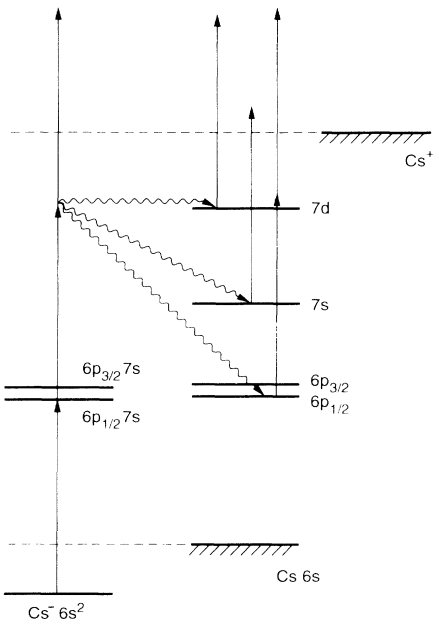

FIG. 1. Simplified energy-level diagram for Cs^- and Cs illustrating excess-photon absorption via the $Cs^-(6p_{1/2}7s)$ Feshbach state. The photon absorption processes lead to double ionization of the Cs^- ion by either a simultaneous removal of the two electrons or a sequential removal via an excited Cs state. For visible irradiation, the photon energy greatly exceeds the detachment limit, as the Cs^- electron affinity is 3803 cm^{-1} [12].

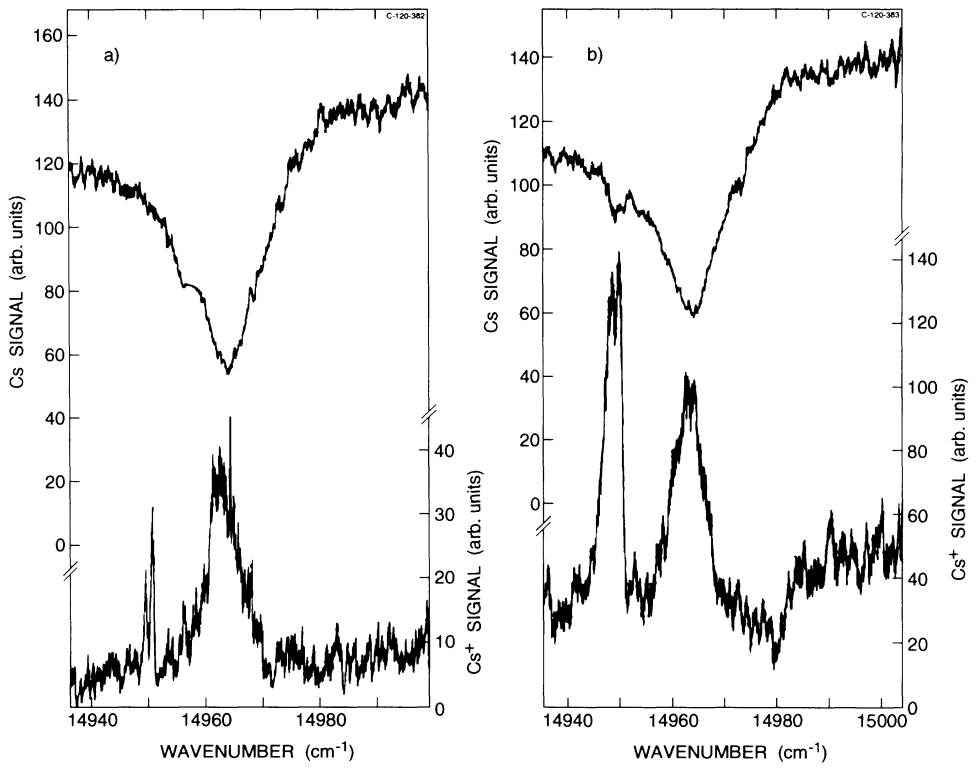

FIG. 2. The Cs^+ signal (lower curves) and the Cs signal (upper curves) vs the laser wavelength in the region of the $Cs^-(6p_{1/2}7s)$ state and for intensities of (a) 1×10^9 W/cm^2 and (b) 6×10^9 W/cm^2. Each spectrum shown is obtained from a single laser-wavelength scan, and via an analog detection scheme.

of $\sim6\times10^9$ W/cm^2. The interaction time of the Cs^- ions with the laser beam is given by the transit time through the laser focus, which in the present experiment is ~0.7 ns. Most data are taken in an analog regime utilizing a gated integrator and a boxcar averager. In measurements of the intensity dependence, digital data collection is used.

The experimental results are shown in Fig. 2. The upper and lower curves show the Cs and the Cs^+ signal, respectively, as the wavelength of the laser is scanned. The Cs signal exhibits the expected pronounced minimum centered around 14963 cm^{-1}. In the Cs^+ spectrum two strong resonances are observed when the laser is tuned either to ~14949 cm^{-1} or to the Fano minimum in the Cs production (~14963 cm^{-1}). The 14949-cm^{-1} resonance is interpreted as resulting from single-photon detachment of $Cs^-(6s^2)$ to $Cs(6s)$ followed by (2+1)-photon ionization via $Cs(11d)$. At the highest intensities, the resonant enhancement is sufficiently strong that depletion of the Cs ground state occurs. This effect is seen as a hole in the Cs spectrum [Fig. 2(b)].

A similar scheme cannot explain the resonance at ~14963 cm^{-1} since single-photon detachment at this wavelength is at the Fano minimum, and three-photon ionization of $Cs(6s)$ is not resonantly enhanced at 14963 cm^{-1}. Also, the two resonances behave differently upon decreasing the intensity. Whereas the $11d$ resonance narrows and splits into the two fine-structure components, the width of the resonance at 14963 cm^{-1} is unchanged (Fig. 2), clearly indicating that the latter resonance is related to a broad autodetaching state. More precisely, when the laser is tuned to ~14963 cm^{-1}, $Cs^-(6s^2)$ absorbs at least two photons via the $6p_{1/2}7s$ Feshbach state. Cs^+ is produced by either direct double detachment after absorption of three photons, or by a sequential removal of the electrons via an excited Cs state (Fig. 1). To possibly clarify whether one of the processes dominates we investigated the intensity dependence of the Cs^+ production at 14963 cm^{-1}. For intensities below $\sim2\times10^9$ W/cm^2 the Cs^+ signal scales approximately as $I^{1.3}$, whereas it saturates at higher values of the intensity. This low-order dependence indicates that a considerable fraction of the Cs^+ ions are created as a result of successive single-photon processes (1+1+1). As shown in Fig. 1, the sequential mechanism occurs as a (1+1+1)-photon process when the $Cs(7s)$ or $Cs(7d)$ state acts as an intermediate state. However, the direct mechanism may also effectivly proceed as a (1+1+1)-photon process due to the possible presence of autodetaching Cs^- states after absorption of the second photon. To further resolve this issue photoelectron spectroscopy may be necessary.

To verify our interpretation of resonant EPD we recorded the Cs^+ signal when the negative cesium ions are exposed to a weak infrared pulse (~9398 cm^{-1}) pri-

or to the intense visible pulse (Fig. 3). The infrared prepulse transforms all Cs^- ions to $Cs(6s)$ so that Cs^+ is solely produced from subsequent multiphoton ionization of $Cs(6s)$. The enhancement at 14949 cm^{-1} does not disappear, establishing that it originates from resonant multiphoton ionization of the ground state of Cs. In contrast, the broad feature at 14963 cm^{-1} completely vanishes, demonstrating that this resonance indeed results from structure in the Cs^- ion.

Figure 3 also enables us to understand Cs^+ production away from the Feshbach resonance at the highest intensities [Fig. 2(b)]. From a comparison of Fig. 3 and Fig. 2(b) it is seen how most of this background disappears when the infrared prepulse is included. Applying the same argument as above, this reveals that the background production of Cs^+ predominantly originates from a sequential removal of the electrons via an excited Cs state. When the laser is tuned above the opening of the $Cs(6p_{1/2})$ channel (14981 cm^{-1}) Cs^+ is likely formed via two-photon ionization of this state. Below the $Cs(6p_{1/2})$ limit, Cs^- absorbs at least two photons, probably quasiresonantly, and enhanced by the near presence of the Feshbach state. At the $Cs(6p_{1/2})$ threshold a dip in the Cs^+ signal is observed. This is due to an increase in the partial cross section for detachment to the $Cs(6s)$ state known as the Wigner cusp [15]. Hence, a larger fraction of Cs^- is transformed to $Cs(6s)$ from where production of Cs^+ is not very likely to occur. This dip strongly supports the interpretation of EPD being responsible for creation of Cs^+.

In order to investigate the importance of the depth of the window resonance on EPD, photon absorption

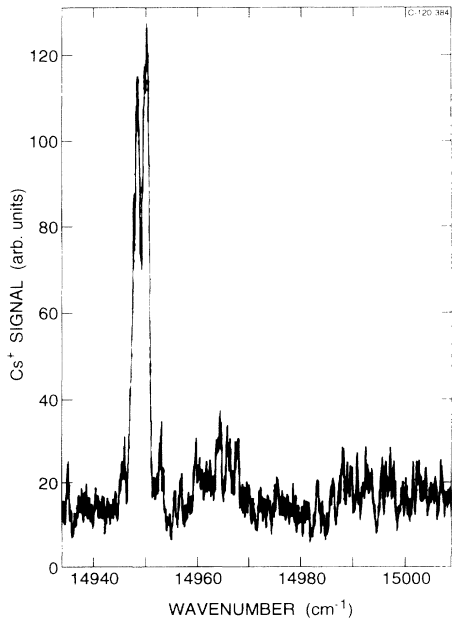

FIG. 3. The Cs^+ signal vs the laser wavelength where an infrared prepulse is applied (see text).

through the shallower $Cs^-(6p_{3/2}7s)$ feature [13,14] below $Cs(6p_{3/2})$ is studied (see the energy-level diagram of Fig. 1). A broad, less significant structure centered around $\sim$15520 cm^{-1} is observed along with a sharper peak due to (2+1)-photon ionization of $Cs(6s)$ through $Cs(20d)$. These data suggest that excess photon absorption via $Cs^-(6p_{3/2}7s)$ is less likely than via $Cs^-(6p_{1/2}7s)$. This may be partly explained by the fact that single-photon detachment at the $6p_{1/2}7s$ position is suppressed considerably more than at the $6p_{3/2}7s$ position [15]. Also, when the $6p_{1/2}7s$ state is excited the wavelength of the laser is close to the $Cs(6p_{1/2}) \rightarrow Cs(7d)$ transition, meaning that it is primarily the $6p_{1/2}$ electron that is excited [18]. This is expected to enhance the photoabsorption cross section significantly. In contrast, a similar near-resonance condition is not present when the $6p_{3/2}7s$ state is excited. An extension of the present available calculations of single-photon detachment [15] to higher-order processes should be feasible [18] and together with photoelectron spectroscopy experiments should lead to a detailed understanding of EPD in Cs^-. Furthermore, Robicheaux and Gao [19] have recently argued that a peak will almost always appear in the two-photon absorption via an autoionizing state, independent of the shape of the one-photon autoionizing resonance. An extension of the present experimental work to other negative ions should provide excellent tests of the dependence of EPD on the Fano profile of the single-photon detachment cross section.

At this point, we would like to address the similarities between our observations and a previous experiment by Gallagher *et al.* [20]. In the present experiment excess photon absorption in a negative ion occurs through a minimum in the single-photon detachment cross section caused by the interference between an autodetaching state and the continuum. In the previous experiment, two-photon absorption from bound Ba $6snd$ Rydberg states to autoionizing Ba $7snd$ states was studied. This process was shown to be very efficient when the two-photon absorption proceeded through a minimum in the single-photon excitation cross section. There the minimum was due to minimal overlap between the wave functions for the outer nd electron in the initial bound state and in the intermediate state.

Future work will be directed to exploiting resonant EPD as a spectroscopic tool. Based on an idea by Crance [21], we plan to study doubly excited ^1S and ^1D states in H^- below the hydrogenic $n=2$ threshold by means of two-photon excitation of $H^-(1s^2)$. In contrast to the present Cs^- work, the resonant enhancement is caused by the final state for two-photon absorption, not by the intermediate state. According to recent calculations [22], the two-photon absorption cross section is resonantly enhanced more than 4 orders of magnitude due to these states. If short picosecond pulses are employed in the experiment, this enhancement ensures that a sufficiently

large fraction of the H^- ions undergoes two-photon excitation before the population of $H^-(1s^2)$ is depleted due to single-photon detachment.

Finally, the present work also touches on the long-standing question as to whether a double ionization regime, where a simultaneous ejection process dominates, can be achieved. This question has been studied extensively for neutral atoms [23,24]. Here, the overall conclusion is that the dominant mechanism is a sequential removal of the electrons via a low-lying state of the singly charged ion. Negative ions may, however, provide good candidates for the observation of direct double ionization by utilizing detachment minima similar to the one described in this work. A negative ion where absorption of one additional photon via such a minimum brings the system above the second ionization limit might possibly give the first clear evidence of simultaneous ejection of the two electrons in a multiphoton absorption process. A recent publication on Si^- [25] indicates that this system may be a particularly interesting candidate.

In conclusion, effective excess-photon detachment via an autodetaching Feshbach state was achieved with long optical pulses and intensities of a few GW/cm^2. This first resonant multiphoton experiment offers new perspectives for future studies of doubly excited states in these systems. In addition, the work is a demonstration of excess-photon absorption to lowest possible order. This is in sharp contrast to above-threshold ionization studies where the higher ionization potentials of neutral atoms typically leads to studies of high-order processes. Finally, future electron spectroscopic measurements in Cs^- or resonant EPD experiments in a more favorable negative ion may lead to the first clear experimental evidence for a dominating contribution of direct over sequential double ionization.

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