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Electron spectroscopy at thresholds in single and double multiphoton ionization of strontium

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Spectroscopy of photoelectrons produced by strontium atoms irradiated by strong picosecond laser pulses reveals excited states of Sr^+ and, for the first time, electrons from stepwise double ionization. Different threshold behaviors are observed for these transitions.

In this paper, we report new results in multiphoton-ionization photoelectron spectroscopy (MPIPES) of strontium atoms irradiated by strong visible picosecond pulses. Different channels for both single and double ionizations can be identified by this method, while they remain obscure when detecting only the ions. In a recent experiment[1] we have used MPIPES to study the double ionization of strontium in an attempt to untangle the physical processes leading to the removal of two electrons. Some of these processes are illustrated in Fig. 1, where the stepwise channels (1)+(5), (2)+(5), (3)+(6), (4)+(7) have been stressed together with the direct channel (8). The question of the competition between stepwise and direct processes (for the production of multiply charged ions in multiphoton ionization) has been considered from the first experimental demonstration as the central one.[2,3] However, the definition usually accepted for the two types of processes suggested that they were physically completely different. This question has been clarified[4] and it has been shown that the stepwise process is nothing but the resonant part of process (8) in which all the possible intermediate energy-conserving ion states are allowed. It should be pointed out that the two types of processes can be distinguished by electron spectroscopy, since the nonresonant one produces a continuum spectrum while the resonant one produces two electrons of well-defined energies. When the resonant and nonresonant processes involve the same number of photons, it is expected that the resonant process will dominate.[4] In previous experiments measuring MPI of Sr (Refs. 1 and 3) the observations clearly suggested that the stepwise (resonant) process was dominating the direct (nonresonant) one. One of the steps believed to be important was a four-photon ionization leaving the ion in the $5^2P_{1/2,3/2}$ states [process (4)]. However, the slow electrons (0.1–0.2 eV) corresponding to the excitation of the $5P_{1/2}$ and $5P_{3/2}$ states of the ion were not detected. Furthermore, electrons ejected in the double-ionization process have not been clearly identified in experiments either on alkaline earths or on rare gases.[5] We present for the first time evidence for both these processes: electron spectra displaying the corresponding components are reported here, as well as their variations around, on the one hand, the $5s^2\,{}^1S_0$–$5P_{1/2,3/2}$ four-photon thresholds and, on the other hand, the $5S^2$–$4p^6\,{}^1S$ five-photon threshold. There are three results of these measurements: (a) they provide proof that excited states of the ion can easily be reached in MPI of two-electron atoms, (b) they confirm that stepwise multiphoton double ionization is the dominant process, and (c) they allow an experimental check of the continuity of the N-photon above-threshold ionization (ATI) process[6] when the energy of $N-1$ photons crosses the continuum limit.

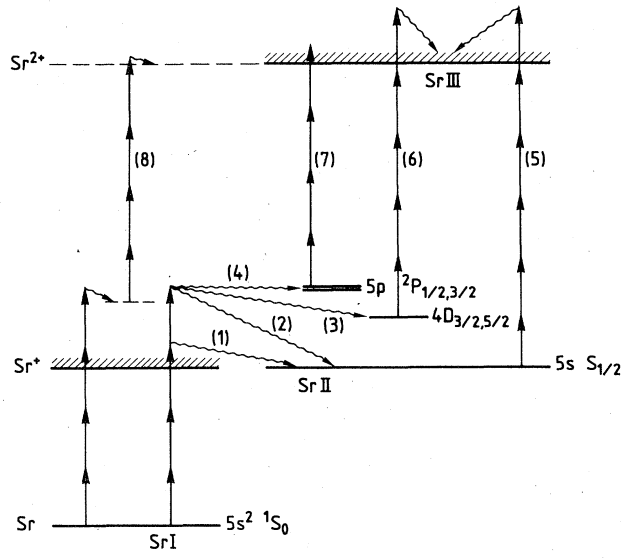

FIG. 1. Energy-level diagram of Sr I and Sr II showing the processes discussed in the text. The process labeled (8) is an example of "direct" or "nonresonant" double ionization.

The experimental setup described before[1-7] has been modified as follows: the start-stop–oscilloscope acquisition device (Lecroy) has been replaced by a true multichannel counter with computer system[6] allowing a higher sensitivity and easy treatment of the data. The average laser power has been stabilized to within ±5% by a servo-controlled Fresnel-prism Glan-polarizer attenuator. Finally, we have used a shorter-focal-length ($f=70$ mm) focusing lens allowing intensities of the order of 10 TW cm^{-2} without stabilization and a few TW cm^{-2} with stabilization.

By adding a small extracting electric field (6.7 V/cm) in the interaction region and keeping the time-of-flight tube 1 V above the interaction-region potential a new component appears in the photoelectron spectra as shown by a typical sample in Fig. 2. Under these conditions the components at 0.9 and 1.3 eV related to processes (1) and (3) are not resolved. The energy of the new peak is in good agreement with the expected structure of two lines (0.1 and 0.2 eV at the wavelength of 559 nm in Fig. 2) corresponding to the

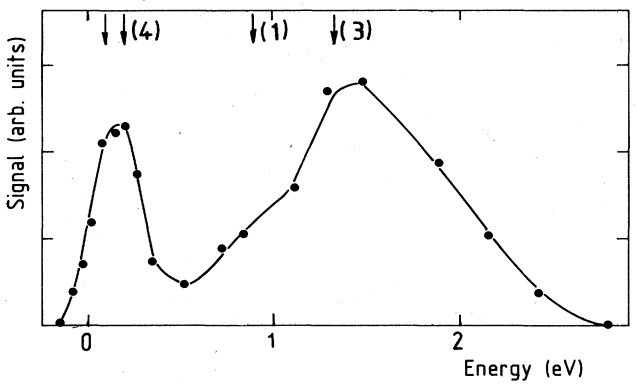

FIG. 2. Typical electron-energy spectrum at 100 Gw cm^{-2}. The numbers inside parentheses refer to the processes illustrated in Fig. 1.

excitation of the ion $5P_{1/2,3/2}$ states. However, the energy calibration of the time of flight is not precise enough to allow an unambiguous identification of this feature. Tuning the laser over the phodamine 6-G range produces resonant variations of this peak due to various intermediate two-electron states, indicating strong configuration mixing. This will be described elsewhere, together with the wavelength dependences of the other peaks reported in our previous experiment.[1] Here we will focus our attention on the variations of this peak around the thresholds of the four-photon $5s^2\,{}^1S_0$–$5\,{}^2P_{1/2,3/2}$ transitions. In order to decrease the overlap with other components of the spectrum, the laser polarization was oriented at 90° from the spectrometer axis. This takes advantage of the angular distribution of the photoelectrons which is peaked along the direction of the laser polarization.[6] The accelerating field deflects the low-energy electrons much more easily, thus resulting in an increase of the corresponding signal relative to the other components of the spectrum. The result is shown in Fig. 3, where the expected threshold behavior is clearly displayed: When the wavelength is larger than 575 nm (the $5P_{1/2}$ threshold is at 574.4 nm), no slow electrons are detected. Between 574 and 568 nm the signal increases up to a sharp maximum and then decreases down to about zero. A similar behavior is observed for wavelengths under 568 nm (the $5P_{3/2}$ threshold is at 567.8 nm). However, it should be noted that the observed variation is the result of a convolution of the ionization probability and the collection efficiency which both vary very rapidly around the threshold. But, nevertheless, the observed variation is a reliable signature for the origin of these slow electrons. This is, to our knowledge, the first direct observation of a threshold in a MPI process which leaves the ion in an excited state, although such a limit had been detected in barium through the enhancement of the auotionization rate of doubly excited states.[8] The positions of the thresholds do not depend on the laser intensity from 70 to 385 Gw cm^{-2}, which proves that the Stark shifts of either the $5s^2\,{}^1S_0$ or the $5P_{1/2,3/2}$ states are negligible within the experimental precision. The identification of the four-photon $5s^2\,{}^1S_0$–$5P_{1/2,3/2}$ transitions completes and confirms the results and conclusions of Refs. 1 and 3.

A large fraction of the electrons from the second ionization are expected to come from five- or six-photon ionization (depending on the laser wavelength) of the $5S_{1/2}$ state of Sr II [process (5) in Fig. 1]. In the case of six-photon

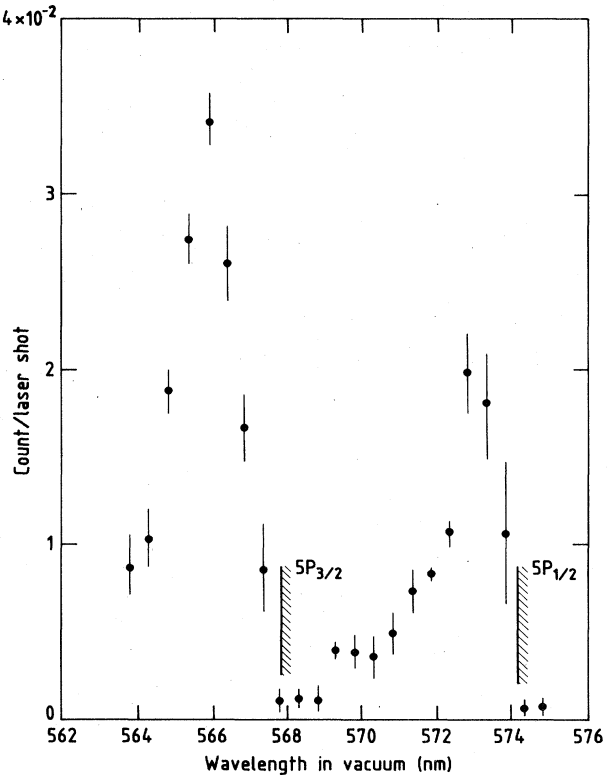

FIG. 3. Wavelength dependence of the 0.1–0.2-eV component around the $5P_{1/2}$ and $5P_{3/2}$ thresholds.

ionization, these electrons have an energy of about 2 eV which makes them easy to detect: By suppression of the accelerating voltages mentioned above, the slow-electron components are cut out of the spectra, and enough resolution is restored to identify a new feature which appears in the spectra at 2.2 eV, provided the intensity is greater than 3 TW cm^{-2} (Fig. 4). However, the spectrometer resolution in this energy range is not high enough to separate the expected peak at 2.2 eV from a component at 1.84 eV expected from the ionization of $4D_{3/2,5/2}$ states of Sr II, as shown in

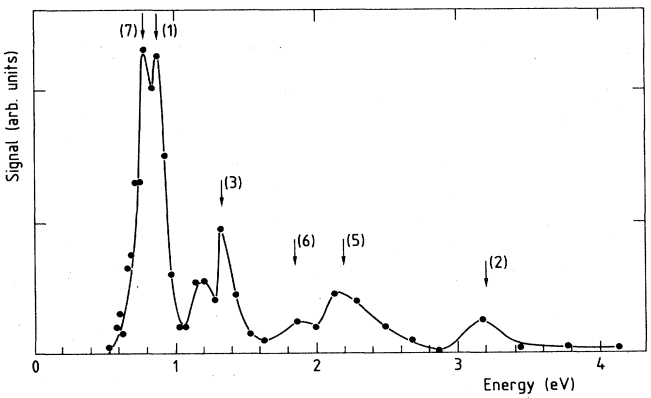

FIG. 4. Typical energy spectrum at 2.8 TW cm^{-2} showing the 2.2-eV component (arrow) corresponding to the second ionization. The numbers in parentheses refer to the processes illustrated in Fig. 1.

Fig. 4. Both of these components are found only at high intensity, which is a good indication of their origin. At this intensity, of a few TW cm^{-2}, the Sr^+-Sr^{2+} ion ratio is about 10%, while the ratio of peak (5) relative to all others is roughly 5%, which indicates that many Sr^{2+} ions are produced through channels (6) and (7). At a wavelength shorter than 562.025 nm (in vacuum) the $5S_{1/2}$ state of Sr II can be ionized by absorption of only five photons. Therefore, for wavelengths shorter than this threshold a peak of very slow electrons should appear in the spectrum. Unfortunately, it is not possible to monitor a slow-electron peak in this wavelength range, since it would not be resolved from the $5P_{1/2,3/2}$ component discussed above. Regarding the variation of the 2.2-eV peak around the five-photon threshold, some prediction can be made: The expected wavelength dependence for the 2.2-eV electron peak would be that of the six-photon cross section when the energy of five photons crosses the continuum limit. Due to the continuity of the wave functions for Rydberg states when $n \rightarrow \infty$ and continuum states when $k \rightarrow 0$, there is no rapid variation of this cross section at the crossing of the continuum limit. The consequence of this general property has been confirmed by several calculations of ATI in hydrogen[9] but had not been checked experimentally so far.[1]

The experimental dependence is shown in Fig. 5, where the arrow points towards the known position of the threshold. In spite of a rather large scatter of the experimental points due to the very small counting rate (of the order of 0.001 count per laser shot, i.e., 0.01 count per second at the maximum stabilized intensity), it is clear that the variation of the amplitude of the electron peak is quite smooth in this region. The signal decreases by about a factor of 2 over the investigated range around the threshold (4 nm). A numerical calculation of the six-photon MPI cross section of hydrogen around the five-photon threshold qualitatively agrees with the observation.[10] However, the continuity of the cross section precludes the threshold test of the origin of the 2.2-eV peak and one has to rely on its energy and intensity dependence to assign it. This first observation of electrons from double ionization is in agreement with our previous conclusions regarding the double-ionization process in strontium atoms by multiphoton absorption.[1]

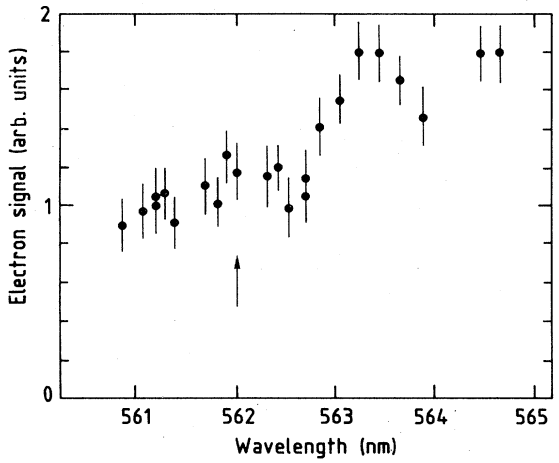

FIG. 5. Wavelength dependence of the 2.2-eV component around the five-photon double ionization threshold (arrow).

To conclude, we would like to emphasize some of the points which have been made. (i) Even when using high-intensity picosecond pulses, not only is the atomic structure not rubbed out, but the ionic states play a dominant role as resonant intermediate steps towards the second-ionization limit. (ii) The direct, nonresonant process (8), albeit small, can probably be detected by MPIPES: there is little doubt that it will be marginal. (iii) Two-electron excited states and configuration mixing are very important ingredients in producing ion excited states. (iv) The perturbative approach, even though considerably complicated by this problem, seems reasonably appropriate to treat the transitions observed in alkaline earths (up to the second ionization) at intensities up to 10 TW cm^{-2}.

Note added. Independently, a similar result was reported with the same interpretation.[11]

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