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# Direct double ionization of the $Ar^+$ $M$ shell by a single photon

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Direct double ionization of the $Ar^+(3p^{-1})$ ion by a single photon is investigated both experimentally and theoretically. The photon-ion merged-beams technique was employed at the Advanced Light Source in Berkeley, USA, to measure absolute cross sections in the energy range from 60 to 150 eV. In this range, three contributions to the double ionization of $Ar^+$ are to be expected: the removal of two $3p$ electrons, of a $3s$ and a $3p$ electron, and of two $3s$ electrons. Among the possible mechanisms leading to double ionization, the TS1 (two-step one) process dominates in the near-threshold region. In TS1, a photoelectron is ejected and, on its way out, knocks out a secondary electron. This two-step mechanism is treated theoretically by multiplying the calculated cross section for direct single photoionization of a given subshell with the calculated $(e, 2e)$ ionization probability for the ejected photoelectron to knock off a secondary electron. The calculated cross section is in very good agreement with the experiment.

## I. INTRODUCTION

Direct, or nonsequential, ejection of two electrons from an atom or a molecule by a single photon is one of the most fundamental few-body processes in atomic physics. This process is different from inner-shell excitation or ionization with subsequent Auger decays, since direct photodouble ionization (PDI) is characterized by the almost simultaneous ejection of two electrons from a neutral or electrically charged atom or molecule that absorbs the incident photon. Characteristic time spans for PDI depend on the mechanism involved and are at most of the order of the flight time of an ionizing electron through an atom, which is typically of the order of or below 100 attoseconds [1]. In contrast, an atomic Auger decay is a much slower process typically taking tens of femtoseconds to proceed [2] but may also take more than several microseconds [3].

Three mechanisms have been discussed and experimentally demonstrated for neutral atoms: the so-called two-step one (TS1), the shake-off (SO), and the quasifree mechanism (QFM) (see Ref. [4] and references therein). In TS1 a photoelectron is ejected which, on its way out, knocks out a second electron. SO is the result of the sudden removal of the photoelectron and the subsequent change of the potential in which the other electrons reside. With a certain probability, one of these electrons may relax to an unbound state; that is, it is shaken off to the continuum. In both of these cases, the photon primarily couples to the dipole formed by one electron and the nucleus. QFM is the quadrupole contribution to photodouble ionization. It is characterized by the back-to-back ejection of two electrons with similar energies while the nucleus is merely a spectator remaining almost at rest.

TS1 dominates the PDI cross section at lower photon energies. At higher energies SO takes over and QFM is generally a very small contribution [4]. TS1 is uniquely facilitated through the electron-electron interaction (EEI). Thus, double photoionization is very sensitive to the details of the EEI. A typical system for studying double photoionization has long been the helium atom, for which numerous experimental and theoretical investigations have been performed (see Ref. [4] and references therein). The theoretical investigations have naturally been extended to heliumlike ions to study the competition of the Coulomb attraction of the nucleus versus the EEI as the charge of the nucleus increases [5]. With increasing nuclear charge the relativistic effects become more important [6].

While numerous experiments on PDI of neutral atoms and molecules have been carried out [7], there are only few experiments yet in which PDI of ions has been investigated (see Ref. [8] and references therein). An advantage of using ion beams as targets for photoionization is the capability for measuring *absolute* cross sections as explained in Sec. II. For atomic targets, typically relative PDI cross sections have been measured, which were then normalized either to theory or to cross sections for photoabsorption. The most recent measurement on photodetachment from $C^-$ ions demonstrated the role of $K$-shell PDI, i.e., double core-hole production, in net multiple (up to fivefold) ionization of an ion with relatively few electrons [9]. The reasons for the scarcity of experimental results for PDI of ions are low cross sections and the low particle densities that can be achieved with ionic targets.

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Considerably larger cross sections are observed for direct double ionization of atoms and ions by electron impact [10].

The present experiment aims at an improved understanding of PDI of ions. By the comparison with a newly developed theoretical approach [11] the power of that theory in predicting PDI cross sections for ions is tested. It is hoped that a broader data base for PDI of ions along isoelectronic or isonuclear sequences will help to establish general scaling laws similar to those found for neutral atoms [12] and for the helium isoelectronic sequence of ions [13]. In particular, PDI of the $Ar^+(3p^{-1})$ ion in its ground-state configuration is investigated. Previously, electron-impact double ionization of $Ar^+$ has been studied experimentally [14,15]. For such processes a satisfying theoretical description is not available to date. $Ar^+$ ions were also targeted in previous single- and multi-photoionization measurements [16,17], however, the aspect of direct photodouble ionization was not addressed in those studies.

## II. EXPERIMENT

The experiments were carried out at the ion-photon-beam (IPB) facility [18] that was previously available at the Advanced Light Source (ALS) in Berkeley, California. The photon-ion merged-beams technique [19,20] was employed. Details of the experimental setup and procedures have been described in detail previously [18,21]. Here, only a brief account of the experiment is provided together with its specific parameters.

$Ar^+$ ions were produced in an electron-cyclotron-resonance ion source. After acceleration to an energy of 6 keV, the ion beam extracted from the source was mass-over-charge analyzed by a dipole magnet. The selected $Ar^+$ beam component was collimated and deflected onto the axis of the interaction region where it was merged with a counterpropagating beam of synchrotron radiation that was made available at beamline 10.0.1.2. By a second dipole magnet the ions were separated from the photon beam. The photon flux was measured by a photodiode. The parent ion beam was collected in a large Faraday cup inside the magnet chamber while the $Ar^{3+}$ product ions were deflected one more time (by 180 degrees out of plane to suppress background from stray particles and photons) before they entered a single-particle detector with a large sensitive area of 15 mm diameter. The available ion-beam current was about 1 $\mu$A. In the investigated energy range of 60 to 150 eV the photon flux varied between approximately 0.7 and $2\times10^{14}$ $s^{-1}$ at a resolution of 200 meV. Background count rates were of the order of 200 $s^{-1}$. The maximum $Ar^{3+}$ signal count rate observed during the measurements was about 1700 $s^{-1}$. It was obtained at approximately 90 eV photon energy where both the cross section and the photon flux were high.

In relative scan measurements the count rate, the photon flux and the ion-beam current were recorded as functions of the photon energy. By subtracting the background count rate measured below the threshold for direct double ionization of $Ar^+$ the true signal rate could be determined. From the measured quantities relative yields of $Ar^{3+}$ photoproducts normalized to photon flux and ion current were obtained. For the measurement of absolute cross sections the overlap of the photon and ion beams had to be quantified. For this purpose a potential of 500 V was applied to a drift tube with a length of $(29.4 \pm 0.6)$ cm coaxially mounted along the merge path with a total length of about 1.4 m. Thereby, the product ions generated inside the drift tube were energy-tagged; that is, they could be separated from ions of the same charge state born outside the interaction region. By applying a nonzero potential to the drift tube, also the effective length of the interaction region is defined. The overall form factor [22] that characterizes the beam overlap was determined from that length and from the results of three area-overlap measurements with horizontal and vertical scanning slits at three positions near the entrance and exit apertures and in the middle of the interaction region. The overall form factors in the absolute cross-section measurements were between 300 and 350 $cm^{-1}$, indicating very good overlap of the photon and ion beams given the beam sizes available at the IPB endstation.

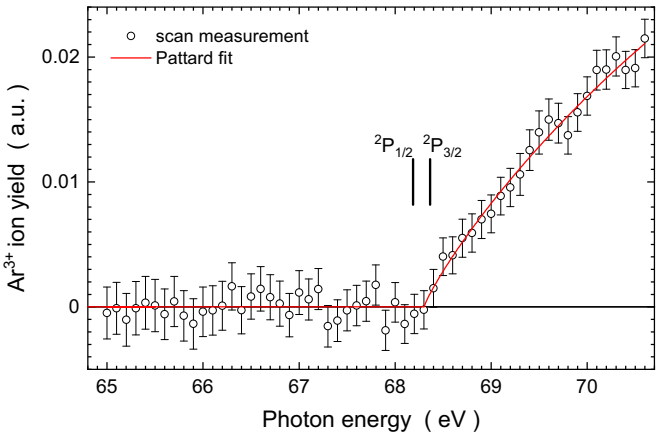


FIG. 1. Ion yield at the double-ionization threshold. The photon-energy resolution was 300 meV. The open circles with one-standard-deviation error bars are from a scan measurement. The red line is a fit to the experimental data on the basis of the Pattard formula [23]. The vertical bars show the lowest double-ionization thresholds for $Ar^+$ in the $^2P_{3/2}$ ground level and the associated (metastable) $^2P_{1/2}$ upper fine-structure level.

Systematic relative uncertainties of absolute measurements were estimated to be $\pm$19% [21]. Statistical uncertainties of each single absolute measurement were reduced to insignificance by choosing long counting times of hundreds of seconds. For energy-scan measurements two-standard-deviation statistical uncertainties at the cross-section maximum were about $\pm$9%. The scan measurements were combined and normalized to the absolute cross sections by multiplication with a suitable factor.

The photon energy axis was calibrated with an uncertainty of $\pm$0.2 eV. Figure 1 shows the result of an energy scan around the expected double-ionization threshold near 68 eV. The $Ar^+$ ion beam used in this experiment is expected to contain ions in two different levels, the $^2P_{3/2}$ ground level and the first-excited metastable $^2P_{1/2}$ level, both forming the ground-configuration fine-structure doublet. For the ground level of $Ar^+$, the minimum energy required to release two electrons is 68.365 eV, for the excited level it is 68.167 eV. These threshold energies can be inferred from the NIST Atomic Spectra Database [24]. They are indicated by the vertical bars in Fig. 1. A fit of

the experimental data with the formula suggested by Pattard for describing PDI cross sections [23] yields an experimental threshold energy of $68.30 \pm 0.13$ eV. The uncertainty of the experimentally determined threshold approximately spans the fine-structure splitting of the ground configuration. The deviation of the experimentally determined double-ionization threshold from the minimum energy needed to release two electrons from the metastable level is less than the uncertainty of the energy axis.

## III. THEORY

The presently employed theoretical model is described in detail in Ref. [11]. In brief, the model treats the TS1 process as a sequence of the single photoionization of the primary target ($Ar^+$ in the present case) followed by the electron-impact ionization of the residual ion ($Ar^{2+}$ in the case under consideration). The approximation is made that the energy is conserved between these two stages of the TS1 process. A complete theoretical description should, in principle, include the virtual excitation and ionization processes that do not conserve the energy in the intermediate state. Such a theoretical description can be achieved for He-like targets [13], lithium [25], and alkaline-earth metal atoms [26]. However, it is not possible at present for many-electron targets such as Ar or $Ar^+$. For the latter targets, a perturbation theory is employed leaving out many-electron correlation processes. In fact, the PDI of the valence shell of Ar was one of the first processes of this kind to be considered theoretically [27,28]. Significantly later, a similar lowest-order perturbation theory (LOPT) treatment was extended to the Ar $L$ shell [29].

The present theory goes beyond the LOPT because many-electron correlations are taken into account both in the single-ionization and the electron-impact ionization stages, but not between them. The PDI cross section is presented in the form

$$\sigma^{2+}(\omega) = \sigma^{+}(\omega)\frac{\sigma^{2+}(\omega)}{\sigma^{+}(\omega)}, \qquad (1)$$

where the ratio of the double-to-single–ionization cross sections is expressed via the inelastic-scattering phase $\mu_{J=1}$ of the photoelectron on the residual ion in the dipole channel [11,30]

$$\frac{\sigma^{2+}(\omega)}{\sigma^{+}(\omega)} = \mu_{J=1}(E) = \mathrm{Im}\Sigma_{J=1}(E). \qquad (2)$$

The latter is equal to the imaginary part of the single-electron Green's function in the same dipole scattering channel. The electron-impact ionization in the dipole singlet channel represents an absorption of a virtual photon which is utilized in the concept of "a poor-man's synchrotron" [31,32]. Importantly, the photoelectron energy is obtained by the energy conservation $E = \omega - I_p$, where $I_p$ is the ionization potential of the primary PDI target and $\omega$ is the photon energy.

The single-photoionization cross section is evaluated using the random-phase approximation with exchange (RPAE) [30]. The inelastic photoelectron scattering is calculated by solving the integral equation for the reducible self-energy part of the one-particle Green's function [33]. Both techniques include many-electron correlations. Numerical implementation of the RPAE and inelastic-scattering techniques is provided by the ATOM suite of programs [34].

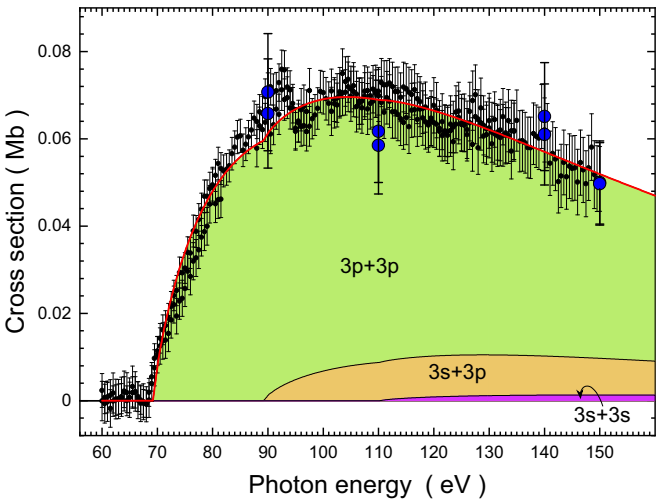


FIG. 2. Absolute cross section for direct double ionization of the ground-configuration $Ar^+$ ion by a singe photon measured at energy resolution 200 meV. The large solid circles with dark (blue) shading and large error bars are the results of absolute cross-section measurements with their total uncertainties. The solid circles with two-standard-deviation error bars were obtained by energy-scan measurements and then normalized to the absolute cross sections. The solid (red) line is the result of the present theoretical calculations shifted in energy by $-1.8$ eV. It is the sum of the partial cross sections for removing two electrons from the $M$ shell, both from the $3p$ subshell ($3p + 3p$), one from the $3p$ and one from the $3s$ subshell ($3s + 3p$), and both from the $3s$ subshell ($3s + 3s$). The individual accumulated contributions to the measured cross section are differently shaded (colored).

The present theoretical approach is applicable both to neutral and electrically charged atoms. The accuracy of the calculations is expected to be somewhat reduced in cases where electron correlations are particularly important, e.g., in processes involving the valence shells of negative ions or neutral atoms. A comparison of the present theory for valence-shell PDI of neutral Ar with experiments [35,36] shows quite reasonable agreement, with minor deficiencies in the low-energy region. For positive ions and especially with increasing ion charge states the outer-shell electrons are more strongly bound by the Coulomb field of the ionic core and the role of many-electron correlation decreases. This diminishes the effect of the virtual intermediate states that do not conserve energy and are neglected in the present model. As a result, the accuracy of the calculations for the $Ar^+$ ion should be quite satisfactory.

## IV. RESULTS

The measured absolute cross sections for single-photon direct double ionization of $Ar^+(3s^2 3p^5\ ^2P)$ are displayed in Fig. 2. The cross-section maximum of approximately 70 kb is reached at a photon energy of about 100 eV. In the investigated energy range extending from 60 to 150 eV, two electrons can be removed above the lowest threshold of 68.19 eV. Also from the NIST tables [24] one can conclude that a $3s$ plus a $3p$ electron can be removed above a minimum threshold of 82.8 eV. The minimum threshold for removing two $3s$

electrons from $Ar^+(3s^2 3p^5\ ^2P)$ has been estimated by the Cowan code [37] to be about 104 eV. Thus, in the investigated energy range all combinations of two electrons from the $3s$ and $3p$ subshells are energetically allowed to contribute to direct double ionization of $Ar^+$.

The individual cross sections for removal of two electrons from the $3p$ subshell ($3p+3p$), one from the $3p$ and one from the $3s$ subshell ($3s+3p$) and both from the $3s$ subshell ($3s+3s$) have been calculated using the method described in Sec. III. These contributions are summed to model the measured double-ionization cross section as shown in Fig. 2. The calculated threshold energies for these contributions are somewhat higher than the numbers resulting from the NIST tables. This is understandable since Hartree-Fock orbital energies are used in the calculation, so that the calculations overestimate threshold energies. In addition, the theory does not include fine-structure splittings. To match the onset of the experimental cross section at about 68 eV the calculated cross sections are shifted by 1.8 eV towards lower photon energies. With this shift, the sum of the calculated contributions is in very good agreement with the measured cross sections.

The ($3p+3p$) contribution (light green shading in Fig. 2) dominates the measured cross section by far. The combination ($3s+3p$) (light brown shading in Fig. 2) contributes 10% to 16% to the total cross section at energies between 100 and 150 eV while the removal of two electrons from the $3s$ subshell ($3s+3s$) (magenta shading in Fig. 2) provides an almost negligible contribution. It should be noted that two vacancies in the $3s$ subshell are not sufficient to energetically allow for the removal of one more electron from the intermediate $Ar^{3+}(3s^0 3p^5)$ ion by an Auger decay. Therefore, the calculated $3s+3s$ contribution fully contributes to the observed PDI cross section without an additional factor accounting for a branching ratio. The same is of course all the more true for the $3s+3p$ contribution with only one single $3s$ vacancy in the intermediate state.

## V. SUMMARY AND OUTLOOK

Absolute cross sections for direct double ionization of the $Ar^+$ valence shell by a single photon have been measured. The cross-section maximum is only about 70 kb, two and a half orders of magnitude smaller than the maximum of all nonresonant contributions to photo single ionization of the $Ar^+$ ion. The measurement of the relatively small cross sections was made possible by the high photon flux available at beamline 10.0.1.2 at the Advanced Light Source and the sensitivity of the photon-ion merged-beams apparatus employed in this experiment. Very good agreement with the experimental results is obtained by applying a recently developed theoretical approach in which the dominant two-step one (TS1) mechanism prevailing in the low-energy regime is described by a product of the cross section for direct single ionization and the probability for the ejection of a second electron by the subsequent $(e, 2e)$ half collision of the photoelectron with the ionic core. The theoretical method is promising to provide very useful results for atomic systems with many electrons where conventional methods such as time-dependent [38] and convergent [39] close-coupling calculations face problems with extremely high computational cost. It will be interesting to see how well the present theoretical approach can predict direct photon-induced double core-hole production, which has become an important subject in studies addressing molecular structures and dynamics.

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