Physical Mechanisms and Scaling Laws of K-Shell Double Photoionization


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We report on the photon energy dependence of the K-shell double photoionization (DPI) of Mg, Al, and Si. The DPI cross sections were derived from high-resolution measurements of x-ray spectra following the radiative decay of the K-shell double vacancy states. Our data evince the relative importance of the final-state electron-electron interaction to the DPI. By comparing the double-to-single K-shell photoionization cross-section ratios for neutral atoms with convergent close-coupling calculations for He-like ions, the effect of outer shell electrons on the K-shell DPI process is assessed. Universal scaling of the DPI cross sections with the effective nuclear charge for neutral atoms is revealed.

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Hollow K-shell atoms are atoms with an empty innermost shell and outer shells occupied. They may be created via K-shell double photoionization (DPI). Because single-photon absorption and two-electron ejection is induced by only one single interaction of the photon with the bound electron, K-shell DPI is one of the most sensitive probes of electron-electron correlations. Multielectron transitions were predicted in 1925 by Heisenberg [1]. Yet, in atomic structure calculations, an accurate theoretical treatment of electron correlations in many-electron systems is still a formidable challenge.

Because of a rather soft, in the perturbation sense, nature of the electron-photon interaction, the probability for creating double 1s vacancies by photon impact is quite low \((-10^{-2} - 10^{-6}\)). As a consequence, experimental data are scarce. With the advent of intense and energy tunable x-ray synchrotron sources hollow atoms have drawn renewed interest on both the experimental and theoretical side. In particular, the investigation of the photon energy dependence of the double 1s vacancy production became accessible. However, data for the double-to-single 1s photoionization cross section ratio \(P_{KK}\) well beyond the broad maxima are not available. The only exception is He [2]. Hitherto, for elements in the range 10 < Z < 20, only few experimental data have been reported. Measurements with conventional x-ray sources were performed for Mg [3] and Ar [4], and for Ne at a photon energy of 5 keV [5]. However, to the best of our knowledge, the photon energy dependence of \(P_{KK}\) had not been determined.

It is generally agreed that absorption of a single photon leads to double 1s ionization via two dominant mechanisms, namely, shakeoff (SO) and knockout (KO). In the SO process, the primary electron is ejected rapidly and a subsequent transition of the remaining electron to the continuum takes place due to a sudden change of the atomic potential. In the case of the electron inelastic scattering, the outgoing photoelectron knocks out the second 1s electron in an (e, 2e)-like electron impact half collision. Initial ground-state correlations are important in SO, while final-state electron interactions govern KO. The two mechanisms have very different photon energy dependences. The separation of the two mechanisms with photon energy is not straightforward and has been a subject of intensive research [6–12].

In this Letter, we present the first investigation of the photon energy dependence of the double K-shell ionization of Mg, Al, and Si. The relative contribution of the initial-state correlations and final-state electron-electron interactions to the K-shell DPI is addressed. We also raise the question as to the role of the outer shell electrons in the K-shell DPI. To this end, the K-shell DPI cross sections for hollow atom production are compared to those of their corresponding He-like counterparts. Universal scaling of the double photoionization cross sections with the effective nuclear charge for neutral atoms is examined.

The experiments were carried out at beam lines ID21 and ID26 at the European Synchrotron Radiation Facility (ESRF), using monochromatic and energy-tunable synchrotron radiation and the Fribourg von Hamos Bragg-type curved crystal x-ray spectrometer [13,14]. The double-to-single photoionization cross section ratios \(P_{KK}\) were deduced from the relative intensities of the resolved hypersatellite \((1s^{-2} \rightarrow 1s^{-1}2p^{-1})\) to the diagram \((1s^{-1} \rightarrow 2p^{-1})\) x-ray transitions. The energy resolution of the spectrometer was comparable to the natural line widths of the measured K x-ray transitions. The incident photon flux was \(\sim 1-3 \times 10^{12} \text{ph/s}\) at both beam lines.

The high-resolution K-hypersatellite x-ray emission spectra of Mg, Al, and Si are shown in Fig. 1. It can be seen that the Kα2 hypersatellite \((1P_1 \rightarrow 1S_0)\) is predomi-
intensities of the $K\alpha_{2}^{h}$ and $K\alpha$ x-ray emission lines employing the expressions: $P_{KK}^{max} = \frac{\omega_{KK}}{\omega_{K}}$ and $\sigma^{2+} = P_{KK}^{max} \sigma^{+}$, where $\omega_{K}$ and $\omega_{KK}$ are the fluorescence yields for the single- and double-hole states [17], respectively, and $\sigma^{+}$ stands for the single $K$-shell photoionization cross section from the NIST database [18].

For the He-like ions, the DPI cross sections were calculated using the convergent close-coupling (CCC) method. In the CCC calculations, the DPI is treated as a two-step process. The first is the full absorption of the photon energy by one electron. The second is the interaction of this electron with the nucleus and the remaining electron which results in the promotion of the remaining electron into the continuum. To describe the first step of single photoionization of Mg$^{10+}$, a 20-term Hylleraas ground state wave function due to Hart and Herzberg [19] was employed. For Al$^{11+}$ and Si$^{12+}$, more accurate results were obtained with a 9-term multiconfiguration Hartree-Fock ground state calculated using the computer code by Dyall et al. [20]. The final states with two electrons in the continuum were described by a close-coupling expansion on the basis of channel functions. Each channel function was a product of a Coulomb wave seeing the asymptotic charge of $Z - 1$, and a positive energy target pseudostate. The size of the basis as well as the number of the Coulomb waves were increased until convergence was achieved. More information on the photoionization CCC formalism can be found in Refs. [21,22].

The double-to-single $K$-shell photoionization ratios $P_{KK}^{max}$ in the peak region of the photon energy evolution were determined to be $2.03(19) \times 10^{-3}$ for Mg, $1.83(20) \times 10^{-3}$ for Al and $1.43(14) \times 10^{-3}$ for Si. Our experimental results are represented in Fig. 2 along with the data for other elements and He-like ions. The double-to-single photoionization cross-section ratios of Mg, Al, and Si are in good agreement with the data for other elements and He-like ions.

To determine the evolution of the double ionization probability with photon energy, the $K\alpha$ x-ray transitions were measured at different incident beam energies from 2.746 to 8.0 keV for Mg, from 3.122 to 7.0 keV for Al, and from 3.6 to 10.0 keV for Si. The ratios of the double-to-single $K$-shell ionization by photoabsorption $P_{KK}$ and the DPI cross sections $\sigma^{2+}$ were determined from the relative

![Graphs showing $K$-hypersatellite x-ray emission spectra of Mg, Al, and Si measured at photon energies in the region of the DPI cross sections maxima.](image-url)
For He-like ions, the nuclear charge incoherent summation of SO and KO terms. For two-electron targets adopting an approach based on an electron-impact ionization of a H-like ion, the single-ionization cross section by electron impact for a He-like ion, the incident electron energy, the ionization potential, and the double photoionization threshold. The KO contribution to the DPI ratio approaches a constant value corresponding to the photoabsorption asymptotic limit of shake-off [24,30]. For neutral Mg, Al, and Si the scaled $P_{KK}/P^\text{max}_{KK}$ values fall on one curve that coincides from threshold up to the maximum with the universal curve by Huotari et al. [12].

In this perspective, it is worthwhile to ask whether the scaling properties of DPI cross sections for two-electron systems, suggested by Kornberg and Miraglia [32], hold for neutral atoms. To this end, first power-law fits to the maximum values of $\sigma^2$ as a function of $Z'$ were performed. As expected, for He-like ions, a $Z'^{-4.08(3)}$ falloff was found; however, for neutral atoms a $Z'^{-3.68(11)}$ dependence was determined [see Fig. 4(b)]. A hint to the difference in scaling properties between two-electron and two-shell photoionization systems should scale with the square of the ionization potentials for a He-like ion and for the same neutral atom, respectively.

To corroborate the above statement, we have fitted the double-to-single $K$-shell ionization ratios for neutral and two-electron targets adopting an approach based on an incoherent summation of SO and KO terms. For $P_{KK}(E)$, we have used the SO expression of Thomas [29] and for $P^\text{SO}(E)$, the analytical form of the universal shape function for double-to-single $K$-shell ionization ratios for neutral atoms (open circles) compared to the CCC calculations (velocity gauge) for the corresponding He-like ions (closed circles) as a function of the scaled excess energy. The best fits to our data (see text) are represented by solid thick lines, the KO by thin solid lines, and the SO by dashed lines. Dotted-dashed lines show the KO terms of the He-like ions scaled by $Z'/Z^4$.

![Diagram](image)

**FIG. 2 (color online).** Double-to-single $K$-shell photoionization cross section ratios versus atomic number. ● present results, ○ [31], ■ [12, 33], △ [34], ▽ [11], × [23, 35], and nuclear electron capture (EC) [36]. For He-like ions, the $P_{KK}$ are from Ref. [22] and this work (open circles), and from Ref. [8] (crosses). The photoabsorption asymptotic limits of Forrey et al. [30] (dashed line) are also shown.

A clue to this finding resides in the proportionality of electron-impact ionization of a H-like ion to the KO term of the double photoionization of the corresponding He-like ion [8,9,26,27]. The KO contribution to the DPI ratio $P_{KK}(E - E^2)$ is proportional to $\sigma_s(E_s - I)$, where $\sigma_s$ is the single-ionization cross section by electron impact for a H-like ion, $E_s$ the incident electron energy, $I$ the ionization potential, and $E^2$ the double photoionization threshold. For the $(e, 2e)$-like electron-impact half collision following single 1s photoionization, the ionization potential corresponds to the binding energy of the remaining $K$-shell electron, $E'$. Using the hydrogenic formula $E' = Z^2$ Ry, where Ry = 13.6 eV, one can deduce the effective nuclear charge $Z'$. For the He-like targets, $Z' = Z$. Since along the hydrogen isoelectronic sequence a universal scaling $\sigma_s f^2 = f(E_s/I)$ works quite well [28], the relative strength of the KO contribution should scale with the square of the ionization potentials for a He-like ion and for the same neutral atom, respectively.

To corroborate the above statement, we have fitted the double-to-single $K$-shell ionization ratios for neutral and two-electron targets adopting an approach based on an incoherent summation of SO and KO terms. For $P_{KK}(E)$, we have used the SO expression of Thomas [29] and for $P^\text{SO}(E)$, the analytical form of the universal shape function for electron-impact ionization of H-like ions of Aichele et al. given by Eq. (5) in Ref. [28]. The $E'$ energies, calculated with the GRASP code [20], the asymptotic SO ratios from [30], and for He-like targets also the $E^2$ values of 3717.0 eV (Mg$^{10+}$), 4385.8 eV (Al$^{11+}$), and 5177.4 eV (Si$^{12+}$) were kept fixed in the fitting procedure. For Mg, Al, and Si, the fits yield $E^2$ values of 2741(35), 3189(23), and 3788(42) eV, respectively. The corresponding GRASP values of 2777, 3294, and 3880 eV are systematically higher, as reported for other elements [15,31]. The results of best fits to our data are shown in Fig. 3. Indeed, the KO terms of He-like ions multiplied by the $Z^2/Z^4$ ratios are close in magnitude to the ones for neutral atoms. Our results suggest that KO dominates near threshold and for intermediate excitation energies but becomes negligible at high energies where the $P_{KK}$ ratio approaches a constant value corresponding to the photoabsorption asymptotic limit of shake-off [24,30]. For neutral Mg, Al, and Si the scaled $P_{KK}/P^\text{max}_{KK}$ values fall on one curve that coincides from threshold up to the maximum with the universal curve by Huotari et al. [12].

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The effective nuclear charge. These results suggest that the range of applicability of the scaling exponents of $\sigma^{2+}$ as a function of $Z^\star$ for neutral atoms, within the experimental uncertainties, exhibit a universal scaling behavior. Further, when comparing the scaled $\sigma^{2+}Z^{3.68}$ for neutral atoms with the $\sigma^{2+}Z^{3.68}$ for the He isoelectronic series, one finds the curves to coincide.

In conclusion, the comparison of our results with other experimental data and theory allows us to separate contributions of the knock-out (KO) and shake-off (SO) mechanisms of the $K$-shell DPI process in Mg, Al, and Si. Our data support the conclusions of Kanter et al. [11] and Huotari et al. [12] for the predominance of KO at near threshold and intermediate photon energies. The effect of outer shell electrons on the double $K$-shell photoionization for neutral low-$Z$ atoms was found to be primarily reflected in the electron scattering contribution due to the change of the effective nuclear charge.

These results suggest that the post-photoabsorption electron-electron correlation is different for neutral atoms and He-like ions. A semiempirical universal scaling of the double photoionization cross sections with the effective nuclear charge for neutral atoms in the range $2 \leq Z \leq 47$ was established. Calculations of the DPI cross sections for neutral atoms will certainly shed new light on electron-electron interactions.

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