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 (CCC) 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.

Due to 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 ($\sim 10^{-2}$–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 knock-out (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}$ 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 $K\alpha_2^h$ hypersatellite ($^1P_1 \rightarrow {}^1S_0$) is predominant, the spin-flip $K\alpha_1^h$ transition ($^3P_1 \rightarrow {}^1S_0$) being dipole forbidden in the LS coupling scheme. In fact, the $K\alpha_1^h/K\alpha_2^h$ ratio probes the intermediacy of the cou-

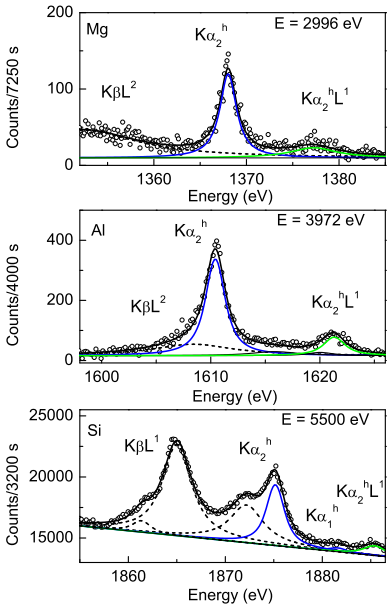

FIG. 1: (color online). K-hypersatellite x-ray emission spectra of Mg, Al and Si (blue solid lines) measured at photon energies in the region of the DPI cross sections maxima.

pling [15]. The $K\alpha_1^h$ could be only observed for Si and the experimental ratio of 0.03(1) was found to be consistent with the computed value of 0.0337 by Costa *et al.* [16]. The K-hypersatellite lines are accompanied by L-satellites $K\alpha_2^h L^1$ resulting from the presence of $2p$ and $2s$ vacancies during the x-ray transitions. In addition, on the low energy side of the $K\alpha_2^h$ x-ray lines the $K\beta L^2$ ($K^{-1}L^{-2}\rightarrow L^{-2}M^{-1}$) satellite transitions can be seen for Mg and Al, and the $K\beta L^1$ ($K^{-1}L^{-1}\rightarrow L^{-1}M^{-1}$) for Si, respectively. Spectral profiles of these multi-vacancy state transitions were determined experimentally by tuning the beam energy below the DPI thresholds. The intensities of the hypersatellites and diagram x-ray line were corrected for sample self-absorption and absorption of the incident x-rays, the photon flux, the spectrometer solid angle, as well as for the detector quantum efficiency.

To determine the evolution of the double ionization probability with photon energy, the $K\alpha^h$ x-ray transitions were measured at different incident beam energies from 2.746 keV to 8.0 keV for Mg, from 3.122 keV to 7.0 keV for Al, and from 3.6 keV 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 σ^{2+} were determined from the relative intensities of the $K\alpha_2^h$ and $K\alpha$ x-ray emission lines employing the expressions: $P_{KK} = \frac{I_{K\alpha^h}}{I_{K\alpha}} \frac{\omega_K}{\omega_{KK}}$ and $\sigma^{2+} = P_{KK}\sigma^+$, where ω_K and ω_{KK} are the fluorescence yields for the single- and double-hole states [17], respectively, and σ^+ 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 multi-configuration 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 $2.03(19)\times10^{-3}$ for Mg, $1.83(20)\times10^{-3}$ for Al and $1.43(14)\times10^{-3}$ 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 $1/Z^{1.6}$-dependence of the P_{KK}^{max} for neutral atoms [11, 29]. However, compared to the calculated P_{KK}^{max} values for two-electron targets of 1.29×10^{-3} for Mg^{10+}, 1.10×10^{-3} for Al^{11+} and 9.46×10^{-4} for Si^{12+}, they lie systematically higher. For

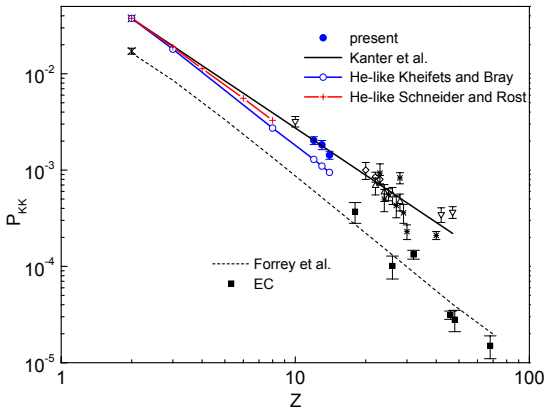

FIG. 2: (color online). Double-to-single K-shell photoionization cross-section ratios versus atomic number. • present results, ⋄[23], ∗[12, 24], △[25], ▽[11], ×[2, 26], and nuclear electron capture (EC) [27]. For He-like ions the P_{KK}^{max} are from Ref. [22] and this work (blue solid line), and from Ref. [8] (red solid line). The photoabsorption asymptotic limits of Forrey *et al.* [28] (dashed line) are also shown.

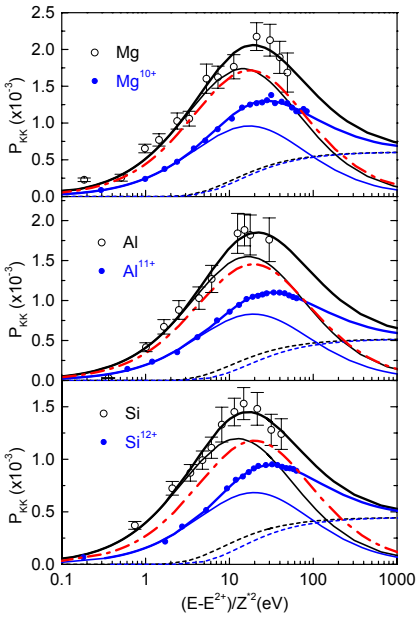

FIG. 3: (color online). Double-to-single K-shell photoionization ratios for neutral atoms (black open circles) compared to the CCC calculations (velocity gauge) for the corresponding He-like ions (blue closed circles) as a function of the scaled excess energy. The best fits to our data (see text) are represented by solid thick lines, the KO by thin solid lines, and the SO by dashed lines. Dot-dashed lines show the KO terms of the He-like ions scaled by Z^4/Z^{*4}. Black and blue lines correspond to the neutral atoms and ions, respectively.

the helium isoelectronic sequence the shake-off ratio in the high-energy limit can be calculated quite accurately provided that electron correlation is accounted for in the initial state wave function [30]. In a recent work on correlated wave functions for the K-shell electrons of neutral atoms and He-like ions [31] it was found that the influence of passive outer shell electrons on electron-electron correlation is very small. The outer shell electrons affect the electron-nucleus interaction but not the ground-state electron-electron interaction. Thus, it is plausible to assume that the photoabsorption asymptotic shake-off values for the He-like ions and neutral atoms are almost the same. Since the KO term predominates in the broad maximum of the P_{KK}, the observed higher values of P_{KK}^{max} for neutral atoms suggest that the electron scattering contribution to DPI is more important than for He-like ions.

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, 32, 33]. The KO contribution to the DPI ratio $P_{KK}^{KO}(E-E^{2+})$ is proportional to $\sigma_e(E_e-I)$, where σ_e is the single-ionization cross section by electron impact for a H-like ion, E_e 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 $\sigma_e I^2 = f(E_e/I)$ works quite well [34], 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 $P_{KK}^{SO}(E)$ we have used the SO expression of Thomas [35] and for $P_{KK}^{KO}(E)$ 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. [34]. The E^+ energies, calculated with the GRASP code [20], the asymptotic SO ratios from [28] 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) eV, 3189(23) eV and 3788(42) eV, respectively. The corresponding GRASP values of 2777 eV, 3294 eV and 3880 eV are systematically higher, as reported for other elements [15, 23]. 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^4/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 shakeoff [28, 30]. For neutral Mg, Al and Si the scaled P_{KK}/P_{KK}^{max} 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 [36], hold for neutral atoms. To this end, first power-law fits to the maximum values of σ^{2+} as a function of Z^* were performed. As expected, for He-like ions a $Z^{*-4.08(3)}$ fall-off was found, however, for neutral atoms a $Z^{*-3.68(11)}$ dependence was determined (see Fig. 4b). A hint to the difference for the scaling exponents of σ^{2+} is already given in the weaker Z dependence of P_{KK}^{max} for neutral atoms of $Z^{-1.61(5)}$ [11, 29]. Indeed, the scaled double photoionization cross sections for Mg, Al, and Si in reduced coordinates $\sigma^{2+}Z^{*3.68}$ against $(E-E^{2+})/Z^{*2}$, where Z^* stands for the effective nuclear charge, collapse onto a single curve (see Fig. 4a). To probe the range of applicability of the scaling, the scaled experimental DPI

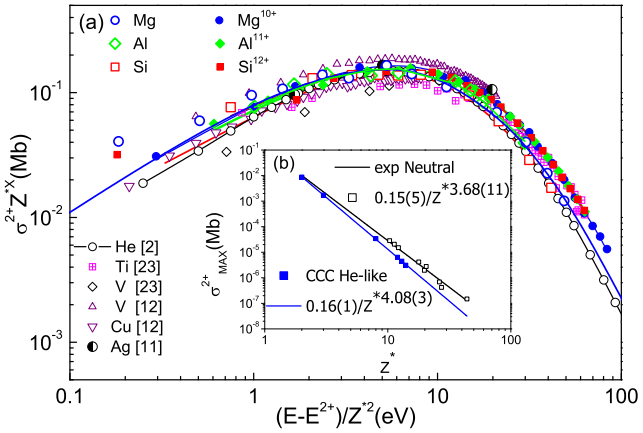

FIG. 4: (color online). (a) Scaled experimental DPI cross sections for Mg, Al, and Si compared to the scaled CCC calculations for He-like ions and experimental data for higher Z elements as a function of the scaled excess energy. A scaling exponent X of 4.08 along the He isoelectronic series and 3.68 for neutral atoms was used (see inset). The curves (solid lines) were deduced from the fitted values of P_{KK} shown in Fig. 3. (b) Shown are the power-law fits to the maximum values of σ^{2+} as a function of Z^*. The error bars are smaller than the symbols.

cross sections for He [2], Ti [23], V [12, 23], Cu [12] and Ag [11] are included on the same plot. The $\sigma^{2+}Z^{*3.68}$ data 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 $\sigma^{2+}Z^{*4}$ 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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