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Single- and double-photoionization data of Na and K corroborate the existence of a universal scaling law for the ratio

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Using monochromatized synchrotron light and the ion time-of-flight method to detect the singly and doubly charged sodium and potassium ions, we have investigated the relative double-photoionization cross-section ratios of Na from 52.4 to 150 eV and of K from 36.5 to 170 eV. We found that both followed a scaling law previously observed for Li and Be, which describes the photon energy dependence of that ratio and allows one to estimate its absolute value. We have also obtained the relative single- and double-photoionization cross sections for Na and K.

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I. INTRODUCTION

Single-photon ionization of atoms may lead not only to singly charged but also to doubly charged ions due to electron correlation among the electrons in an atom. The double-to-single photoionization ratio is a convenient way to quantify this correlation. While many experiments and theoretical investigations have dealt with the double photoionization of helium (see, e.g., [1–3]), the study of double-photoionization processes in heavier, simple elements, which are not noble gases, has been lagging behind. Although there are some investigations on double photoionization for heavier elements, their complexity (i.e., several double-ionization thresholds within a few eV) often does not make them attractive for an investigation attempting to understand the double-ionization process in which two electrons are emitted simultaneously.

In previous double-photoionization studies of lithium [4,5] and of beryllium [6,7] a scaling law for the double-to-single photoionization ratio of He, Li, Be, and possibly molecular hydrogen [10] has been found. At the time, it was hoped that this scaling law would allow us to predict the double-to-single photoionization ratios for other elements. However, it has been tested only for the emission of two s electrons, but not for electrons with an angular momentum larger than zero. The double-to-single photoionization ratios provided for sodium ($2p^6 3s$) by Rouvellou *et al.* [8] and Journel *et al.* [9] seem to indicate that this prediction is true, as mentioned in Ref. [10]. However, there was still some uncertainty due to the scatter in their values. There has also been research done on the single and double photoionization of potassium ($3p^6 4s$) [11,12]. However, those authors did not measure the ratio but the double-photoionization cross section which, unfortunately, does not go down to zero at threshold as it should. This lack of accuracy makes both the available Na and K data unsuitable for a rigorous test of the scaling law.

Therefore, we have performed precise measurements of the double-to-single photoionization cross-section ratios of K and Na in order to extend the test of the scaling law. The ratio for K was measured from 36.5 eV, which is near the double-ionization threshold (35.97 eV), to 170 eV photon energy. The ratio for Na was measured from its double-ionization threshold to higher photon energies, spanning the range of 52.4–150 eV. In this paper we compare the newly acquired double-to-single photoionization ratios of Na and K to those obtained in previous experiments and to the scaling-law model.

II. EXPERIMENT

A. Experimental setup

The Na and K experiments were performed on the 6 m Toroidal Grating Monochromator [13] at the Synchrotron Radiation Center (SRC) in Stoughton, WI (USA). The beamline has an energy range of 8–200 eV having an energy resolution of 0.13 eV at 65 eV for 200 μm entrance and exit slits. Second-order light was present at the lower energies, which we suppressed with appropriate filters. We employed an Al filter for the 36.5–70 eV range and a Si_3N_4 filter for the 70–100 eV range. The experiments covered only the region below 170 eV, which is well below the maximum photon energy of the beamline, thus avoiding any stray (i.e., not monochromatized) light contribution in the experiments.

As Fig. 1 shows, the monochromatized photon beam passed through a differential pumping stage which holds our filter arrays. A capillary connected the differential pumping stage and the main chamber and was aligned so that the photon beam came through it with a minimal photocurrent on the capillary (typically less than 100 pA).

The photon beam entered the interaction region in the main chamber, where it photoionized either the Na or K atoms produced by a resistively heated oven. The Na was heated to a temperature of about 190 °C while the K was heated only to about 140 °C. A pulsed electric field accelerated the photoions toward a drift tube with a Z stack of microchannel plates (MCPs) at its end [14]. The electric field was applied for 1.9 μs every 20 μs.

In Fig. 2 we display a typical sodium time-of-flight spectrum demonstrating a low background and undisturbed ion

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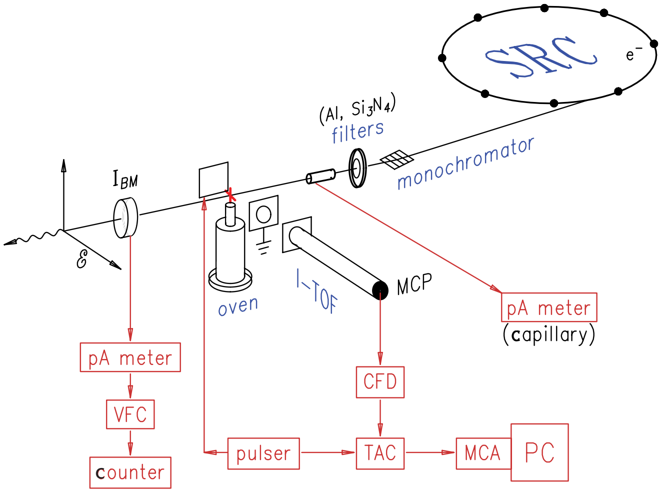

FIG. 1. (Color online) Sketch of the experimental setup (VFC: voltage-to-frequency converter, CFD: constant-fraction discriminator, TAC: time-to-amplitude converter, MCA: multi-channel analyzer, I-TOF: ion time-of-flight spectrometer, MCP: microchannel plate).

peaks. The countrate was about 2000–3000 Hz in the singly charged ion peak with a signal-to-noise ratio of about 32. Employing an ion extraction pulse rate of 50 000 Hz our count rate was low enough not to have dead-time problems of the electronic. The dead time was monitored throughout the experiments as it was displayed by the data acquisition program.

B. Experimental parameters

The Na and K used in the experiment were 99.5% pure and originally packaged in a glass ampoule under argon. They were handled inside a helium-filled glovebag to avoid reactions with the ambient air. The filled crucibles were transferred directly from the glovebag to the helium-filled chamber, typically being exposed to open air for less than a couple of minutes. The background pressure inside the experimental chamber during the experiment never exceeded 1×10^{-8} mbar during our measurements, and was typically in the $(3-7)\times10^{-9}$ mbar range. The contaminants in the spectrum, such as molecular nitrogen, water, and their fragments, had peaks that were well separated from the Na or K peaks.

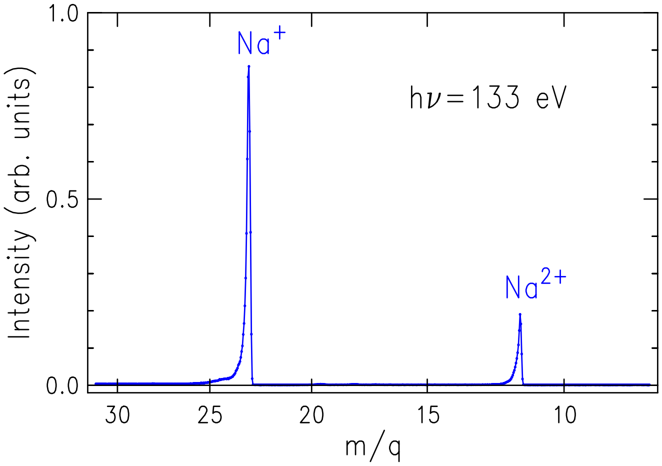

FIG. 2. (Color online) Sodium time-of-flight spectrum taken at 133 eV photon energy showing the singly and doubly charged ion peaks. Note that the abcissa shows the mass-to-charge ratio on a nonlinear scale.

The measurements of individual spectra lasted for 30 min in the threshold region of K, and only 10 min at the highest energies, where the double-photoionization signal strength was higher. The measurements for Na were typically 15 min long at all energies, except very close to threshold, where they lasted for up to 1 h.

The preamplified (×10) MCP pulse was processed by a constant-fraction discriminator (CFD) with its threshold set to 28 mV, its lowest value. The signal of a singly charged ion is on the average weaker than that of a doubly charged ion and, thus, is more discriminated against if the CFD threshold is too high.

In order to measure the correct ratio, we varied our MCP voltage from 2700 to 3800 V to find the voltage where the ratio became constant and singly and doubly ionized atoms are equally well detected. We used a MCP voltage of 3300 V throughout our Na and K experiments.

III. DATA ANALYSIS

We extracted the areas of the singly and doubly charged ion peaks in our time-of-flight spectra using direct numerical integration or an asymmetric Voigt profile fit. At most energies, the spectra were taken repeatedly and only the averaged value of the measurements at each photon energy is shown.

Helium, neon, argon, and xenon gases were used to calibrate the beamline's photon energy. They were kept at a steady pressure via a regulator and needle valve. We found that a photon energy correction was not necessary because the beamline was accurate within its energy resolution. We tested the energy calibration of the beamline by measuring the ion signal while scanning the monochromator across the Ar $3s\rightarrow4p$ resonance at 26.606 eV [15], the Ne $2s\rightarrow3p$ resonance at 45.5442 eV [16], the He $1s^2\rightarrow2s2p$ resonance at 60.1503 eV [17], and the Xe $4d\rightarrow6p$ resonance at 65.110 eV [18], which are shown in Fig. 3. For the He, Ne, and Ar resonances we selected the singly charged ion signal whereas for the Xe resonance we selected the doubly charged ion signal for determining the resonance position.

Helium was used during the Na and K experiments in order to derive the respective partial Na^+ and K^+ cross sections by using the area of the corresponding singly charged ion peak $S(h\nu)$ at a photon energy $h\nu$, the area of the singly charged He peak $H(h\nu)$ of the same spectrum, and the nonresonant part of the He cross section [19] $R(h\nu)$. We normalized our relative Na cross section by a factor N to fit our data to the data given in Refs. [8,20] near the Na $2s\rightarrow3p$ resonance at 66.372 eV. The K data were left unchanged, i.e., $N=1$. The formula

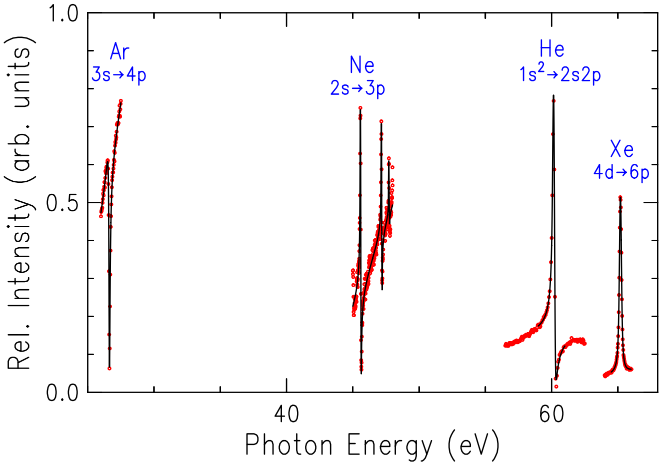

FIG. 3. (Color online) Ar, Ne, He, and Xe resonances used for the energy calibration of the beamline (dots) along with their Fano profile fit curves (solid line).

$$\sigma^{+}(h\nu) = N\frac{S(h\nu)}{H(h\nu)}R(h\nu) \tag{1}$$

was used for the derivation of the relative cross section for singly ionized Na and K assuming that the densities of He and the metal vapor are constant over time.

The double-photoionization cross sections of Na and K, σ^{2+}, were calculated using the area of the double-ionization peak $D(h\nu)$, the area of the single-ionization peak $S(h\nu)$, and the single-ionization cross section σ^{+} employing the formula

$$\sigma^{2+}(h\nu) = \frac{D(h\nu)}{S(h\nu)}\sigma^{+}(h\nu). \tag{2}$$

IV. RESULTS AND DISCUSSION

A. Sodium

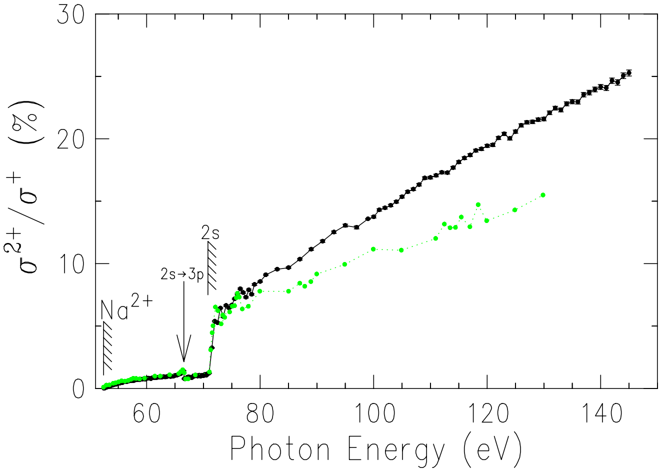

FIG. 4. (Color online) Double-to-single photoionization cross-section ratio of Na. Our data are connected by a solid line and the data from Ref. [8] are connected by a dotted line.

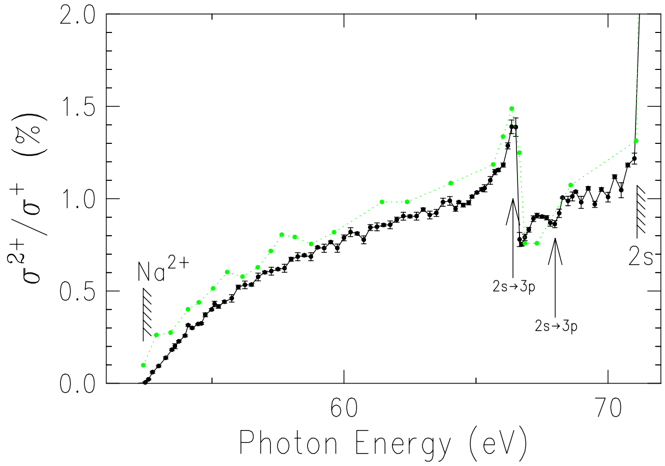

FIG. 5. (Color online) Near-threshold region of the double-to-single photoionization cross-section ratio of Na. Our data are connected by a solid line and the data (gray) connected by a dotted line are from Ref. [8]. The main features are the two $2s \rightarrow 3p\ ^2P$ resonances at about 66.4 and 67.8 eV, respectively.

Figure 4 shows our double-photoionization cross section data, along with those of Rouvellou *et al.* [8]. The two sets show good qualitative agreement with the major features, such as the Na^{2+} threshold, the $2s \rightarrow 3p$ transition, and the $2s$ threshold, matching very well. The higher values of the double-to-single photoionization cross-section ratio at the lower photon energies of Rouvellou *et al.* [8] could possibly be the result of second-order light, making the double-ionization cross section appear bigger. Similarly, the lower ratios of Rouvellou *et al.* at higher photon energies could be the result of stray light affecting the data near a grating's upper limit. Stray light typically favors single ionization, causing an overall reduction of the ratio.

The ratio exhibits a sharp rise starting at the double-ionization threshold at 52.39 eV with its slope becoming more gradual as the energies get into the 60 eV region. It is strongly affected by the $2s \rightarrow 3p$ resonance at about 66.4 eV, and the one at around 67.8 eV. The ratio reaches about 1% just below the $2s$ threshold at 71.08 eV; then it shoots up with a very steep slope, and continues to rise at a fairly constant rate to 25% at 145 eV, with a few minor excursions right above the $2s$ threshold. Figure 5 shows the low-energy region of Fig. 4 with the $2s \rightarrow 3p$ resonances shown in greater detail. Note that our Na^{2+} data go to zero at the double-ionization threshold indicating the absence of second-order light in our experiment.

We determined the double- and single-photoionization cross sections of Na as described in Sec. III, which are shown in Fig. 6. In order to put the data on an absolute cross section scale, we scaled our *total* cross section, comprising the single- and double-photoionization cross sections, to the data of Codling *et al.* [20] at 74 eV. Figure 7 shows the resulting data along with the data of other authors. From the total cross section we extracted the single- and double-ionization cross sections shown in Fig. 6 and in Fig. 8 for the $2s \rightarrow 3p$ resonance region.

For a quantitative analysis of the $2s \rightarrow 3p$ resonance we have applied the Fano formula [21] with an additional slowly varying background σ_b to our data

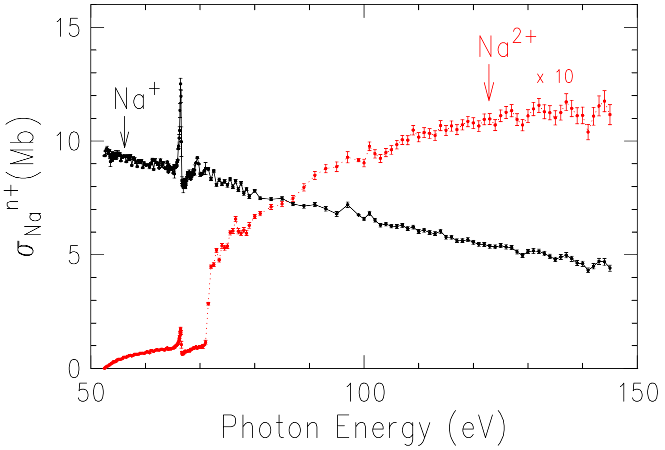

FIG. 6. (Color online) Double- and single-photoionization cross sections of Na. The data of the single-ionization cross section are connected by a solid line, while the double-ionization cross section data are connected by a dotted line and multiplied by a factor of 10.

$$\sigma = \sigma_a \frac{(q+\epsilon)^2}{1+\epsilon^2} + \sigma_b, \tag{3}$$

where $\epsilon = 2(E - E_0)/\Gamma$. Here, q is Fano's profile parameter, E is the photon energy, E_0 is the energy position of the resonance, and Γ is the resonance width. σ_a represents the part of the continuum cross section that interacts with the discrete level. In order to fit a Fano profile to the resonance, Eq. (3) was convoluted with a Gaussian band pass of 0.13 eV. The nearby Xe $4d \rightarrow 6p$ resonance at 65.11 eV allowed us to correctly calibrate the monochromator band pass for the Na resonance at 66.48 eV. Listed in Table I are our Fano fit parameters compared to those presented in Refs. [9,20]. The value for the $2s \rightarrow 3p$ resonance accepted by the National Institute of Standards and Technology (NIST) [22] is 66.372 eV, which is closer to our value than to that of Journel *et al.* Furthermore, while the Fano fit parameters derived by Journel *et al.* were done with a monochromator band pass of 0.31 eV, our band pass was only 0.13 eV. It should further be mentioned that the Γ provided by Codling *et al.* [20] has been derived assuming a q value of −2.0, which may, considering their error bar, give an incorrect Γ value.

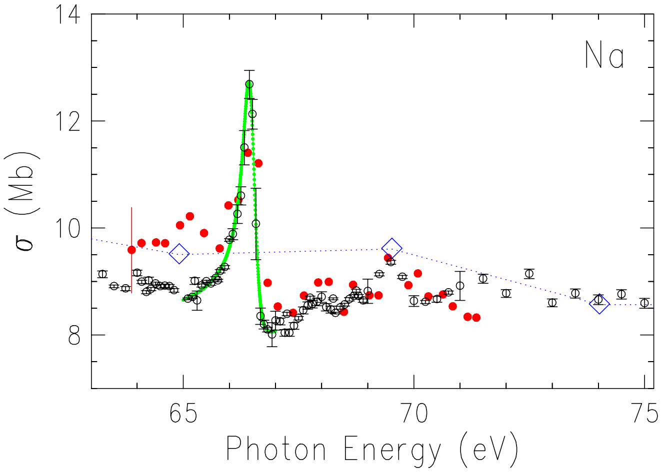

FIG. 7. (Color online) Total cross section of Na around the $2s \rightarrow 3p$ transitions. Our data, open circles; Journel *et al.* [9], filled circles (one data point is shown with a typical error bar); Codling *et al.* [20], open diamonds. The Fano profile fit to our data is shown as a gray solid line.

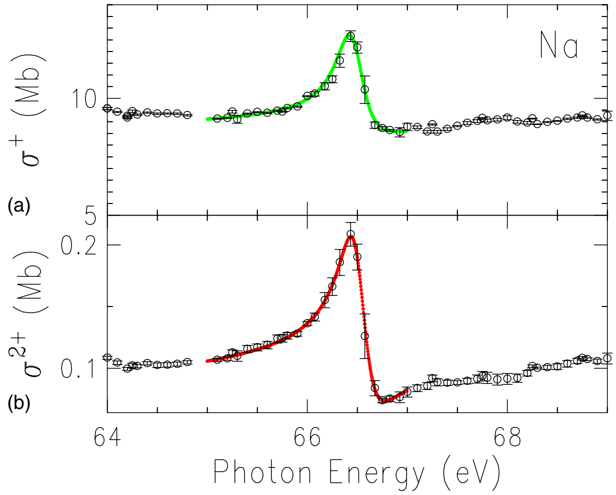

FIG. 8. (Color online) (a) Single-ionization and (b) double-ionization cross sections of Na around the $2s \rightarrow 3p\ ^2P$ resonance, with their corresponding Fano profile fits (solid lines).

B. Potassium

In Fig. 9 the double-to-single photoionization cross-section ratio of K is displayed from 36 to 170 eV. The ratio rises slowly to ≈0.5% as the energy increases toward the $3s$ threshold that appears at 40.6 eV. Then the ratio spikes upward to ≈3.5% followed by a decrease with a gradual slope up to about 46 eV, and has again a sharp rise in the 46–65 eV region, after which it essentially levels off to a plateau at about 30% all the way to 170 eV.

The low-energy region with a clearly visible $3s \rightarrow 5p\ ^2P$ resonance, displayed in Fig. 10, shows our data and the data extracted from the paper by Koide *et al.* [11]. Their ratio was calculated from the double- and single-photoionization cross sections after subtracting a constant value from their double-ionization cross section. The resulting ratio is in qualitative agreement with our ratio, however, there is an unexplained

TABLE I. Fano parameters for the $2s \rightarrow 3p\ ^2P$ resonance in the total Na cross section together with the values of Journel *et al.* [9] and of Codling *et al.* [20]. Here, E_0 is the resonance position, Γ the resonance width, q the Fano parameter, and σ_a the nonresonant cross section [see Eq. (3)].

Parameter	This work	[9]	[20]
E_0 (eV)	66.48(1)	66.64(2)	
Γ (eV)	0.28(2)	0.23(1)	0.34
q	−3.0(1)	−1.73(4)	−2.0(5)
σ_a (Mb)	6.13(6)	9.6(6)	

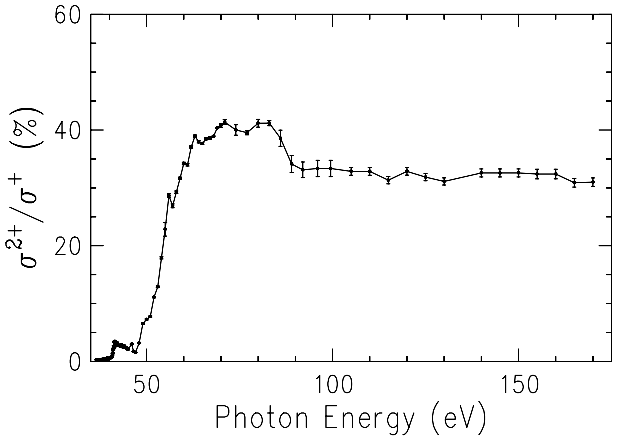

FIG. 9. Double-to-single photoionization cross-section ratio for K from 36.5 to 170 eV.

energy offset in the data by Koide *et al.* which could be the result of an improper energy calibration of their data. Because of that energy offset, we see the $3s \rightarrow 5p$ resonance at 39.3 eV, whereas Koide *et al.* data shows the resonance at 38.9 eV. Unfortunately, there seem to be no other data available to compare to.

Figures 11 and 12 show our relative single- and double-photoionization cross sections of K from 36.5–70 eV. A comparison of our data with those presented by Koide *et al.* shows qualitative agreement but, as mentioned above, there is an energy and cross-section offset between the two sets of data. It should be noted that the resonances seem to be more pronounced and visible in the single-photoionization cross section than in either the double-photoionization cross section or the ratio. The $3s \rightarrow 4p\ ^4P$ resonance appears at 36.7(2) eV, the $3s \rightarrow 4p\ ^2P$ resonance appears at 37.7(2) eV, and the $3s \rightarrow 5p\ ^4P$ resonance appears at 39.3(2) eV. The last resonance is also clearly visible in the double-ionization cross section, though it is not as obvious.

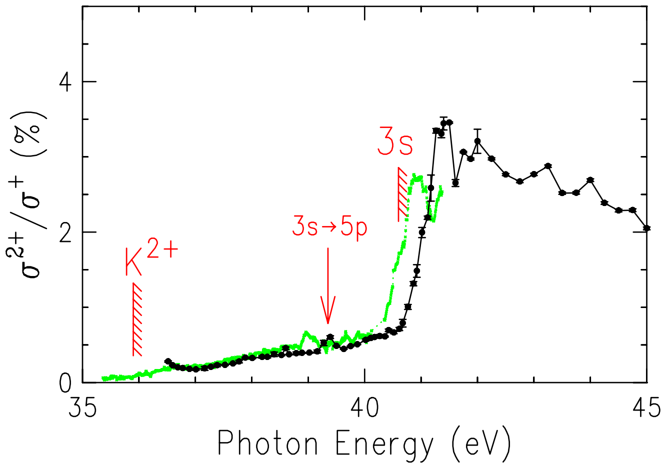

FIG. 10. (Color online) Double-to-single photoionization cross-section ratio for K in the near-threshold region. The data (black) connected by a solid line represent our results while the dots (gray) represent the data from Koide *et al.* [11].

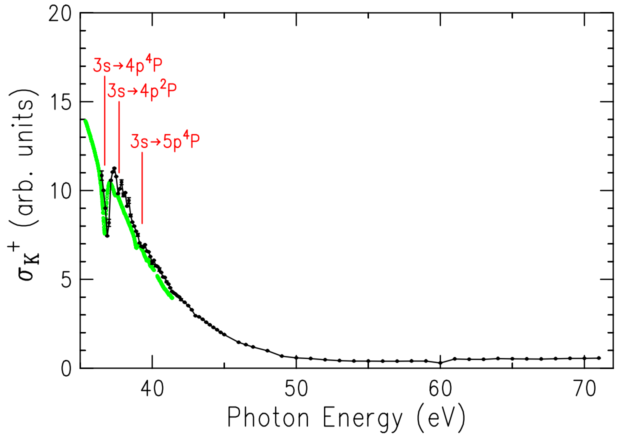

FIG. 11. (Color online) Single-photoionization cross section of K. Our data are represented by black dots connected with a solid line, while the data of Koide *et al.* [11] are represented by gray dots. The resonances visible in the cross section are indicated.

C. Scaling law

In order to test the scaling law that has been found to be valid for He, Li, and Be, we put our Na and K ratio data on an energy axis in units of ΔE [6,10] with

$$\Delta E = E^{2+} - E^{+} \quad (4)$$

where E^{2+} and E^{+} are the ionization thresholds for double and single ionization, respectively. If the two electrons originate from two different shells, the ionization threshold of the more strongly bound electron is used, possibly because the stronger bond electron has typically a larger cross section. The maximum nonresonant double-to-single photoionization ratio $R_{\max}$ for a given double-ionization process, can be expressed according to the scaling law [10] by

$$R_{\max} = c(\sqrt{E^{2+}} - \sqrt{E^{+}}) \quad (5)$$

where the constant $c \approx 0.01\ \text{eV}^{-0.5}$, and is assumed to be the same for all elements.

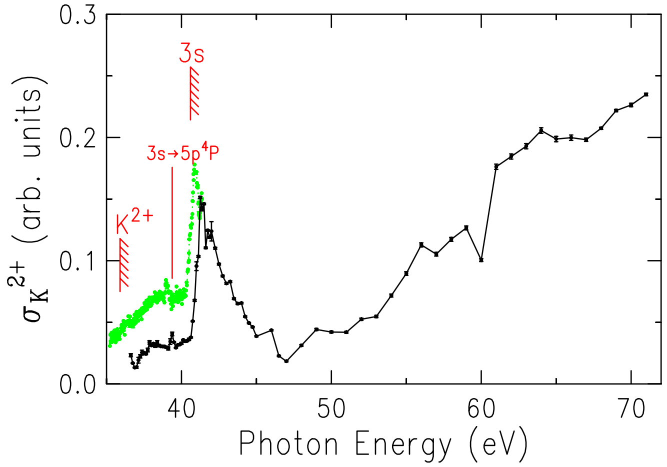

FIG. 12. (Color online) Double-photoionization cross section of K. Our data are represented by black dots connected by a solid line, while the data of Koide *et al.* [11] are represented by gray dots. The resonance and thresholds visible in the cross section are indicated.

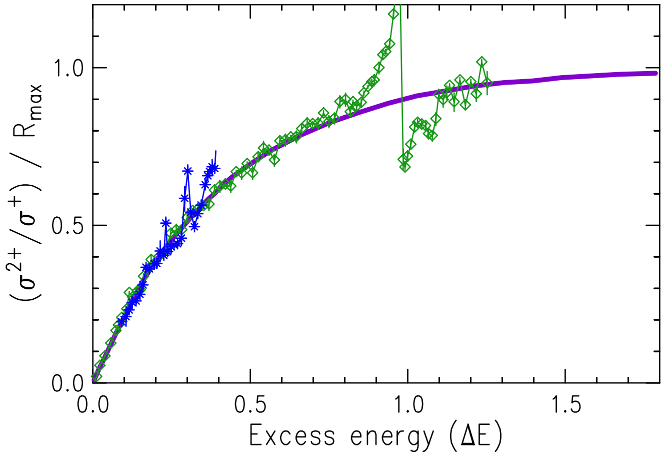

FIG. 13. (Color online) Our Na (open diamonds) and K (asterisks) ratios and the helium ratio curve of Samson *et al.* [23] (solid line) all scaled by a factor $R_{\max}$ so that the maximal ratios do not exceed 1.0. The energy axis is in units of $\Delta E = E^{2+} - E^{+}$ where E^{2+} and E^{+} are the thresholds for double and single ionization, respectively.

Note that, as far as we know, $R_{\max}$ is the maximum of the ratio of the double-ionization process that involves only the two outermost electrons in an atom. And, of course, autoionization and Auger decay processes are not included in this model. This means we have to restrict the test of the model to the energy region below the first inner-shell threshold that allows Auger decay. More information about the model can be found in Refs. [6,10].

Indeed, as Fig. 13 demonstrates, the scaling law, which has been demonstrated to work well for He, Li, Be, and possibly molecular hydrogen [6,10], can also be applied to Na and K. Interestingly, the double-ionization process of interest here involves a *p* and an *s* electron—in contrast to previous cases where two *s* electrons were involved. Obviously, the angular momenta of the ejected electrons do not make a difference in the scaling law. As Table II shows, the values of $R_{\max}$ predicted by the model are close to our experimental results, more so for Na than for K. Because the maximum ratio is reached only at a photon energy where Auger processes take place, we have fitted the Li, Be, Na, and K data to the He curve and so determined the values of $R_{\max}$. The error bars (except for He) are statistical error bars based on the fit to the He data, i.e., all ratio values have been used to determine $R_{\max}$ except for the resonance regions present in the Na and K data. The systematical error of $R_{\max}$ is estimated to be 0.05%. The shapes of the curves for Na and K conforms to the shape of the curve for He with the autoionization resonances clearly disturbing the picture.

TABLE II. $R_{\max}$ predicted by the model presented in [10] along with the experimental values for He [23], Li [4], and Be [6] and our present values for Na and K. Except for He, the error bars are based on the statistical deviation of *all* ratio values from the He data.

Element	Model	Expt.
He	3.93%	3.76(8)%
Li	0.98%	1.081(2)%
Be	2.19%	2.14(1)%
Na	1.08%	1.117(2)%
K	1.03%	1.12(1)%

Outside the resonances, the Na data fit the curve almost perfectly all the way up to its 2*s* threshold where the scaling law is no longer applicable. The K data, on the other hand, do not fit the scaling law quite as well and exhibit the largest deviation from the model but seem to generally be in decent agreement up to its 3*s* threshold.

So far it is not clear to us why the scaling law works so well. Nevertheless, it indicates that the mechanism of the double-photoionization process is the same for different atoms. A possible explanation why our energy scaling works so well may be found in the similarity between electron-impact ionization and double photoionization [24] and in a scaling law for electron-impact ionization [25]. Samson [24] discovered that the double-to-total photoionization ratio of an atom is proportional to the electron-impact ionization cross section of the corresponding ion. Since the double-to-single photoionization ratio is small, the double-to-*total* photoionization ratio is approximately the same as the double-to-*single* photoionization ratio. Now, the single-ionization cross section σ_e by electron impact for the hydrogen isoelectronic sequence is a function of the incident electron energy E divided by the ionization potential I, i.e., $\sigma_e = F(E/I)$ [see Eq. (4) in [25]]. In the case of single ionization of a singly charged ion, the corresponding ionization potential is then the double-ionization potential (since we end up with a doubly charged ion) minus the single-ionization potential (since we have an already singly ionized target). As a result of combining the proportionality found by Samson and the scaling law for electron-impact ionization, one may expect some similarity between the double-to-single photoionization ratios of different targets if the energy axis is given in units of the energy difference between the double- and single-ionization thresholds. Although it seems surprising that it does not matter whether *s* or *p* electrons are involved, it can be understood in terms of the average location of the electron. Obviously, it is not important where the electron can be located (according to the orbital) but where it is located on the average, which is given by its binding energy, which, in turn, determines the energy scale.

This, however, does not explain the scaling of the maximal ratio. The square root of the first ionization threshold is proportional to $Z_{\text{eff}}/n_{\text{eff}}^2$ with Z_{eff} the effective nuclear charge and n_{eff} the effective principal quantum number. However, the meaning of an effective principal quantum number for the double-ionization threshold is unclear and theoretical investigations of this scaling law are called for.

V. CONCLUSION

We have measured the double-to-single photoionization cross-section ratio of Na in the 52.4–150 eV range and that of K in the 36.5–170 eV range. Furthermore, we have determined the relative single- and double-ionization cross sections of both elements. While our data are in qualitative agreement with previous measurements on Na [8] and K [11] they also reveal problems and inaccuracies in the earlier measurements. The improved accuracy of our data allows us now to test the applicability of a scaling law introduced in [6,10], finding that the ratios of both Na and K follow that scaling law in the appropriate energy regions. In contrast to previous tests of the scaling law, a *p* electron participates in the double ionization of Na and K. Overall, the scaling law seems to be very robust and can indeed be applied to a wide variety of elements. We hope that our universal scaling law stimulates theorists to join our efforts to find the underlying physics of this effect.

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