Double ionization of rare gases. II. Ion formation by photon impact*†

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Using synchrotron radiation and a new two-grating grazing-incidence monochromator specially designed to remove higher-order components, the relative intensities of double-photoionization transitions have been measured in helium, neon, and argon between 70 and 200 eV by ion spectrometry. Extensive tests of a new ion spectrometer were undertaken to eliminate any charge discrimination for the ions. These new results resolve discrepancies existing between previous experimental data and favor, in general, the earliest results of Carlson for photon energies higher than 100 eV. For helium and neon they are in good qualitative agreement with theoretical calculations that include electron-electron correlations. In particular they agree very well with the many-body perturbation-theory approach of Chang and Poe in the case of neon.

I. INTRODUCTION

The interaction of low-energy photons with an atom can lead to the simultaneous emission of two electrons. When the primary ionization process takes place in an *inner* shell, the sudden ejection of an inner electron causes a change in the effective charge experienced by electrons in the outer shells. The simultaneous emission of one outer electron can thus result from this variation of charge. This process, called electron shakeoff, has been widely studied,[1-5] and the experimental results have been shown to be in agreement with calculations based on the use of single-electron wave functions in the sudden approximation theory.[6,7]

When the absorption of a photon takes place in the *outer* shell of an atom, the change experienced by the electrons of this same shell is too small to account for the large observed probability of double ionization; it has been shown experimentally by several methods (multiple photoionization processes,[8-12] double Auger process[13-15]) that the intensity observed for multiple ionization is much larger than expected from the assumption of a single-particle model (e.g., Hartree-Fock) and that this is strongest for the electrons of the outermost shells. These multiple transitions are strongly influenced by electron correlations, only a part being covered by shakeoff transitions. The double-photoionization cross section is thus a direct criterion for the importance of electron correlations. In order to avoid the problems connected with the experimental determination of absolute values for the single- or double-photoionization cross sections, in most cases only the ratio R of doubly to singly charged ions has been determined. Here we are dealing with both relative and absolute values. In the case of charge spectroscopy, the single-ionization cross section can contain a remarkable amount of ionization and simultaneous excitation processes.

From a theoretical point of view, the multiple-photoionization process is a fundamental process and theory should include electron correlations. But, up to now, this has been done only in two cases: for He, where several different approaches were used (highly correlated Hylleraas-type wave functions,[16,17] many-body perturbation theory[18]), and for Ne, with the many-body perturbation theory.[19,20]

The existing experimental results for the ratio R of Carlson[8] and Van der Wiel and Wiebes[9] on multiple-photoionization processes in the outer shell of noble-gas atoms (He, Ne, Ar) disagreed systematically by almost 100%. Carlson used filtered x radiation from an x-ray tube as a photon source and a mass spectrometer for the charge analysis of the photoions. Van der Wiel simulated a photon process by measuring the energy loss ΔE of small-angle-scattered high-energy electrons in coincidence with the photoions which were analyzed by mass spectrometry. In the limit of zero momentum transfer, the energy loss ΔE corresponds to the absorption of a photon with energy $h\nu = \Delta E$. The relative abundances of multiply charged ions of Ne, Ar, and Kr has also been determined by Lightner *et al.*[10] using a balanced-filter technique to obtain monoenergetic x rays. Here the lowest photon energy was 280 eV. At this energy their value for Ne is close to that of Carlson. New measurements of the threshold behavior of the double-photoionization process up to 107 eV photon energy in Ne, Ar, Kr, and Xe with discrete uv lines by Samson and Haddad[12] could not lead to a definite conclusion, because they were limited

at 107 eV; besides, they found excellent agreement with the data of Van der Wiel and Wiebes in the case of neon, but not for argon.

Theoretical results on[16,17] He and[19,20] Ne favored the measurements of Carlson, but these theories made several approximations, such as selection of the velocity form for the matrix element[16,17,19,20] or neglect of the electron correlations in the final state.[16,17]

Under these circumstances it was desirable to carry out a new experiment with two essential features: the use of a continuous photon source associated with a monochromator delivering a monochromatized photon flux with well-defined energy and variable bandpass in the energy range from 40 to 200 eV, and the construction of a specially designed ion spectrometer which was intensively tested with respect to possible charge discriminations.

II. EXPERIMENTAL

A. Apparatus

In the energy range of interest, only the synchrotron radiation emitted in electron accelerators has the required property to provide a continuous and intense flux of photons with well-defined energy. Moreover, the radiation emitted by a storage ring is the best source for this type of experiment because of the very high stability of the flux and the position of the photon beam.

The synchrotron radiation emitted by the ACO storage ring was consequently used at the new laboratory Laboratoire pour l'Utilisation du Rayonnement Electromagnétique (LURE) recently created to use the radiation emitted by the Orsay storage rings.[21] The characteristics of the ACO radiation (maximum flux around 200 eV, lifetime of a 100-mA electron beam on the order of 20 h, stability of the orbit within 20 μm) were perfectly suited for this experiment.

This continuous radiation was monochromatized in a double-grating grazing-incidence spectromonochromator specially designed to furnish with synchrotron radiation a flux of photons free of the higher-order components diffracted by the gratings. Higher orders are greatly enhanced with synchrotron radiation because of the shape of the spectral distribution. In addition, in our experiment there would be no possibility of distinguishing in the ion spectrometer between ions produced by photons of different energies. The fundamental characteristics of the monochromator that allows the delivery of a photon flux free of higher-order components at the exit slit were established more than ten years ago by Jaeglé,[22] and have been utilized successfully in many experiments.[23-25] The basic characteristics of this monochromator have already been described,[26] and a detailed description of the apparatus will be given elsewhere[27]; only some aspects are listed here. Basically two features are used:

(i) The radiation diffracted by the grating is reflected by a mirror which selects a narrow-wavelength band out of the diffracted spectrum and makes it convergent onto a second diffraction grating. Both gratings work alternately in negative and positive order.

(ii) The incidence angle on the first grating can be changed continuously over a large angular range, which allows one to choose the best working conditions at a given wavelength range by properly adjusting the critical angle.

Figure 1 shows the principle of the monochromator. Figure 1(a) gives an example of the two-grating setting. A prefocusing mirror PM focuses the almost parallel light coming from the storage ring on the entrance slit S_1 of the monochromator. After diffraction or reflection on the first grating G_1, the intermediate mirror M, and the second grating G_2, the monochromatic light passes the exit slit S_2. All optical elements, except the prefocusing mirror, are mounted on a horizontal 50-cm-radius Rowland circle. Our experiment required the monochromatized radiation to remain fixed in position and direction as the photon energy was varied. Thus the conventional exit slit is replaced by a mirror–knife-edge combination of the Codling type.[28] The wavelength scan is accomplished by driving M and G_2 with stepping motors. Simultaneously, the exit slit is allowed to rotate through one-fourth of the angle that is traversed by the second grating. An electro-optical encoder mounted on the axis of the Rowland circle gives the value of the angles with a relative accuracy of 0.001°.

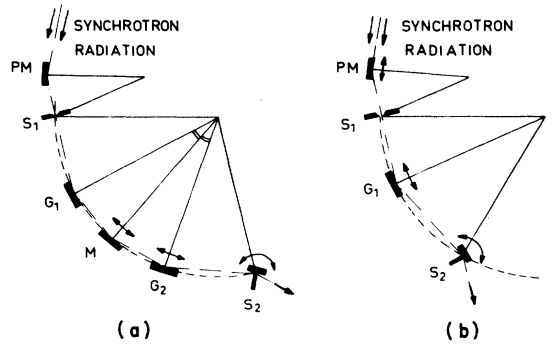

FIG. 1. Optical layout of the apparatus. (a) Two-grating setting: PM: prefocusing mirror; S_1 and S_2: entrance and exit slits; G_1 and G_2: gratings; dashed line: Rowland circle. (b) One-grating setting. In both cases the direction of the monochromatized radiation is kept fixed by rotating the mirror exit slit when the wavelength range is scanned.

Figure 1(b) shows a scheme of the monochromator when it is working in the one-grating mode, in the case where the suppression of higher-order components does not require the two-grating system. In this case the wavelength scan is accomplished by driving G_1 along the Rowland circle together with an automatic motion of the prefocusing mirror.

For the determination of the relative intensity of differently charged ions produced by photoionization, the ionization chamber and the ion spectrometer were connected to the exit slit of the monochromator. This apparatus, the operating procedure, and the checks concerning the problem of charge discrimination are described in the preceding paper.[29] In the photon-impact experiment, the replacement of the electron beam by a photon beam required only the following minor modifications: The photon beam was detected and monitored by a photomultiplier sensitized with sodium salicylate, and diaphragms with large openings and properly adjusted potentials prevented photoelectrons originating in the beam-defining diaphragm and the photon detector to enter the ionization chamber.

B. Operating procedure

Experimental setting

Only two different settings of the exit slit of the monochromator were necessary to cover the entire energy range. These two settings did not give, of course, the best working conditions for all wavelengths of the range used. However, they gave the possibility of covering the entire energy range of interest without the need of having to readjust the whole ion spectrometer too frequently.

In the low-energy range (70–125 eV) the two-grating system [Fig. 1(a): two 576-lines/mm platinum-coated gratings with a blaze angle of 1°, grazing angle on the first grating of 14°] was needed to remove higher-order components at the exit slit. Between 70 and 85 eV, the (−1, +2) and (−2, +1) components[30] did not strike the mirror M; the (−2, +2) component was suppressed by several reflections with grazing angles lower than the critical angles. Between 85 and 125 eV, all higher-order components were removed in this manner. An example of the direct spectrum recorded with the photomultiplier placed behind the ion chamber, at 62 cm from the exit slit, with two diaphragms in between, is shown in Fig. 2. At this position, the spectrum is an image of the shape of the flux of photons emerging from the ion chamber. The reduction factor of the photon flux due to the diaphragms introduced along the path of the monochromatized radiation was estimated to be around $\frac{1}{3}$ at the maximum of the spectrum. The direct spectrum at the exit slit of the monochromator, its interpretation as well as the testing procedure used to adjust the monochromator, will be described elsewhere.[31] Here we only note the importance of the blaze angle in giving the maximum intensity in the region of particular interest.

In the high-energy region (100–200 eV) the one-grating mode [Fig. 1(b): one 1200-lines/mm platinum-coated grating used in the first positive order with a blaze angle of 1°, grazing angle varying between 6.5° and 8°] was able to achieve the suppression of higher-order components by a proper choice of the incidence angle. This mode was chosen preferentially because of the higher photon flux delivered. In this setting, no monochromatized light was detected for wavelengths below 50 Å: the insertion of a carbon foil in the direct spectrum showed that the small signal detected by the photomultiplier was given by stray light. Consequently no higher-order component was present at photon energies higher than 125 eV. In order to avoid possible contamination by stray light at higher energies, all measurements in this energy range were carried out with a carbon foil inserted in the direct spectrum. An example of the direct spectrum recorded behind the ionization chamber is shown in Fig. 3, together with the spectrum transmitted through a carbon foil. These curves were recorded with a freshly platinum-coated grating. The influence of the critical angle is particularly evident. Other spectra recorded with gratings possibly covered with hydrocarbon layers showed a decrease of the output intensity

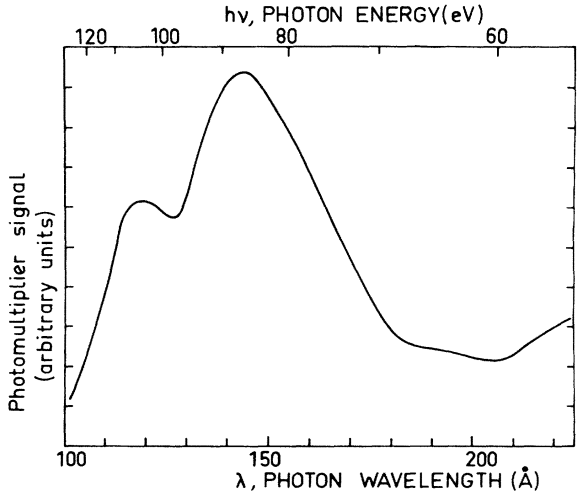

FIG. 2. Light output behind the ion chamber, recorded with the photomultiplier in the two-grating setting, with two 576-lines/mm platinum-coated gratings, as a function of the photon energy $h\nu$. Grazing angle on the first grating = 14°; angular position of the exit slit on the Rowland circle = 102° (0° = position of the entrance slit).

and a shift of the critical wavelength to higher wavelengths. These conditions achieved the suppression of higher-order components at photon energies even lower than 125 eV.

For both settings we determined the size of the photon beam in the middle of the ionization chamber with a Kodak SC5 film; the size is about 2 mm high and about 4 mm wide. It crosses the ionization chamber well within the limits for the region of the source volume giving no reduction for the transmission of the ions to the detector.[29]

Energy calibration

The relative energy scale, provided by the encoder, was adjusted to the absolute energy scale by performing an absorption experiment. Thin foils of Al and Si, about 1000 Å thick, were inserted into the photon beam and their mass absorption coefficient was measured, scanning the $L_{II,III}$ absorption edges. We adjusted the energy position of the edges to the values given for Al by Codling and Madden[32] [L_{II}: 73.15(2) eV; L_{III}: 72.72(2) eV] and for Si to the average between the values determined by Ershov and Lukirskii,[33] Gähwiller and Brown,[34] and Melchart *et al.*[35] [L_{II}: 99.7(1) eV; L_{III}: 100.3(1) eV]. Using these values we achieved the absolute calibration of the energy scale with an accuracy of 0.01°, which gave an uncertainty of 0.1–0.7 eV in photon energy, depending upon the energy range. No special effort was made to get a better accuracy.

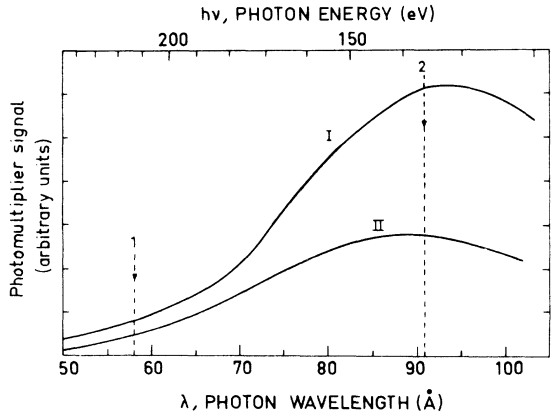

FIG. 3. Light output behind the ion chamber, recorded with the photomultiplier in the one-grating setting, with a 1200-lines/mm platinum-coated grating used in the first positive order, as a function of the photon energy $h\nu$. Curve I: direct spectrum; curve II: spectrum transmitted through a carbon foil. Angular position of the exit slit on the Rowland circle: 37°; vertical dashed lines: position where the wavelength is equal to the critical wavelength calculated with $\delta = 5.0\times10^{-6}$ (line 1) and $\delta = 2.5\times10^{-6}$ (line 2).

Photon flux

In all experiments, the ACO storage ring was operated at 536 MeV with a maximum circulating current of 100 mA. For each setting we measured at one photon energy the absolute flux of photons in two ways: (i) by comparing the number of singly charged ions produced in Ne with a known electron current in the electron gun[29] and with the unknown flux of photons, taking into account the different cross sections for photons[36] and electrons[37], and (ii) by measuring directly with an electrometer the current of photoelectrons emitted from a gold foil under photon impact, taking into account the photoelectric yield of the gold foil.[38] Both methods gave results in good agreement, that is, within 20%. In the two-grating mode, we measured a flux of 5×10^{6} photons sec^{-1} Å^{-1} at 170 Å (73 eV) and, in the one-grating mode, 5×10^{8} photons sec^{-1} Å^{-1} at 82 Å (150 eV) in the ionization chamber with the beam-defining diaphragms.

Widths of, typically, 50 μm were chosen for the exit slit of the monochromator, giving a bandpass of several eV; $\Delta E/E$ was generally kept between 3 and 4%, thus providing a higher photon flux.

Operating conditions of the ion spectrometer

These conditions were the same as described in the case of ion production by electron impact.[29] In particular, the target-gas pressure was kept low enough, in the 10^{-5}-Torr (10^{-3} N m^{-2}) region, to achieve a negligible dependence of the ratio R on the pressure. The entire ion peak, as detected by the channeltron, was scanned in each case and equal parts of the plateau were used for the evaluation of the data.

Measurements

In all cases the photon energy was too low to create ionization in any other but the outermost shells. In steps of 25 eV, we measured the ratio R of doubly to singly charged ions between 70 and 200 eV for neon and argon and between 100 and 150 eV for helium. The influence of the H_2^+ ion peak was found to be negligible in the case of helium. For each photon energy several sequences of counting (doubly-singly-doubly...) were performed.

Owing to the very low background of the channeltron (which was as low as 4 counts/min) and the high stability of the electron beam in the storage ring (in energy, intensity, and position), we were able to measure R with counting rates as low as 20 counts/sec for He^+, 5 counts/sec for Ne^+, and 4 counts/sec for Ar^+. Under the best conditions, the highest counting rate achieved was 380 counts/sec for Ne^+ at 150 eV with the carbon foil inserted

in the photon beam.

In the one-grating mode, it was possible to accumulate enough events to make the statistical error much lower than the intrinsic error inherent in the ion spectrometer (7%),[29] and the final error, determined by linear addition of both errors, was less than 9% in all cases. In the two-grating mode, the counting rate was, of course, much lower and, for the ratios determined with this setting, the final errors vary between 9 and 13%.

III. RESULTS AND DISCUSSION

The values measured for the relative intensities of double-ionization transitions (ratio R) are given in Table I for He and in Table II for Ne and Ar.

For He, all results were obtained in the one-grating mode and with larger bandpass, because of the lower cross section. For Ne and Ar, the results obtained with the two-grating setting in the low-energy region are listed in the upper part of the table. For one photon energy, in Ne at 125 eV, data were collected in both modes; both results are in agreement within the error limits. The value adopted for this energy is the weighted average of these two results.

Figures 4–6 show our results in comparison with earlier experimental results and theoretical calculations.

For He our results lie between those of Carlson[8] and Van der Wiel and Wiebes.[9] Byron and Joachain[16] obtained R, calculating the double-photoionization cross section and using theoretical values of Salpeter and Zaidi[39] and Stewart and Webb[40] for the corresponding single-ionization cross sections. The main part for processes of ionization and simultaneous excitation is included in this theoretical ratio.[41] These theoretical results are in qualitative agreement with our values, if the velocity formulation for the matrix element is used. The length formulation, as anticipated by Byron and Joachain, does not agree well with experiment. The fact that their theoretical curve in the velocity formulation seems to be more in favor of Carlson's results is not significant, because the neglect of correlations in the final state could be a simplification. The curve calculated by Amusia[18] is valid only for high photon energy. In the energy region of our measurements, Brown's curve[17] is rather close to Byron and Joachain's curve and was not plotted in Fig. 4.

Comparison of our data on Ne with Van der Wiel and Wiebes's data[9] shows that their values are significantly higher than ours and that this discrepancy is more and more pronounced with increas-

TABLE I. Relative intensities of double-ionization transitions in helium. Values are given in percent and relative to intensity of single-ionization transitions. Quoted errors are determined as explained in Sec. II. of the text. Data were collected in the one-grating mode.

Photon energy (eV)	Bandpass (eV)	He^{++}/He^{+}
100.0(2)	6	2.4(2)
125.0(3)	10	3.7(3)
150.0(4)	14	4.1(4)

TABLE II. Relative intensities of double-ionization transitions in neon and argon. Values are given in percent and relative to intensity of single-ionization transitions. Data collected with the two-grating system are listed in the upper part of the table. The bandpass is sometimes different for neon and argon owing to the differences in the cross sections.

Photon energy (eV)	Bandpass (eV)	Ne^{++}/Ne^{+}	Bandpass (eV)	Ar^{++}/Ar^{+}
73.0(1)	2	1.5(2)	2	18.1(2.0)
100.0(2)	3	5.4(5)		
115.0(3)			4	18.0(2.3)
125.0(3)	5	7.9(9) [a]		
125.0(3)	5	9.4(8) [a]	5	21.2(1.8)
150.0(4)	4	11.2(1.0)	7	19.0(1.6)
175.0(6)	6	12.6(1.1)	9	20.7(1.9)
200.0(7)	12	11.5(1.1)	12	19.1(1.7)

[a] The weighted value used in Fig. 5 is 8.9(8).

Comparison of our data on Ne with Van der Wiel and Wiebes's data[9] shows that their values are significantly higher than ours and that this discrepancy is more and more pronounced with increas-

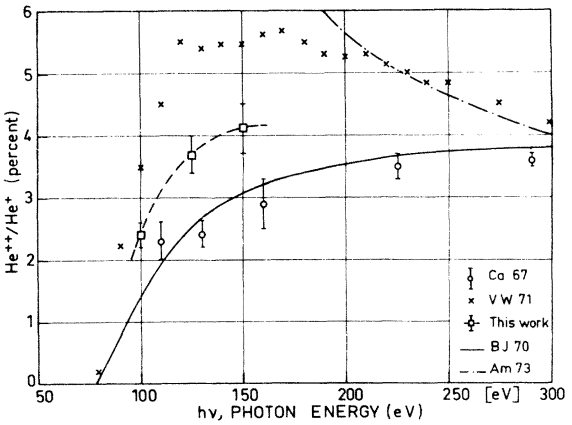

FIG. 4. Relative intensity of double-photoionization transitions in He as a function of the photon energy $h\nu$. Values are given in percent and refer to the intensity of single-ionization transitions. Earlier experimental values are from Refs. 8 (Ca 67) and 9 (VW 71). Theoretical results are from Refs. 16 (BJ 70, velocity form) and 18 (Am 73). As in Figs. 5–7 dashed line represents our choice of bridging the gap between the experimental points.

ing photon energy. On the other hand, our data agree well with Carlson's results in the high-energy part of the curve for photon energies higher than 100 eV. At lower energy, Carlson's data tend to be too high, as might be expected,[20] since his results average over the wide bandpass of his filtered photon source. Agreement with Samson and Haddad's results[12] is reasonably good, although the slope of their curve is somewhat different from the line we can draw through our points. The agreement with the theoretical curve calculated by Chang and Poe[20] is good; only the slope of their curve is slightly different. But the wave functions used in their treatment may not be well adapted for the description of the inelastic internal collision process. Therefore better wave functions could modify the results at lower photon energies.[42] Such a good agreement is obtained, although one has to consider the following points as given by Chang and Poe: First, their results for the single-photoionization cross section describe the experimental data only within 5–10%. Second, the contribution from the final ionic configuration $2s2p^5$ is neglected. We estimated the relative contribution of the $2s$ subshell to double photoionization by assuming that this relative contribution varies in the same way as the relative contribution of the $2s$ subshell to the simultaneous photoexcitation and ionization transitions (transitions with $2s2p^5 \epsilon l$, $n'l'$ final states compared with $2s^2 2p^4 \epsilon l$, $n'l'$ final states).[43] Using the values given in Ref. 43, we calculated that this contribution should vary from 5% at 100 eV to 10% at 200 eV. But this correction for the double-photoionization cross section, which would increase R, is partly compensated by a correction for the single-photoionization cross section. Owing to the influence of processes involving ionization and simultaneous excitation, about 3% at 100 eV up to about 6% at 200 eV (Ref. 43) should be added to the calculated single-ionization cross section. Third, although they took into account the physical effects that are expected to be most important (namely, ground-state electron-electron correlations, core rearrangement, virtual Auger transitions, and inelastic internal collision), they did not include other physical effects involving more than two orbital electrons or possible interactions between electrons of different shells. Finally, they employed the velocity formulation of the theory. Nevertheless, the good agreement with our experimental results shows that they used an approach well adapted to the investigation of multiple processes in photoionization.

For argon, there exists no theoretical calculation of the double-photoionization cross section. The comparison of our data with the various experimental results confirms the tendency observed in the case of neon: good agreement with Samson and Haddad at low energy and with Carlson at high energy, and a large discrepancy with Van der Wiel and Wiebes[9] over the entire energy range up to 200 eV.

Our results for the energy dependence of the relative double-photoionization cross section are shown in Fig. 7 on a normalized energy scale. This figure shows that while the general behavior is the same for the three gases (fast rise of the curve followed by a plateau), the details of the curves are quite different, namely, in regard to the initial slope of curve and the point where the plateau starts. It should be noted also that this plateau is reached at much lower photon energies than in the corresponding cases of simultaneous

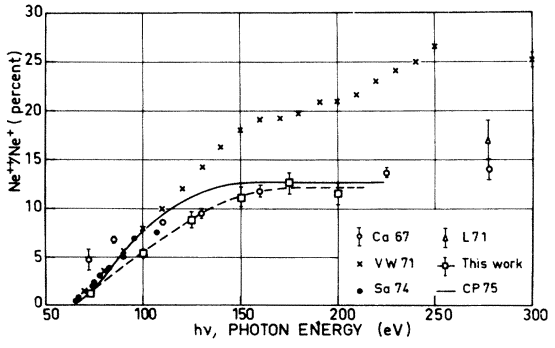

FIG. 5. Relative intensity of double-photoionization transitions in Ne as a function of the photon energy $h\nu$. Values are given in percent and refer to the intensity of single-photoionization transitions. Earlier experimental values are from Refs. 8 (Ca 67), 9 (VW 71), 10 (L 71), and 12 (Sa 74). Theoretical curve is from Ref. 20 (CP 75).

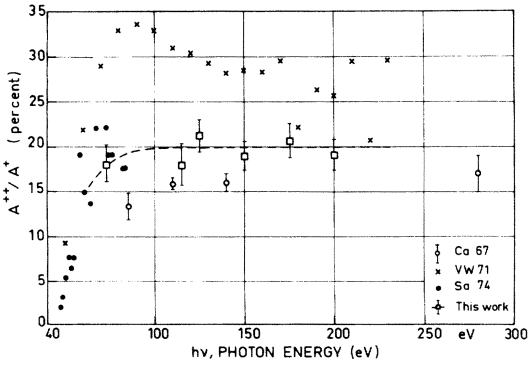

FIG. 6. Relative intensity of double-photoionization transitions in Ar as a function of the energy $h\nu$. Values are given in percent and refer to the intensity of single-photoionization transitions. Earlier experimental values are from Refs. 8 (Ca 67), 9 (VW 71), and 12 (Sa 74). There is no theoretical calculation for argon.

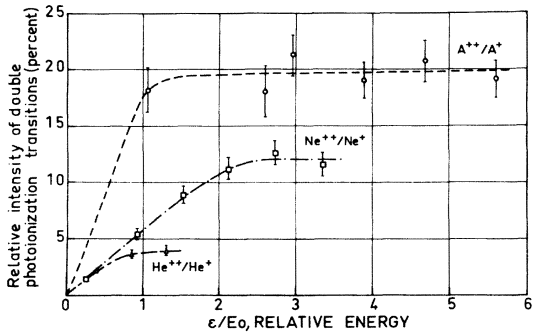

FIG. 7. Energy dependence of the relative intensity of double-photoionization transitions in He, Ne, and Ar as a function of the relative ionization energy ϵ/E_0. $\epsilon = h\nu - E^*$ is the excess energy of the photon $h\nu$ over the energy necessary for the double-ionization transitions; E_0 is the energy necessary to remove a second outer electron in the presence of one hole in the outer shell. Values are given in percent and refer to the intensity of single-photoionization transitions.

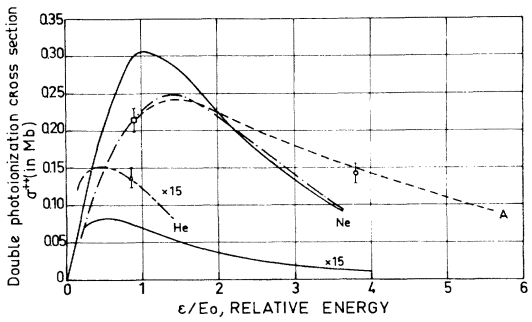

FIG. 8. Absolute values of the double-photoionization cross section in He, Ne, and Ar as a function of the relative ionization energy, defined as in Fig. 7. Dashed lines: experimental curves extracted from Fig. 7; the magnitude of the error is indicated on each curve, and the error bar does not include the error on total photoabsorption cross section. Full lines: theoretical curves for He (Ref. 16, velocity form) and Ne (Ref. 20).

photoexcitation and ionization in helium[44] and neon.[43]

But even on a normalized energy scale the variation of the relative double-photoionization cross section is not fully representative of the strength of electron correlations. Only the absolute value σ^{++} for double photoionization can give accurate information about electron correlations, since the different behavior of the curves representing the ratio R might be due to different variations of the single-photoionization cross sections as a function of the energy. Neglecting the triple photoionization in this energy range (threshold for triple ionization in Ne at 126.6 eV and in Ar at 84.3 eV), we determined σ^{++} using our R values and the total photoabsorption cross section as determined in other experiments.[45] The results are plotted in Fig. 8 as a function of the photon energy on the normalized energy scale, together with the theoretical curves for[16] He and[20] Ne. From this it can be seen that the double-photoionization cross section in He is much smaller than those for the outer-shell p (and s) electrons in Ne and Ar, which are of the same order of magnitude.

In He, the experimental and theoretical curves show rather a large discrepancy at lower energies but seem to agree better at higher energies. In Ne, there is still a small discrepancy at lower energies but theory and experiment agree very well at higher energy. Thus in general the many-body perturbation–theory approach of Chang and Poe seems to be able to describe correctly the *absolute* magnitude of the correlation effects.

During the initial phases of this work with synchrotron radiation, we had several discussions with Van der Wiel concerning the topic of multiple ionization and, especially, the discrepancy between his and our results for the ratio R in the case of electron impact (preceding paper). In the course of our experiments using the synchrotron radiation, Van der Wiel repeated his quasiphoton measurements in a different way and sent us his latest, revised values prior to publication.[46] There now exists gratifying agreement between his new results and our values for He and Ar over the entire energy range and for Ne up to 175 eV.

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