

Measurements of absolute Ar 3s photoionization cross sections

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Cross sections for the photoionization of the argon 3s electron are presented from threshold up to 100 eV. In order to cover this whole energy range with comparable accuracy, we used two complementary methods: absolute measurements by photon-induced fluorescence spectroscopy (PIFS) and relative measurements by time-of-flight (TOF) photoelectron spectroscopy (PES). After calibration of the relative PES data by the most recent absorption results, the data of these independent measurements agree well with each other. The experiments were carried out with special emphasis to the threshold region (PIFS) and the region of the cross sectional minimum (TOF) where, in addition to the partial cross sections, the angular distributions of the photoelectrons were determined. The results are compared with published data and discussed with respect to matrix-element dependencies and correlation effects.

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I. INTRODUCTION

The photoionization of the argon 3s electron has attracted a continuous interest over the last twenty years. Already in 1972 it was theoretically predicted [1], and in 1974 experimentally proved [2], that the collective response of the argon *M*-shell electrons gives rise to a drastic change in the energy dependence of the 3s photoionization cross section, compared to independent particle calculations which show no minimum above threshold [3]. The intershell interaction was proven to be the dominant correlation term shifting the cross sectional minimum, also known as a "Cooper" minimum, to higher energies. Many different methods for the calculation of electron correlations were subsequently developed. The argon 3s photoionization cross section became one of the prominent case studies in atomic photoionization [4–12]. Nevertheless, it took more than ten years until another kind of correlation, namely the influence of doubly excited states on the 3s cross section, was experimentally demonstrated [13–15]. A first, we believe, but not the complete calculation of this influence was given recently by Wijesundera and Kelly [10].

Besides this fundamental interest in the 3s photoionization cross section, as a demonstration of electron correlation effects, it was widely used for normalization of other experimental data in the field of photoionization. All cross sections for the production of satellite states [16] and for the $3s3p^5\ ^1,^3P$ double photoionization [17] are given in relation to the 3s cross section.

So far, the experimental Ar 3s photoionization cross sections were predominantly determined using photoelectron spectroscopy data (PES) being normalized to total absorption cross sections [18–23]. This method requires the capability to measure photoelectrons at very different energies, also including the low energy part of a spectrum

and thus a correct knowledge of the detection efficiency over a wide range of photoelectron energies.

In order to obtain photoelectron spectra at such widely varying kinetic energies, we used time-of-flight (TOF) photoelectron spectroscopy in our experiments (Fig. 1). As described below these were especially carried out in the region of the cross sectional minimum to test the different theoretical calculations (e.g., from Refs. [1,10,11]) in this region which are very sensitive to correlation effects. At higher excitation energies special efforts have been made to take the portion of the double ionization continua into account as correctly as possible. For normalization we used the total absorption cross sections of helium from West and Marr [24] and the accurate total absorption cross sections of argon, which were given

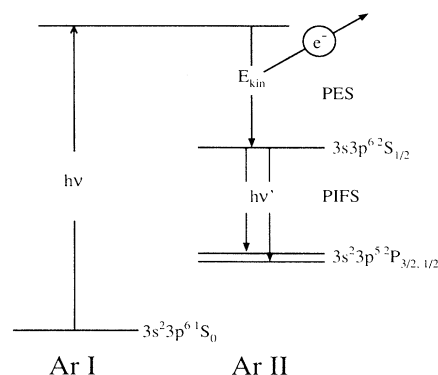


FIG. 1. Schematic Ar 3s photoionization process. The diagram shows the outgoing electron e^- which can be detected using photoelectron spectroscopy (PES) and the subsequent fluorescence decay. The emitted photons $h\nu'$ are measured using photon-induced fluorescence spectroscopy (PIFS).

very recently by Samson *et al.* [25] with an error as small as $\pm 3\%$.

In an alternative approach we used photon-induced fluorescence spectroscopy (PIFS). For the data presented here, we used a normalization to accurate emission cross sections for electron-impact induced line radiation. The results are absolute cross sections for the photoionization of the argon 3s electron, which are independent of any PES data or total absorption data. Moreover, PIFS overcomes one of the main problems of PES; the detection efficiency for the fluorescence photons of a given state is independent of the exciting photon energy. This advantage especially applies to the threshold region; therefore, the experiments based on PIFS were carried out with special emphasis to this region.

II. PIFS MEASUREMENTS

Using PIFS, the photoionization cross section σ of the argon 3s electron follows from the intensity of the $3s3p^6^2S_{1/2} \rightarrow 3s^23p^5^2P_{3/2,1/2}$ transitions ($h\nu = 92.0$ and 93.2 nm) as a function of the exciting photon energy ($h\nu$), as shown in Fig. 1.

The measurements were carried out at the toroidal-grating monochromator TGM-4 beam line at BESSY, Berlin. The experimental setup was described in detail elsewhere [17], and only a short description is given here. The monochromatized synchrotron radiation ($\Delta E \approx 90$ meV) passes through a differentially pumped target gas cell, where the argon atoms are ionized. The produced fluorescence radiation is dispersed by a vacuum ultraviolet monochromator and then detected by a position sensitive detector.

The photons of the exciting beam are collected in a Faraday cup. To convert the secondary electron current of its cathode into an absolute photon flux, the quantum efficiency of the cathode has to be known. These data were obtained in two ways. At first, the cup was calibrated by the laboratory of the PTB (Physikalisch Technische Bundesanstalt) at BESSY in the range of 36–120 eV with an error of $\pm 14\%$. This was done one month before the measurements. Second, we performed an *in situ* comparison with an aluminum cup calibrated by the National Institute of Standards and Technology in the range of 25–120 eV [28]. This yielded efficiencies with an error of $\pm 16\%$. Both data sets agreed within $\pm 2\%$.

In a first step, PIFS yields cross sections for the emission of fluorescence radiation from an excited state: the fluorescence signal normalized to the photon flux gives relative emission cross section, since the quantum efficiency of our monochromator-detector system is unknown. To get the absolute emission cross sections we normalized our data to the absolute emission cross sections for electron-impact induced line radiation. This method is described in detail elsewhere [29]. To use it here, our experimental setup was supplemented by an electron gun. The exciting photon beam was replaced *in situ* by an electron beam, keeping all experimental parameters constant during this exchange. For the normalization we used emission cross sections for electron-impact induced line radiation with a low error of $\pm 8\%$, determined very recently by Jans *et al.* [30]. The total error

of the presented cross section is $\pm 18\%$, mainly caused by the error in the determination of the exciting photon flux.

To equalize the determined emission cross section with the photoionization cross section for the argon 3s electron, three points have to be kept in mind. As the $3s3p^6^2S_{1/2}$ state is the lowest excited state of Ar II, no further branchings exist besides the observed ones. Second, some cascades originating from other excited states (satellite states) are possible above 35 eV. From known cross sections for these satellite states [16] and from an estimation of their possible branchings we estimate an upper limit of 5% for the influence of these cascades. Since we did not correct the data for these cascade effects, the present data may be interpreted as an upper limit for the photoionization cross section. Finally, due to the radial symmetry of the $^2S_{1/2}$ state the emitted fluorescence shows no angular distribution. Therefore the observed signal is proportional to the photoionization cross section, although we were not using the “magic” observation angle where the intensity is polarization independent.

III. TOF MEASUREMENTS

The Ar 3s partial cross sections and angular distribution anisotropy parameters β were measured in the photon energy region between 36 and 77 eV using a time-of-flight (TOF) electron spectrometer. The experimental setup consists of one analyzer measuring the electrons emitted along the main direction of the electric field vector and another looking under the magic angle (54.7°) with respect to the electric polarization vector of the photon beam. The entire vacuum chamber including the analyzers can be rotated around the photon beam in order to determine the photon’s degree of linear polarization and the angular distribution of the emitted electrons. The experiments were carried out partly at the 5.6-m toroidal grating monochromator (TGM) at HASYLAB and partly at the 14.9-m TGM-5 at BESSY which is located behind an undulator. Kinetic energy spectra of the emitted electrons were taken at several photon energies in the above mentioned energy range. For the Ar 3s measurements we used a mixture of argon and helium as a target gas. In order to obtain a precise ratio of this mixture in the interaction region, we took complete spectra of pure argon and helium under the magic angle at 77 eV photon energy with electron energies down to 0.2 eV. The argon spectrum was normalized to the recently published total photoionization cross section of Samson *et al.* [25], who quote an accuracy of $\pm 3\%$. From this normalization we determined a value of 0.157(8) Mb for the Ar 3s cross section 0.85(4) Mb for the Ar 3p cross section. The given accuracies include the uncertainty of the absorption values. For the normalization of the helium spectra we used the total absorption cross section from West and Marr [24], who gave a most probable error of 3.5%. The He 1s cross section was found to be 0.73(4) Mb also including the errors from the absorption. Taking these cross sections into account we could determine the mixing ratio in the interaction region from an argon-helium spectrum at the same photon energy (77 eV) with an accuracy better than 11%. Since the mixture is in-

TABLE I. List of all Ar 3s cross sections measured using the PIFS method. The numbers in parentheses indicate the uncertainty of the last digit(s).

$h\nu$ (eV)	σ (Mb)	$h\nu$ (eV)	σ (Mb)
28.40	0.003(1)	33.21	0.48(9)
28.90	0.000(1)	33.29	0.42(8)
29.00	0.003(1)	33.36	0.37(7)
29.10	0.000(1)	33.44	0.37(7)
29.20	0.073(13)	33.52	0.36(6)
29.30	0.89(16)	33.60	0.34(6)
29.40	0.87(16)	33.68	0.27(5)
29.50	0.90(16)	33.75	0.20(4)
29.60	0.90(16)	33.83	0.44(8)
29.70	0.88(16)	33.92	0.37(7)
29.90	0.88(16)	33.99	0.44(8)
30.00	0.89(16)	34.07	0.37(6)
30.10	0.88(16)	34.15	0.34(6)
30.20	0.88(16)	34.23	0.31(5)
30.29	0.84(15)	34.32	0.31(5)
30.30	0.84(15)	34.40	0.54(9)
30.35	0.83(15)	34.48	0.58(10)
30.41	0.81(15)	34.56	0.48(8)
30.48	0.78(14)	34.65	0.43(7)
30.54	0.77(14)	34.73	0.53(9)
30.61	0.72(13)	34.81	0.38(6)
30.67	0.73(13)	34.90	0.33(5)
30.74	0.73(13)	34.98	0.26(4)
30.80	0.64(12)	35.07	0.30(5)
30.87	0.84(15)	35.15	0.33(5)
30.93	0.80(15)	35.24	0.29(5)
31.00	0.70(13)	35.32	0.24(4)
31.07	0.63(12)	35.41	0.21(4)
31.13	0.62(11)	35.50	0.17(3)
31.20	0.64(12)	35.58	0.18(3)
31.27	1.03(19)	35.67	0.26(4)
31.34	0.86(16)	35.76	0.21(4)
31.40	0.79(14)	35.85	0.20(3)
31.47	0.75(14)	35.94	0.17(3)
31.54	0.67(12)	36.03	0.20(3)
31.61	0.97(18)	36.12	0.19(3)
31.68	0.88(16)	36.21	0.14(2)
31.75	0.80(15)	36.30	0.14(2)
31.82	0.76(14)	36.39	0.16(3)
31.89	0.68(12)	36.48	0.13(2)
31.96	0.69(13)	36.58	0.082(14)
32.03	0.67(12)	36.90	0.140(23)
32.10	0.66(12)	37.90	0.096(16)
32.17	0.62(11)	38.90	0.056(9)
32.25	0.59(11)	39.90	0.027(4)
32.32	0.67(12)	40.00	0.022(4)
32.39	0.61(11)	45.00	0.021(3)
32.46	0.60(11)	50.00	0.059(10)
32.54	0.57(10)	55.00	0.098(16)
32.61	0.57(10)	60.00	0.12(2)
32.68	0.59(11)	65.00	0.14(2)
32.76	0.49(9)	70.00	0.16(3)
32.83	0.54(10)	75.00	0.16(3)
32.91	0.57(10)	80.00	0.16(3)
32.98	0.55(10)	90.00	0.15(3)
33.06	0.51(9)	100.00	0.14(2)
33.13	0.52(9)		

dependent of the incident photon energy the Ar 3s cross section could then be normalized to the He 1s cross sections [24], which below the $n=2$ threshold (65.40 eV) and outside of well-known resonances [31] are equal to the total cross sections of helium. For each photon energy we have taken spectra at eight different angles with respect to the polarization vector of the incoming photon beam. The angular distributions of the He ($\beta=2$) and Ar 3s ($\beta\approx 2$) lines are very similar so that uncertainties in the degree of linear polarization or in the tilt of the polarization plane have negligible influence on the cross section measurements.

IV. RESULTS AND DISCUSSION

The cross sections for the photoionization of the argon 3s electron, obtained by PIFS and TOF, as described above, are summarized in Tables I and II, respectively. They are compared in Fig. 2(a) and 2(b) for the threshold region and in Figs. 3(a) and 3(b) for the ‘‘Cooper’’ minimum region of the cross section up to 100 eV with most of the previous experimental data. Only some of the theoretical data are included, since a detailed comparison of these was given recently by Saha [11].

The available experimental data close to threshold are those of Samson and Gardner [2], which are in quite good agreement with the present PIFS data. Our determined threshold value for the cross section of 0.86(15) Mb is within the error bars also in good agreement with most of the theoretical predictions, as shown in Fig. 2(b). But in contrast to these, the data obtained by PIFS indicate a rather flat energy dependence within 1 eV above threshold. Further experiments with reduced error bars are necessary to state a preference for one of the other theories near threshold.

Further PES data are available only above 31 eV. The lack of data below and the discrepancies just above this energy result from the problem of the determination of the detection efficiency for low energy electrons. This again demonstrates the special ability of PIFS for threshold measurements, since no detection efficiency variation occurs due to the variation of the exciting photon energy.

TABLE II. List of all Ar 3s cross sections measured using the TOF method. The numbers in parentheses indicate the uncertainty of the last digit(s).

$h\nu$ (eV)	σ (Mb)
36.20	0.19(4)
38.20	0.080(15)
40.00	0.028(5)
41.40	0.013(3)
43.05	0.010(2)
44.10	0.015(3)
45.11	0.024(5)
47.12	0.036(7)
49.07	0.059(11)
50.13	0.064(12)
51.14	0.072(13)
53.08	0.092(17)
55.08	0.11(2)
77.00	0.157(8)

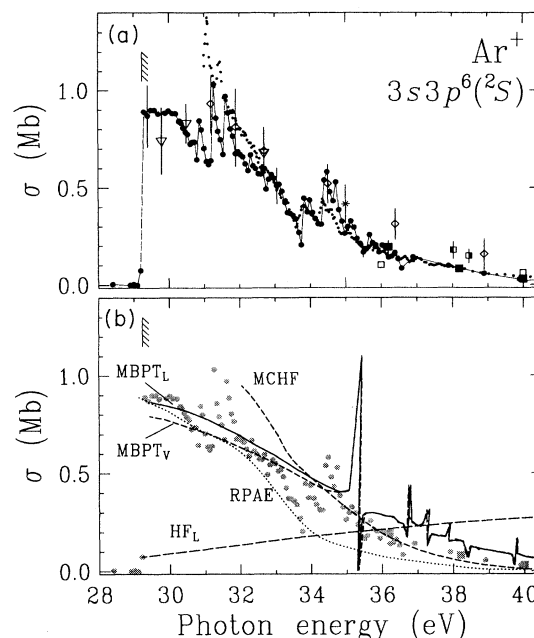


FIG. 2. Partial cross sections of the Ar 3s $\rightarrow \epsilon p$ transition are shown as function of the incident photon energy from the threshold (29.24 eV) up to 40.5 eV. (a) compares experimental data: \bullet , this work PIFS (only representative error bars are shown); \blacksquare , this work PES; ∇ , Samson and Gardner [2]; \diamond , Houlgate *et al.* [19]; \square , \square , Adam *et al.* [21], normalized to West and Marr [24] and Samson [26], respectively; \circ , Adam, Morin, and Wendin [13]; \square , Adam, Morin, and Wendin [23]; $*$, Kammerling [27]. In order to guide the eye in the resonance region, the PIFS data points are connected by a solid line. In (b) our experimental data (shown as gray areas) are compared to some selected theoretical calculations: Hartree-Fock (FH_L) Kennedy and Manson [3] (length form); random-phase approximation with exchange (RPAE) Amusia, Ivanov, and Chernysheva [1]; many-body-perturbation theory ($MBPT_{L,V}$) Wijesundera and Kelly [10] (length and velocity form, respectively); multiconfiguration Hartree-Fock (MCHF) Saha [11].

In our TOF experiment special care was taken with respect to the low energy transmission function. Our recent TOF and PIFS data agree well with each other in this energy range.

As observed by Schartner *et al.* [14] and independently by Adam, Morin, and Wendin [13] and more recently by Wills *et al.* [15], strong resonance structures appear at about 31 eV and some weaker ones at higher energies. They result from the autoionization of doubly excited atomic states. These were firstly found by Madden, Ederer, and Codling [34], who saw resonance structures in the total absorption cross section. The available experimental data show that only some of the atomic states do influence the 3s cross section strongly, which is discussed in detail in Refs. [14,15]. Up to now only Wijesundera and Kelly [10] included resonance structures in their calculations. Unfortunately, they only paid attention to doubly excited atomic states between 34.2 and 40.7 eV, so that the especially strong resonances around 31 eV were

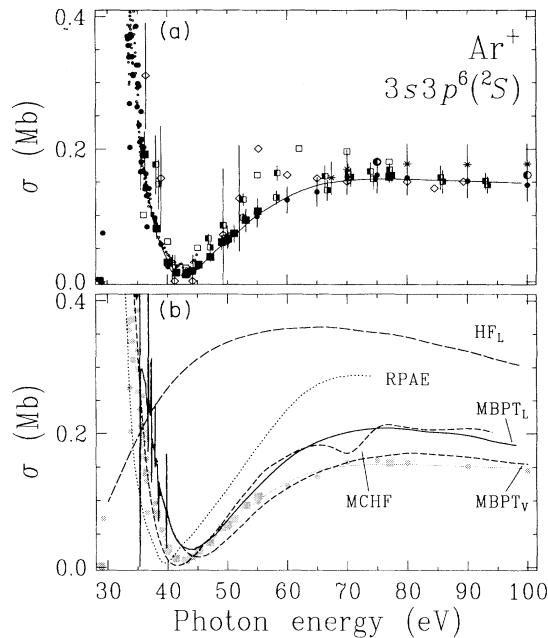


FIG. 3. Same as in Fig. 2 but with a wider energy range. Additional experimental results are from Wuilleumier *et al.* [22] (\bullet). In order to clarify the picture (a), above 51 eV the error bars from Houlgate *et al.* [19] (\diamond) are omitted and an eye-guiding solid line is drawn through our data points above the resonance region [both being shown as gray areas in (b)].

not considered.

TOF experiments were done with special respect to the “Cooper” minimum region of the cross section. Our data confirm the statement of Wijesundera and Kelly [10], that due to the contribution from the imaginary part of the dipole matrix elements for the transition $3s \rightarrow \epsilon p$, the cross section for $3s$ photoionization does not fall to zero. In contrast, most of the other calculations find a zero value as a result of the compensation of contributions from different transition amplitudes. The minima measured by other groups tend also to stay above zero intensity.

Above the “Cooper” minimum the $3s$ photoionization cross section rises to a maximum at about 80 eV followed by a slow decrease again. Most discrepancies in the experimental data occur around 60 eV. As demonstrated already by Adam *et al.* [21] they do occur, because differing data for the total argon absorption cross section determined by West and Marr [24] and by Samson [26] were used for the normalization of the PES data [21] and references therein]. For the evaluation of our recent TOF data we used the total-helium-absorption data from West and Marr [24] and the total-argon-absorption cross section at 77 eV recently published by Samson *et al.* [25]. The resulting cross sections agree with those of Adam *et al.* [21] normalized to the older data of Samson [26]. Both data sets are confirmed by the cross sections resulting from our PIFS measurements, being obtained independently of any total absorption data. We think that the present data overcome the uncertainties within this energy range.

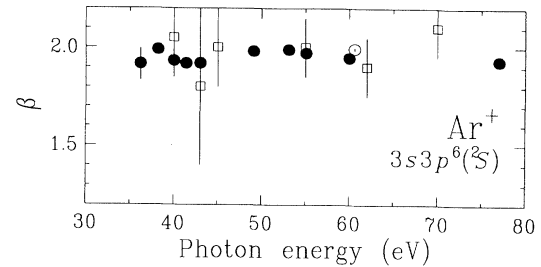


FIG. 4. Angular distribution anisotropy parameters β of the Ar $3s \rightarrow \epsilon p$ transition as a function of the photon energy. The theoretical value is $\beta=2$, the experimental data are \bullet , this work PES (only one representative error bar is shown); \square , Adam, Morin, and Wendin [23]; and \circ , Krause *et al.* [16].

Above 80 eV all experimental data agree within their error limits. In this energy range most calculations differ significantly from the experimental data, namely they are too large. Only the velocity form of Wijesundera and Kelly’s calculations [10] shows a very good agreement.

Figure 4 shows the angular distribution anisotropy parameter β for the energy range between 35 and 77 eV. Our results agree with all of the former measurements within mutual error bars, although our data points have substantially reduced error bars (Table III). The results confirm the photon energy independent β value of 2 for nearly all studied photon energies. If there exists a variation from this value in the vicinity of the “Cooper” minimum this variation would be very small, not exceeding 0.1 β units. This is in marked contrast to the behavior of the Xe $5s$ photoline where a distinct variation of the β parameter in the Cooper minimum has been observed [32,33]. This shows that besides the different importance of relativistic effects also the correlational effects in both elements might be very different. Theoretical data on this subject do not exist for comparison but would be very helpful in understanding these differences in more detail.

V. SUMMARY

Absolute cross sections for the photoionization of the argon $3s$ electron were presented. Our independently

TABLE III. Ar $3s$ angular distribution anisotropy parameter β . The numbers in parentheses indicate the uncertainty of the last digit(s).

$h\nu$ (eV)	β
36.20	1.91(6)
38.20	1.99(6)
40.00	1.93(6)
41.40	1.92(6)
43.05	1.92(10)
49.07	1.98(6)
53.08	1.99(6)
60.00	1.95(6)
77.0	1.93(6)

determined PIFS and TOF data agree well with each other. PIFS allowed a scan over the threshold. The assumption that the cross section does not fall to zero at the "Cooper" minimum was confirmed by our TOF measurements. The present data overcome the discrepancies in the experimental data near 60 eV and demonstrate the need for including doubly excited atomic states at low energies and the failure of most theories to properly calcu-

late the 3s partial cross section for photon energies above the "Cooper" minimum.

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