Observation of Resonances in the Ar-3s Photoionization Cross Section

K.-H. Schartner, B. Möbus, and P. Lenz
I. Physikalisches Institut der Justus-Liebig-Universität, D-6300 Giessen, Federal Republic of Germany

H. Schmoranzer and M. Wildberger
Fachbereich Physik, Universität Kaiserslautern, D-6750 Kaiserslautern, Federal Republic of Germany
(Received 1 August 1988)

Resonancelike structures have been observed by means of photon-induced fluorescence spectroscopy in the Ar-3s photoionization cross section between the threshold and the Cooper minimum. They coincide with resonances classified in published absorption spectra of Ar by $3s^2 3p^4 nln'l'$ excitations. Moreover, structures were found to correlate with resonances in the satellite production also studied by photon-induced fluorescence spectroscopy.

PACS numbers: 32.80.Fb

Resonance phenomena are prominent features of the interaction of photons with atoms. They are present in strong ionization channels,[1] as well as in weak satellite processes,[2] and stem from Rydberg series of the neutral atom. Interchannel interactions cause these discrete states to autoionize into continuum states which are degenerate in energy with states reached by direct photoionization from the ground state. The investigation of resonances can yield detailed information about the manifold of electron correlations.

Resonancelike phenomena have not been observed so far in the photoionization of the Ar-3s electron[3-5] though absorption spectra of Ar show pronounced window and absorption resonances above the Ar-3s threshold.[6] Also, until very recently,[7] the published calculated Ar-3s photoionization cross sections[8-10] showed a smooth passage from threshold on through the Cooper minimum. Thus they reproduced the main experimental features known so far. The calculations by Wijesundera and Kelly[7] include for the first time two-electron excitations which strongly influence the Ar-3s photoionization cross section in the form of narrow resonances. Though the selected doubly excited Ar configurations taken into account in these calculations are clearly not sufficient —as will be shown in the following—the calculations certainly underline again the importance of electron correlations for an understanding of the photon atom interaction. It is demonstrated hereby that even a well studied process like the Ar-3s photoionization is not understood completely. The implication of the presented results will hold also for photoionization of the outer s shell of the heavier rare gases. Moreover, new experiments with Ar, Kr, and Xe will be stimulated.

We report in the following the first experimental evidence of the presence of resonances in the energy dependence of the Ar-3s photoionization cross section. They have been observed for photon energies between the threshold and the Cooper minimum. Photon-induced fluorescence spectroscopy (PIFS) was used in the experiments. First results of an application of PIFS to Ar have been reported recently.[11]

The simplified level scheme of Ar and Ar^+ in Fig. 1 might be helpful for an understanding of the application of PIFS for studies of the Ar-3s photoionization process. PIFS allows the study of the photoionization of the Ar-3s electron by a measurement of the intensity of the $3s\,3p^6\,{}^2S_{1/2}$-$3p^2 3p^5\,{}^2P_{3/2,1/2}$ transitions. Correspondingly, the energy dependence of the satellite lines can be investigated by PIFS. Only a short description of the experimental setup is given here. The monochromatized synchrotron radiation ($\Delta E = 150$ meV) traverses a differentially pumped target gas cell. The target pressure is kept at 10 mTorr. The photons are collected in a Faraday cup. The secondary-electron current from its cathode is used for normalization of the fluorescence signal. The latter is obtained from a position-sensitive detector mounted in the focal plane of a home-built vacuum ultraviolet monochromator of the Pouey 28° type, which is used to disperse the fluorescence radiation. The beam of impinging photons, which has a diameter of 0.5 mm, serves as the entrance slit of the monochromator.

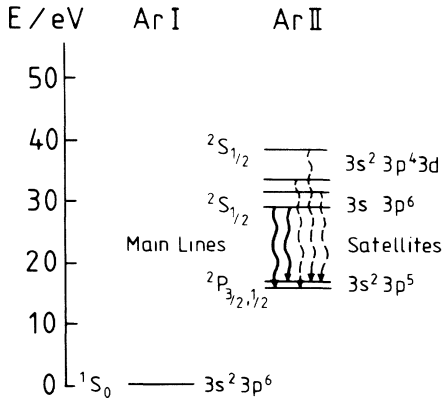

FIG. 1. Simplified energy level diagram of Ar I and Ar II.

Thus, high luminosity is achieved at a spectral resolution of 0.8 nm. Details can be found in Refs. 11 and 12.

In the previous application of PIFS, which concentrated on the double ionization of Ar into the $3s3p^5$ configuration,[11] the energy of the impinging photons was increased from threshold on to 40 eV in steps of 1 eV and to 120 eV in steps of 5 eV. Fluorescence spectra were recorded between 65 and 96.5 nm. In a further application of PIFS to Ar from threshold to 36 eV, the width of the energy steps was now decreased to 0.07 eV. Both experiments used synchrotron radiation from the storage ring BESSY at Berlin, which was monochromatized by a toroidal grating monochromator.

In Fig. 2 fluorescence spectra for a few energies of the impinging photons around 33 eV have been reproduced. The displayed spectra have not been corrected for the relative detection efficiency of the monochromator detector combination. The dominant lines in Fig. 2 result from the Ar II $3s3p^6\,^2S_{1/2}$-$3s3p^5\,^2P_{3/2,1/2}$ transitions. The lines at shorter wavelengths are classified as satellites.

Two runs in the mode of fluorescence excitation spectroscopy have been carried out, i.e., the signal of the lines at 91.9 and 93.2 nm was integrated for a preset time of 100 s. In between these two runs the storage ring was refilled with electrons. Figures 3(a) and 3(b) reproduce the respective signal variation with photon energy. The signal is normalized to the integrated secondary-electron current, which was recorded along with the fluorescence signal to ensure that no structures in the signal were introduced by the normalization procedure. In a third run, complete spectra like those displayed in Fig. 2 were accumulated in the PIFS mode. The line intensity was determined afterwards [Fig. 3(c)]. In this case the background signal could be subtracted. The accumulation time of the spectra varied from 200 to 600 s. Thus several injections were necessary to obtain the energy variation of the line intensities. In the threshold range, a few spectra only were registered with the intention of the energy-scale calibration. A threshold value of 29.24 eV was assumed. The scale of the normalized signals in Figs. 3(a)–3(c) was chosen to resemble the number of accumulated counts, allowing a judgment of the statistical uncertainties of the data points. The error bars (2σ) have been indicated in Figs. 3(a) and 3(b), whereas they are of the order of the dot size for Fig. 3(c).

The results in Figs. 3(a)–3(c) display the expected decrease of the fluorescence intensity towards the Cooper minimum. (So far, the secondary-electron current from the Faraday cup cathode cannot be converted to the photon flux with satisfactory accuracy. A decrease of the photon-induced secondary-electron emission of $\lesssim 20\%$ between 30 and 40 eV might be expected from the literature data.) As a new observation, narrow structures

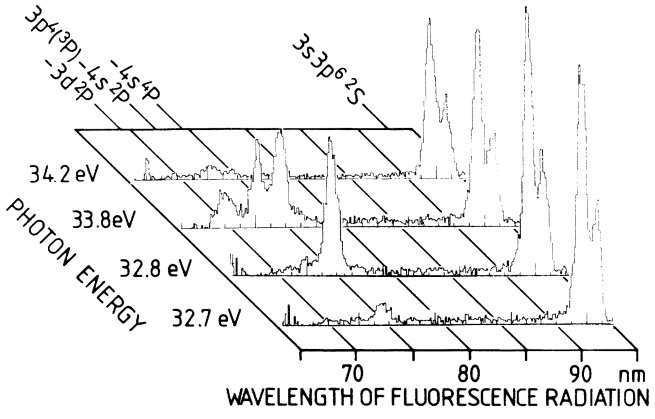

FIG. 2. Fluorescence spectra with the energy of the exciting photons as parameters.

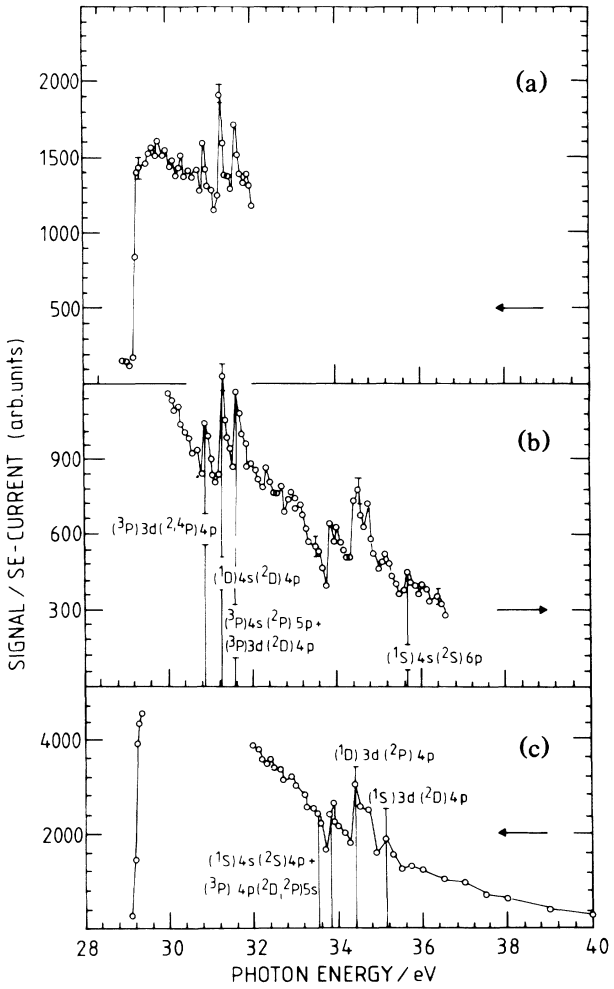

FIG. 3. (a)–(c) Intensity of the $3s3p^6\,^2S_{1/2}$-$3s^23p^5\,^2P_{1/2,3/2}$ transitions (normalized to the secondary-electron current from the Faraday cup) as functions of exciting photon energy. The arrow indicates the direction of the photon energy variation.

show up in the process of Ar-3s photoionization. The assumption of a proportionality between fluorescence signal and ionization cross section within the narrow energy range of the observed structures is certainly justified (see above).

Resonances in the photoionization continuum of Ar have been studied with high-resolution absorption techniques by Madden, Ederer, and Codling.[6] In addition to the prominent resonances due to the 3s-3p excitation into the $3s3p^6np$ states, a large number of rather strong absorption resonances above the Ar-3s threshold have been observed. They have been explained to result from two-electron excitations into $3s^23p^4nln'l'$ states, which were classified by quantum defect calculations. In Figs. 3(a)–3(c) the position and classification of strong features in the absorption spectra have been marked. Table I contains the respective numerical values. We state a strong correlation between the structures in the Ar-3s photoionization cross section which are presented here, and the resonances seen in the photoabsorption spectra.[6]

There exists only one very recent calculation of the Ar-3s photoionization cross section which takes into account the influence of two-electron excitations.[7] These ones—listed in Table I—show up as resonances in the Ar-3s photoionization cross section. We mention that the strongest effect is caused by the $3s^23p^4(^1D)$-$4p(^2P)5s$ excitation at 35.25 eV. This resonance is within the energy range of our investigation. Its energy as given by Wijesundera and Kelly[7] is in good agreement with the value from Madden, Ederer, and Codling.[6] Moreover, we observe a small structure at 35.1 eV. At this energy, Madden, Ederer, and Codling[6] have classified resonances from two-electron excitations into the $3s^23p^4(^1D)3d(^2D)4p$ states. The resonance calculated by Wijesundera and Kelly[7] is rather narrow (a width $\lesssim 50$ meV can be estimated from Fig. 5 of Ref. 7). Moreover, it has a dispersion-type profile, so that this resonance will be partially washed out in the measurement in view of the bandwidth of the impinging photons of ≈ 150 meV. The situation is different for the only other resonance in the calculation which is within the investigated energy range. It results from the $3s^23p^4(^3P)4p(^2P)5s$ state and was predicted by Wijesundera and Kelly[7] for 34.2 eV while Madden, Ederer, and Codling[6] have found an energy value of 33.6 eV. Neither at 34.2 nor at 33.6 eV do we observe a pronounced peak. On the other hand, this resonance also does not show up in the Ar-3s cross section calculated by Wijesundera and Kelly.[7]

In addition to sharp maxima in Figs. 3(a)–3(c), sharp minima of the window type are visible, which seem to be correlated with narrow intensity maxima of satellites of the type $3s^23p^4nl$. Satellites are known to show strong resonances in their excitation cross section. This has been observed by Becker and co-workers by photoelectron spectroscopy[2,13] and shows up in the calculations by Wijesundera and Kelly.[7] Becker *et al.*[13] observed that the relative contribution of the $3s^23p^4(^3P)4s^4P$ satellite to the main line $3s3p^6$ peaks strongly close to 34 eV. This effect is also demonstrated in Fig. 2. A first analysis of the satellite intensities in the spectra which

TABLE I. Comparison of absorption resonances from Ref. 6 and calculated resonances in the Ar-3s photoionization cross section from Ref. 7 with the structures observed in the present investigation.

Configuration	Energy value (eV) Ref. 6	Ref. 7	Present analysis
$3s^23p^4(^3P)3d(^2P)4p$	30.7, 30.84		30.9
$(^3P)3d(^2D)4p$	31.67, 31.69		
$(^3P)3d(^2F)4p$	31.6		31.6
$(^1D)3d(^2S)4p$	?		
$(^1D)3d(^2D)4p$	?		
$(^1D)3d(^2P)4p$	34.4		34.4
$(^1S)3d(^2D)4p$	35.02, 35.12		35.1
$3s^23p^4(^3P)4s(^2P)4p$	29.03, 29.22		
$(^1D)4s(^2D)4p$	31.23		31.25
$(^1S)4s(^2S)4p$	33.5		
$(^3P)4s(^2P)5p$	31.44, 31.45		
$(^1D)4s(^2P)5p$	32.84		
$(^1S)4s(^2S)5p$	?		
$(^1S)4s(^2S)6p$	35.75		35.65
$3s^23p^4(^1D)4d(^2S)4p$		36.56	
$(^1D)5d(^2S)4p$		37.10	
$(^1D)3d(^2S)5p$		37.10	
$(^1D)4d(^2S)5p$		39.51	
$3s^23p^4(^1D)4p(^2P)5s$	35.25	35.24	35.1
$(^3P)4p(^2P)5s$	33.6	34.20	

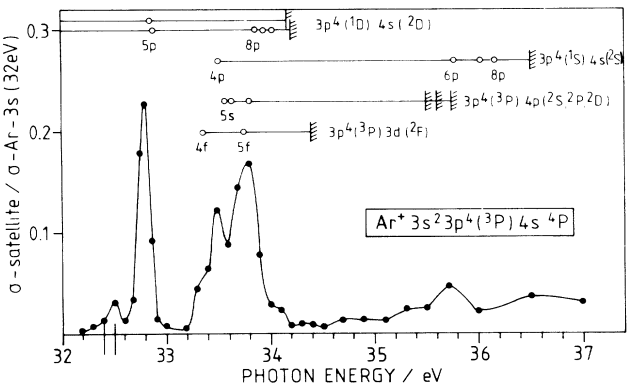

FIG. 4. Intensity of the $3s^23p^4(^3P)4s^4P$-$3s^23p^5{}^2P$ transitions (normalized to the secondary-electron current from the Faraday cup) as functions of exciting photon energy in units of the $3s3p^6{}^2S_{1/2}$-$3s^23p^5{}^2P_{1/2,3/2}$ intensity at 32 eV. Excitation thresholds are indicated. Resonances $3s^23p^4nln'l'$ are taken from Ref. 6.

are the basis of Fig. 3(c) reveals an additional resonance at 32.8 eV with an experimental half-width of 150 meV.

This new resonance is present in Fig. 4, where the intensity of transitions from the $3s^2 3p^4(^3P)4s\,^4P$ satellite (73.7–74.8 nm) as a function of the photon energy is shown. The ordinate of Fig. 4—fluorescence signal divided by secondary-electron current—allows the determination of the cross section for production of the satellite relative to the one of Ar-3s ionization. The latter is set equal to 1 at 32 eV for this comparison. (The relative wavelength dependence of the quantum efficiency of the fluorescence detection has been obtained by a comparison with electron-impact–induced fluorescence radiation from Ar.[14] The resonance at 32.8 eV is probably correlated with the narrow intensity decrease at 32.8 eV in Figs. 3(b) and 3(c). Furthermore, a correlation of the satellite production between 33.5 and 34 eV and the structure in Figs. 3(b) and 3(c) in this energy range appears likely.

We summarize that new resonancelike structures in the Ar-3s photoionization cross section and satellite production have been observed. They could be classified by comparison with published absorption spectra. From a comparison with existing recent calculations, we conclude that the number of coupled excitations has to be increased, especially by inclusion of more two-electron excitations. However, also more experimental work will be necessary to determine the shape of the observed spectral features. This demands still higher resolution of the impinging photons, and is coupled with the need of higher synchrotron radiation flux as expected from existing and forthcoming wiggler/undulator systems.

From the presented results for Ar, one can expect that resonance structures should be present also in the energy dependence of the Kr-4s and the Xe-5s photoionization cross sections. For the latter, resonances close to threshold have already been observed.[15]

We have appreciated helpful discussions with Dr. U. Becker and Dr. R. P. Madden. K.-H.S., P.L., and B.M. gratefully acknowledge the continuous interest of Professor Dr. A. Scharmann in their work. Technical support by the staff of BESSY is gratefully acknowledged. This work has been funded by the German Federal Minister for Research and Technology under Contract No. 05 352 AX I0.

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