

## High-resolution pre-edge structure in the inner-shell ionization threshold region of rare gases Xe, Kr, and Ar

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The total ion and/or electron yield spectra of rare gases Xe, Kr, and Ar have been recorded around the  $4d^{-1}$ ,  $3d^{-1}$ , and  $2p^{-1}$  ionization thresholds, respectively, with very high photon energy resolution at the Finnish beam line at the MAX I storage ring. Linewidths of the excited states are determined for higher excitations than earlier. The widths are observed to differ slightly for each core-excited state. [S1050-2947(96)04710-5]

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### I. INTRODUCTION

The absorption spectra of rare gases were one of the first experiments that showed the usefulness of synchrotron radiation in spectroscopic experiments already in the early 1960s [1]. However, the synchrotron radiation excited absorption spectroscopy has not been able to compete in accuracy with the electron energy-loss spectroscopy that uses electron bombardment excitation. This has been due to relatively poor photon energy resolution of the monochromatized radiation. The continuous development of the instrumentation of synchrotron radiation beam lines has changed the situation. Today the best monochromators can provide adequate photon intensity with photon energy resolution so high that the synchrotron radiation excited spectra can overcome the electron energy-loss spectra.

The electron energy-loss results of King and co-workers [2,3] have served a long time as a reference for the linewidths of the resonantly excited states in rare gases. They have been applied, e.g., when testing the performance of new monochromators. Although the resolution has improved, very few more accurate values for the linewidths have been given. Also, the widths obtained in Refs. [2] and [3] contain only a few low resonances. In this paper we shall present very high resolution total yield spectra of rare gases Xe, Kr, and Ar near the  $4d^{-1}$ ,  $3d^{-1}$ , and  $2p^{-1}$  ionization thresholds, respectively. The aim is to give more accurate values for the linewidths of the excited states and to extend the studies also to higher resonances that mainly have lacked reliable values so far. Recently, Masui *et al.* [4] have given more accurate values for the widths of the Xe  $4d^{-1}np$ ,  $n=6-9$ , resonances. Those values greatly differ from the values given by King *et al.* [2].

Accurate widths for the resonances are of general interest since these values are used, as mentioned above, when determining the photon energy resolution of new monochromators. By using high-intensity undulator radiation sources, the photon energy bandwidths comparable to or even smaller than the lifetime widths can be used in resonance Auger spectroscopy (Auger resonant Raman spectroscopy [5]). The knowledge of exact natural widths has become important since they are needed when trying to avoid line-shape distortion effects in the experimental spectra [6]. Moreover, the

deexcitation spectra of the higher resonances near threshold can give complementary information for the studies of the postcollision interaction effects. But, to consider the different possible initial-state effects in the deexcitation process [7] we must be able to distinguish the overlapping resonances with some reliability.

### II. EXPERIMENT

The experiments were carried out on the Finnish beam line [8] at MAX I synchrotron radiation laboratory in Lund, Sweden. Synchrotron radiation from a short-period undulator [9] was monochromatized by a modified SX-700 plane grating monochromator [10]. The beam line has been designed mainly for gas-phase experiments. Thus a permanent differential pumping system containing also a refocusing toroidal mirror has been installed after the monochromator.

The total electron or ion yield spectra of Xe, Kr, and Ar were recorded by using the time-of-flight (TOF) mass spectrometer described in [8]. The sample gases were introduced through a capillar tube directly into the excitation region. The pressure in the TOF chamber was about  $2 \times 10^{-5}$  mbar during the measurements, but probably much higher in the gas jet.

The undulator spectrum is composed of sharp peaks, which means that the photon intensity varies as a function of the photon energy. This has been taken into account by recording the photon flux simultaneously and dividing the total yield intensity by the flux. The recorded photon energy range in different measurements was also kept rather short, not more than 3 eV in Xe and Kr and 6 eV in Ar, in order to minimize inaccuracies due to varying photon intensity.

### III. DATA HANDLING

The total experimental width of each line was obtained by a least-squares fit of Voigt functions. The observed linewidth is a convolution of the natural lifetime width of the excited state and the experimental broadening. The lifetime width is supposed to be of a Lorentzian shape. Actually, the interference between the autoionization (i.e., participator) decay channel of the excited state and the direct photoionization channel should lead to an asymmetric Fano profile for the absorption line [11], as in the well-known case of He

$1s^2 \rightarrow 2s2p$  absorption. However, the inner-shell resonance excitations in the rare gases have appeared to decay prominently via the spectator process and therefore the interference effects between the direct and autoionization channels are expected to be so small that they do not distort the line profiles [7,12–14].

The experimental broadening function is a convolution of three different factors: the source, the exit slit width, and the slope errors of the optical elements of the monochromator. The source and the slope errors are supposed to give a Gaussian broadening function, whereas the slit contribution should be a step function in the case of a single (exit) slit. When using very small slit widths the experimental contribution can be estimated to be close to a Gaussian shape. Especially in the cases of Xe and Kr, the experimental contribution to the total measured linewidth is so small that the measured line shape is almost Lorentzian. Thus the error made when supposing the total experimental contribution to be Gaussian has a negligible effect on the deconvoluted Lorentzian (natural) width. In the case of Ar, the effect can be more significant since at higher photon energies the photon band is broader and also the relative contribution of the slit to the total broadening function increases. This has been taken into account when estimating the error limits of the width values.

As mentioned earlier, the recorded photon energy range at a time was kept short because the varying photon intensity as well as the possible instabilities in the circulating electron beam of the storage ring might lower the experimental resolution. Two or more recordings were needed to cover the whole threshold region and the photon bandwidth had to be estimated for each recording separately. Therefore the measurements were carried out as follows. First, the strongest or the most isolated resonance— $4d_{5/2}^{-1}6p$  in Xe,  $3d_{5/2}^{-1}5p$  in Kr, and  $2p_{3/2}^{-1}4s$  in Ar—was taken as a reference line and recorded alone by using a very short (1.2 eV) photon energy range. The natural Lorentzian width of that line was determined carefully by using the procedure described in [15]. Then, the next resonances were recorded together with the reference line. The photon bandwidth was determined by dividing the total width of the reference line into the Lorentzian and Gaussian contribution, where the Lorentzian width was set to be the same as the natural lifetime width obtained above. The Gaussian contribution, i.e., the photon bandwidth in that recording, was then obtained using the numerical information on the widths of different shapes given by Lee [16]. After obtaining the photon band width, the natural (Lorentzian) widths of the other resonances were determined by using again the table by Lee, but now with the help of total widths and the Gaussian photon bandwidth.

The highest resonances could not be recorded together with the reference line. In those cases the photon bandwidth was estimated with the help of the “middle range” resonances, whose natural widths were obtained as described above. This means, of course, that the accuracy in the estimation gets poorer.

When comparing the energies of the resonance excitations to those of King *et al.* [2], we found that the photon energy scale of our monochromator is slightly compressed, about 4 meV/eV. This has been taken into account when determining the total full widths at half maximum of the measured lines.

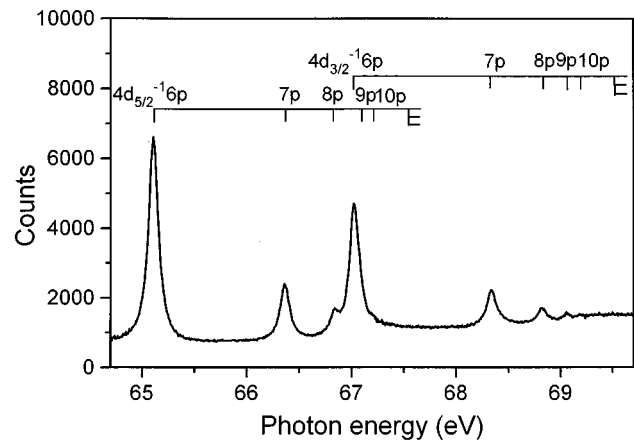


FIG. 1. Total yield spectrum of Xe; an overview, formed by combining two different recording ranges.

The possible errors in the width values mainly consist of difficulties in estimating the background properly. The error limits have been estimated by fitting the spectra with slightly different background shapes. In the case of Ar, the increased effect of uncertainties in the shape and width of the photon band has also been taken into account.

## IV. RESULTS

### A. Xenon

The total yield spectrum of Xe at the  $4d \rightarrow np$  excitation photon energy range is shown in Figs. 1 and 2. The width of the incoming photon beam was restricted by using a 2-mm horizontal aperture before the monochromator. Also the illuminated area of the refocusing mirror was diminished by a 2-mm vertical aperture. The photon bandwidth was found to be 6 meV in the measurement of the  $4d_{5/2}^{-1}6p$  resonance. Absorption resonances up to  $10p$  could be fitted individually rather reliably in both  $4d$  series. Higher resonances are fitted with curves that represent more or less a sum of the resonances converging to the ionization edge.

The results for the natural widths of measured lines are given in Table I together with the values given by Masui *et al.* [4] and King *et al.* [2]. Our values for the natural widths of the  $4d_{5/2}^{-1}np$ ,  $n=6,7$ , excited states are slightly larger than the values by Masui *et al.*. The results do not meet even within the error limits. If the results of Masui *et al.* were right, the resolution of our monochromator would be much worse than indicated by the measurement for the krypton  $3d^{-1}5p$  excited states (see below). Therefore, we conclude that the values given by Masui *et al.* are slightly too low. Our results for the rest of the  $4d_{5/2}^{-1}np$  series meet the results of Masui *et al.* within the error limits.

Our results show a tendency of slight decrease in widths when going on to higher resonances in both series ( $4d_{5/2}^{-1}np$  and  $4d_{3/2}^{-1}np$ ). If this tendency is real, it indicates that the Auger decay widths differ somewhat for each core-excited state. Such a difference could arise from various reasons, as discussed, e.g., in Refs. [17,18]. First, the absolute Auger decay rates are found to depend on the core-electron orbitals, which, furthermore, are sensitive to the electronic structure. The continuum orbital is affected when the field it

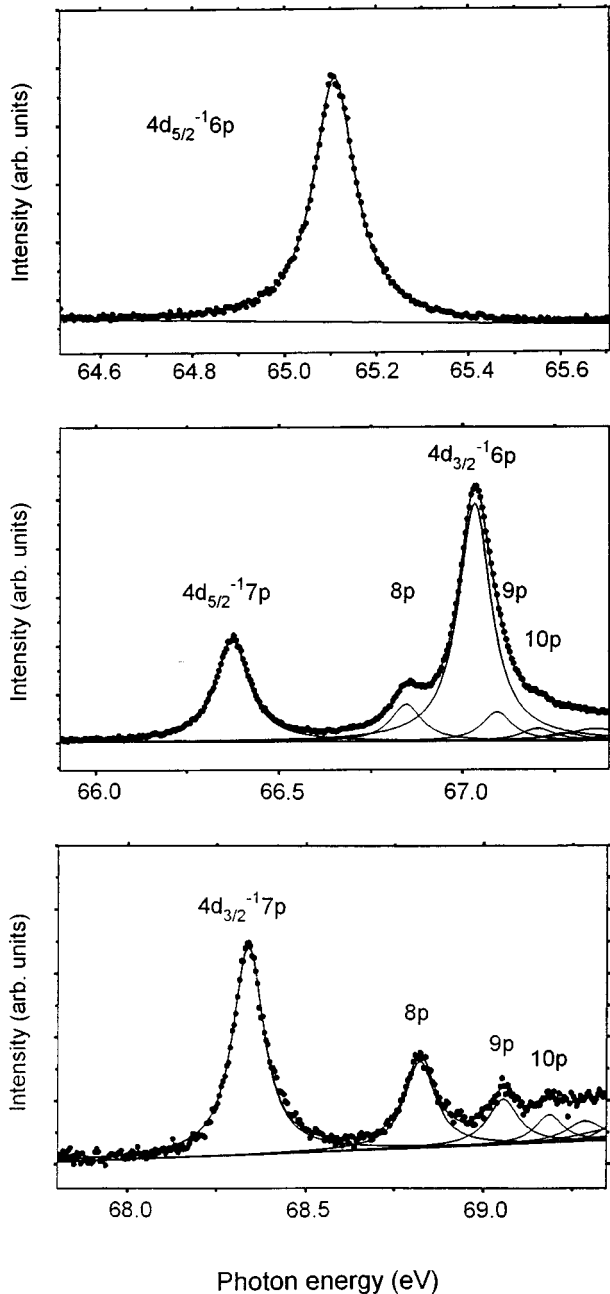


FIG. 2. Some details of the total yield spectrum of Xe. Top, the  $4d_{5/2}^{-1} \rightarrow 6p$  excitation region; middle, the  $4d_{5/2}^{-1} \rightarrow np, n \geq 7$  and the  $4d_{3/2}^{-1} \rightarrow 6p$  excitation region; and bottom, the  $4d_{3/2}^{-1} \rightarrow np, n \geq 7$  excitation region. The solid lines correspond to the least-squares fit of Voigt functions to the experimental points.

experiences changes. Second, the decay rates depend on the kinetic energy of the transitions, as demonstrated clearly for Coster-Kronig transitions [19]. The energy dependence of Auger amplitudes might be pronounced especially at low kinetic energies. In the case of Xe, the transition to the  $5s^{-2}np$  final states have energies up to about 10 eV. The relative group decay rates to the  $5p^{-2}np$ ,  $5s^{-1}5p^{-1}np$ , and  $5s^{-2}np$  final states may thus change, depending on the principal quantum number  $n$  of the spectator electron. The decay spectra show that the intensity ratio of the  $4d^{-1}6p \rightarrow 5p^{-2}np$  and  $4d^{-1}7p \rightarrow 5p^{-2}np$  resonance Au-

TABLE I. Natural widths (meV) of the Xe  $4d^{-1}np$ ,  $n=6,7,8,9$  excited states.

State	King <i>et al.</i> [2]	Masui <i>et al.</i> [4]	This work
$4d_{5/2}^{-1}6p$	$111 \pm 4$	$106.3 \pm 0.5$	$109.8 \pm 1.0$
$7p$	$128 \pm 9$	$106.0 \pm 0.5$	$109 \pm 1$
$8p$		$104 \pm 1$	$103 \pm 3$
$9p$		$104 \pm 2$	$98 \pm 8$
$4d_{3/2}^{-1}6p$	$119 \pm 8$	$104 \pm 1$	$107 \pm 1$
$7p$	$133 \pm 15$	$106 \pm 5$	$106 \pm 3$
$8p$		$110 \pm 3$	$105 \pm 3$
$9p$		$106 \pm 3$	$104 \pm 5$

ger transitions is almost the same as the intensity ratio of the  $4d^{-1}6p$  and  $4d^{-1}7p$  excited states in the yield spectrum. For higher  $4d^{-1}8p$  resonances the relative decay rate to the  $5p^{-2}np$  final states is higher than could be expected from the corresponding absorption probability in the yield spectrum [7,12]. It seems that the decay of the higher excited state leads more probably to  $5p^{-2}np$  final states than the decay of the lower excited state. This could lead to a variation of the lifetime of the resonantly excited state. The results would indicate that the lifetime of the excited state is longer when the excited state has higher probability to decay to a lower-energy state. One has to, of course, bear in mind that the tendency is very slight compared to the error limits and therefore very definite conclusions cannot be made. Especially for the  $4d_{3/2}^{-1}np$  series, a small change in the background estimation gives different values for the linewidths. We have chosen a slightly increasing background caused by the onset of the giant resonance.

Masui *et al.* found ‘‘a marginal hint’’ for an increase in widths on going from the  $4d_{3/2}^{-1}6p$  to the  $4d_{3/2}^{-1}7p$  excited state. Instead of broadening, our results for the  $4d_{3/2}^{-1}7p$  excited state show a slight asymmetry with a small tail on the high photon energy side. The asymmetry is clearly seen even though the peak is fitted by using an increasing background. The asymmetry could be due to the excitation of  $f$ -type states. In Xe the  $4d$  absorption cross section has a huge resonance above the ionization threshold. This giant resonance is caused by excitations to  $\epsilon f$  continuum states. Since the giant resonance has some discrete character in elements after Xe it is possible that near the  $4d$  threshold it has some discrete character also in Xe. This could therefore be responsible for the asymmetry observed near the  $4d_{3/2}^{-1}7p$  excitation peak. At higher resonances there could be same kind of asymmetry, but the statistics of the spectrum do not allow us to draw further conclusions.

## B. Krypton

The total yield spectrum of krypton at the  $3d^{-1}np$  resonance excitation range is depicted in Figs. 3 and 4. In these measurements the width of the incoming photon beam was restricted by using 2-mm horizontal and 2-mm vertical apertures before the monochromator. The photon bandwidth was found to be 8 meV in the measurement of the  $3d_{5/2}^{-1}5p$  resonance. Also in Kr, resonances up to  $10p$  could be fitted individually rather reliably in both series. Curves on the

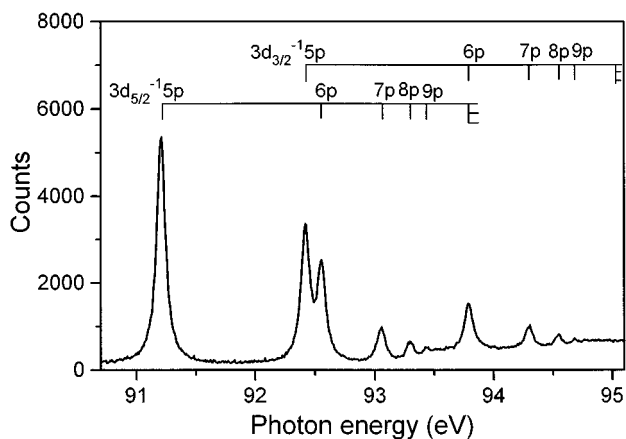


FIG. 3. Total yield spectrum of Kr; an overview, formed by combining two different recording ranges.

higher-energy side represent again more or less a sum of the resonances converging to the ionization edge.

The natural widths obtained are given in Table II. As can be seen, the width of the  $3d_{5/2}^{-1}5p$  excited state, which has largely been used in the determination of the monochromator resolution, is in good agreement with the earlier determined values [2,20].

At the  $3d_{3/2}^{-1}6p$  resonance an asymmetry can be observed at the high photon energy side. This is most probably due to the  $3d_{5/2}^{-1}$  ionization threshold lying almost exactly under the resonance (93.79 eV). The asymmetry could be fitted rather satisfactorily with a curve of arcus tangential shape. The  $3d_{3/2}^{-1}6p$  excited state seems to be broader than could be expected from the  $3d_{5/2}^{-1}np$  series. This is also probably due to the underlying  $3d_{5/2}^{-1}$  ionization threshold and difficulties in estimating the threshold shape properly.

The widths of the excited states show the same trend as in Xe: they seem to decrease when going on to higher resonances in series. As in Xe, the resonance Auger electrons have rather small energies, which can lead to changes in the relative intensity distribution of the resonance Auger transition to the  $4p^{-2}np$ ,  $4s^{-1}4p^{-1}np$ , and  $4s^{-2}np$  final states as a function of the initial excited state. The decay spectra of the excited states again support this explanation: the relative intensity of the transitions to the  $4p^{-2}np$  final states in the decay of the excited states is getting stronger with increasing  $n$  [13]. Due to large error limits, any definite conclusions cannot be made.

### C. Argon

The total yield spectrum of argon near the  $2p^{-1}$  ionization threshold is shown in Figs. 5 and 6. In these measurements the width of the incoming photon beam was restricted by using a 2-mm horizontal aperture before the monochromator.

In argon the background increases more strongly after the first ionization edge than in Kr and Xe. The reason for this is that the continuum states are predominantly of  $d$  character and the  $2p \rightarrow \epsilon d$  channel follows unbrokenly the  $2p \rightarrow nd$  series below the ionization edge, whereas in krypton and xenon the continuum states above the edge are predominantly of  $f$  character. The results of the deconvolution of the

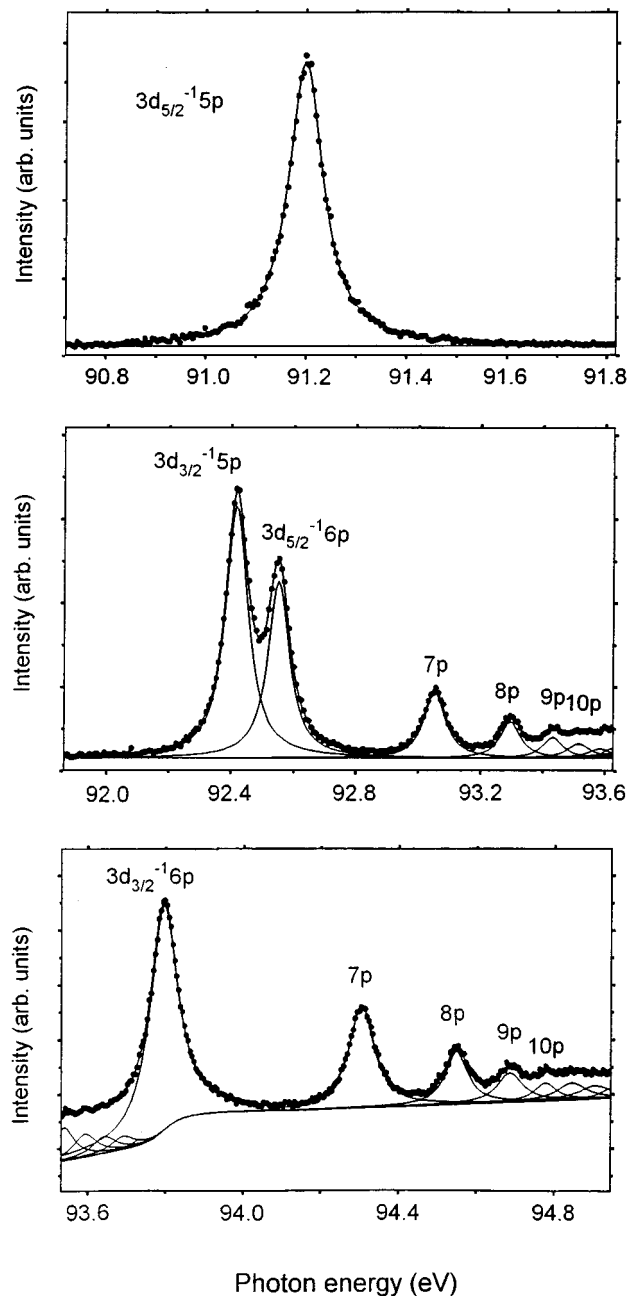


FIG. 4. Some details of the total yield spectrum of Kr. Top, the  $3d_{5/2}^{-1} \rightarrow 5p$  excitation region; middle, the  $3d_{5/2}^{-1} \rightarrow np, n \geq 6$  and the  $3d_{3/2}^{-1} \rightarrow 5p$  excitation region; and bottom, the  $3d_{3/2}^{-1} \rightarrow np, n \geq 6$  excitation region. The solid lines correspond to the least-squares fit of Voigt functions to the experimental points.

measured lines are given in Table III. The inaccuracies in the values are higher than in the cases of Xe and Kr because of the difficulties in obtaining the accurate shape and width of the monochromatized photon band. The monochromator broadening should increase as  $E^{3/2}$ . The resolution estimate (according to the measurement of the width of the krypton  $3d_{5/2}^{-1}5p$  excited state at 91.2 eV) should lead to a bandwidth of about 30 meV at 244 eV. The experimental broadening is therefore so large that the effects of possible errors on the estimated shape of the photon band are not negligible. Anyway, our results suggest a smaller width for the  $2p_{3/2}^{-1}4s$

TABLE II. Natural widths (meV) of the Kr  $3d^{-1}np$ ,  $n=5,6,7,8,9,10$ , excited states.

State	King <i>et al.</i> [2]	This work
$3d_{5/2}^{-1}5p$	$83 \pm 4$	$83 \pm 1$
$6p$		$79 \pm 2$
$7p$		$79 \pm 2$
$8p$		$76 \pm 3$
$9p$		$78 \pm 5$
$10p$		$75 \pm 8$
$3d_{3/2}^{-1}5p$		$83 \pm 2$
$6p$	$98 \pm 12$	$84 \pm 3$
$7p$		$77 \pm 3$
$8p$		$70 \pm 5$
$9p$		$70 \pm 8$
$10p$		$68 \pm 8$

excited state than has been suggested earlier in [3].

Opposite to the cases of Xe and Kr, the natural widths in Ar seem to increase with increasing  $n$ . Kinetic energies of the  $2p^{-1}$  Auger transitions in Ar are around 200 eV. At such high kinetic energies the Auger amplitudes may not show any pronounced energy dependence.

Above the  $2p_{3/2}$  ionization threshold the lines show clear asymmetry. Especially the peaks corresponding to the  $2p_{1/2}^{-1}3d$  and the  $2p_{1/2}^{-1}4d$  states are so asymmetric that additional peaks had to be fitted in both cases. This asymmetry has also been observed in an earlier study by Nakamura *et al.* [21]. They explained it to be due to interference between the direct photoemission channel and the autoionization decay channel leading to the same final states. As pointed out above, this interference should lead to an asymmetric Fano profile. However, the studies of the decay spectra of the  $2p_{1/2}^{-1}3d$  excitation have shown that the resonance Auger process, where the excited  $3d$  electron stays as a spectator during the decay process, strongly dominates over the autoionization (or participator) process [14]. Therefore the asymmetry caused by the interference should be very small, if detectable. We suggest that the asymmetry is partly due to background and partly due to excitations to the

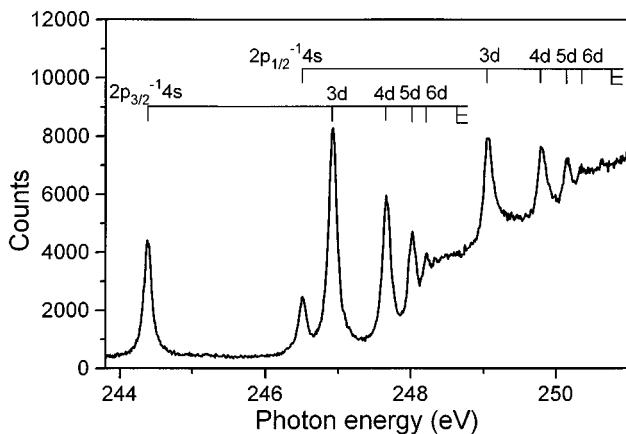


FIG. 5. Total yield spectrum of Ar; an overview, formed by combining two different recording ranges.

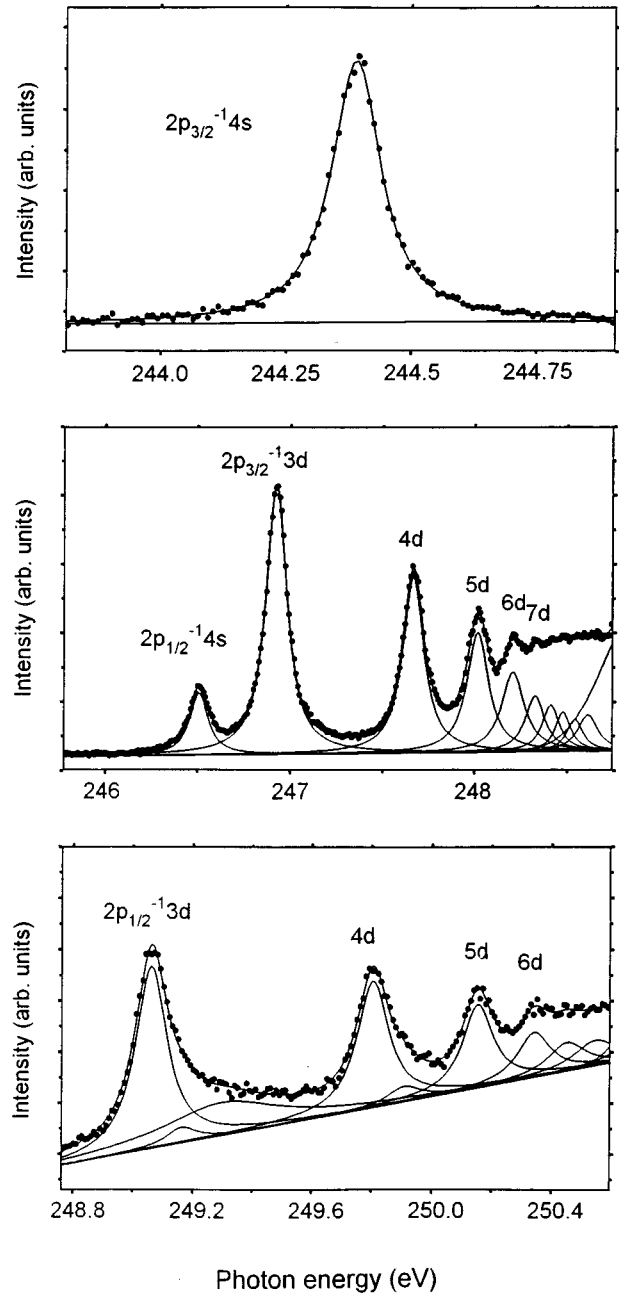


FIG. 6. Some details of the total yield spectrum of Ar. Top, the  $2p_{3/2}^{-1} \rightarrow 4s$  excitation region; middle, the  $2p_{3/2}^{-1} \rightarrow nd, (n+2)s$ ,  $n \geq 3$ , and the  $2p_{1/2}^{-1} \rightarrow 4s$  excitation region; and bottom, the  $2p_{1/2}^{-1} \rightarrow nd, (n+2)s$ ,  $n \geq 3$  excitation region. The solid lines correspond to the least-squares fit of Voigt functions to the experimental points.

$2p_{1/2}^{-1}(n+2)s$  states that should lie near the  $2p_{1/2}^{-1}nd$  states. The broad peak near the  $2p_{1/2}^{-1}3d$  peak in Fig. 6 can be connected to the background caused by the  $2p_{3/2} \rightarrow \epsilon d$  electrons. We have estimated a linear shape for the background as the first approximation, which certainly is not exactly correct. The two narrower peaks could indicate excitations to  $(n+2)s$  states. The fit of the spectrum gave these peaks relative intensities of about 10%, which is in good agreement

TABLE III. Natural widths (meV) of the Ar  $2p^{-1}4s$  and  $2p^{-1}nd$ ,  $n=3,4,5,6$  excited states.

State	Shaw <i>et al.</i> [3]	This work
$2p_{3/2}^{-1}4s$	$116 \pm 4$	$114 \pm 2$
$3d$	$118 \pm 4$	$125 \pm 3$
$4d$	$142 \pm 14$	$129 \pm 5$
$5d$		$133 \pm 10$
$6d$		$139 \pm 10$
$2p_{1/2}^{-1}4s$		$109 \pm 3$
$3d$	$132 \pm 10$	$115 \pm 5$
$4d$	$132 \pm 10$	$117 \pm 10$
$5d$		$112 \pm 10$
$6d$		$133 \pm 10$

with the value of 12% estimated by King *et al.* [2]. The inaccuracies in the background and in the positions of the weak  $2p_{1/2}^{-1}(n+2)s$  states do not allow us to give any proper estimations for the widths of those states. In the  $2p_{3/2}$  series, the  $(n+2)s$  states lie so near the  $nd$  states that they could

not be fitted separately. However, the fact that the determined linewidths for the  $2p_{3/2}^{-1}nd$  excited states are clearly broader than those for the  $2p_{1/2}^{-1}$  series indicates that the  $(n+2)s$  excitations are not negligible in the  $2p_{3/2}^{-1}$  series.

## V. CONCLUSION

We have measured the absorption structures in Xe, Kr, and Ar at the energy range of the  $4d^{-1}$ ,  $3d^{-1}$ , and  $2p^{-1}$  thresholds, respectively, with higher resolution than before. The widths of the excited states seem to decrease slightly as a function of  $n$  in Xe and Kr. This could be connected to the variation in the relative group decay rates of the excited states when the transition energy is low, as it is in Kr and especially in Xe. To confirm this, further studies of the decay processes of these excited states are needed.

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