

## Photoionization of Kr 4s: III. Detailed and extended measurements of the Kr 4s-electron ionization cross section

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**Abstract.** Absolute Kr 4s-electron photoionization cross sections as a function of the exciting-photon energy were measured by photon-induced fluorescence spectroscopy (PIFS) at improved primary-energy resolution. The cross sections were determined from threshold to 33.5 eV and to 90 eV with primary-photon bandwidths of 25 meV and 50 meV, respectively. The measurements were compared with experimental data and selected theoretical calculations for the direct Kr 4s-electron photoionization cross sections.

### 1. Introduction

The photoionization of the Kr 4s-electron has been intensively investigated theoretically in the past twenty years (Kennedy and Manson 1972, Amusia *et al* 1973, Huang *et al* 1981, Parpia *et al* 1984, Tulkki *et al* 1992, Schmoranzert *et al* 1993, Sukhorukov *et al* 1994). It turned out that electron correlations play an important role for the description of the 4s-electron photoionization cross section as a function of the exciting photon energy. Experimentally there have also been some efforts (Samson and Gardner 1974, Aksela *et al* 1986, Tulkki *et al* 1992, Schmoranzert *et al* 1993, Ehresmann *et al* 1994, Langer 1993) to determine these cross sections. However, there is up to now no work which covers the entire exciting photon energy range between the 4s-ionization threshold and, e.g. 90 eV, which is above the minimum of the cross section. Thus this work has two aims:

(i) a detailed measurement of the exciting-photon energy dependence of the Kr 4s-electron photoionization cross section near threshold with the best achievable resolution, using photon-induced fluorescence spectroscopy (PIFS);

(ii) a detailed compilation of the data from our most recent measurements at higher energies to provide a complete set of reliable cross sections for comparison with theory. Special emphasis has been laid on the cross sections around the minimum of the Kr 4s-electron photoionization cross section function.

Section 2 gives a brief description of the experimental set-up for photon-induced fluorescence spectroscopy (PIFS). The method of determining the Kr 4s-electron photoionization cross section from the spectroscopic data is described in section 3

including a detailed discussion of the experimental uncertainties. The results are presented in section 4.

## 2. Experimental set-up

The general features of PIFS and its applications have been described previously (Schartner *et al* 1988a b, 1990, Schmoranzner *et al* 1990, 1993). In particular the present set-up has been outlined by Schmoranzner *et al* (1993) and thus we only mention the major differences to this latter work.

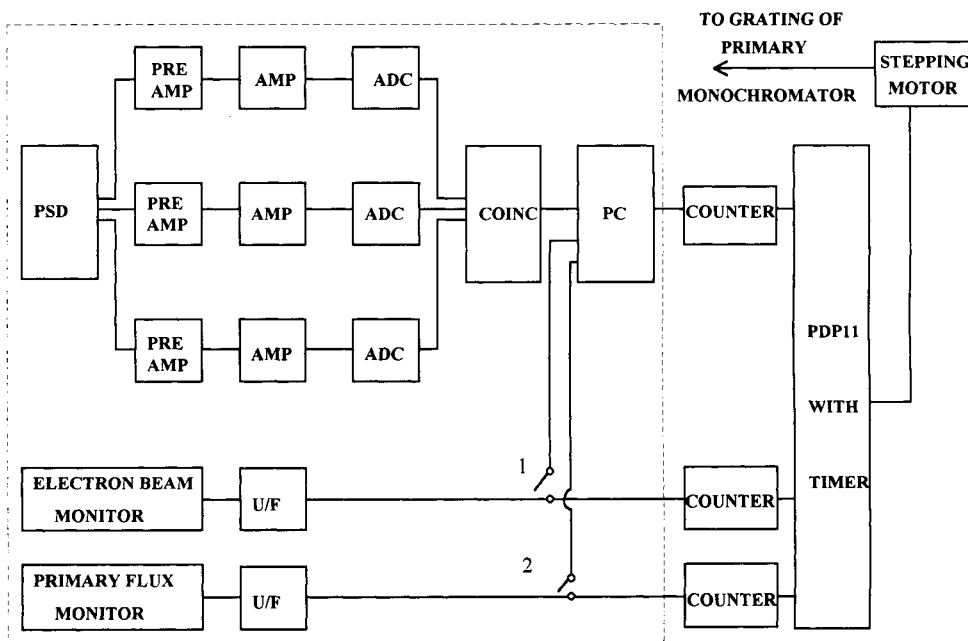


Figure 1. Schematic diagram of the present data acquisition system for PIFS.

Figure 1 shows a schematic diagram of the most important components for the data acquisition. The dashed box with closed switches 1 and 2 represents the configuration used for the multiplex data collection used previously (Schmoranzner *et al* 1993). The three signals which provide the spatial information of a detected photon are amplified, digitized and sent to a PC collecting the data. The energy of the exciting photons is set to a certain value and a fluorescence spectrum is evaluated from the positions of the detected photons. The time for the data collection is determined by a preset for the primary flux. Thus the individual spectra recorded at a certain excitation energy are automatically normalized to the primary photon flux. The signal of the electron beam monitor of the electron storage ring is also recorded by the PC.

In order to reduce fluorescence intensity variations caused by slow changes of the exciting-photon beam position the so-called scanning mode (instead of the multiplex mode) was used for the present investigation. The primary flux monitor, the electron beam monitor and the signal output were fed to three counters which were connected with a timer and a second computer (see figure 1). This second computer controls the energy of the incoming photons via a stepping motor for the grating of the primary

monochromator. The timer controls the time of data collection. Using this set-up the primary monochromator is set to a certain energy and then the signal of the fluorescence lines of interest is collected for typically 10 to 20 s per energy point. This procedure is automatically repeated for the energy interval of interest containing typically 50 energy points. To obtain good statistics several of these scans are added. Due to the short time needed for one scan variations of fluorescence intensity caused by slow changes of the exciting-photon beam position were averaged out.

The synchrotron radiation from the storage ring BESSY, Berlin, was monochromatized by a toroidal grating monochromator. The bandwidth of the monochromatized synchrotron radiation was set to 25 meV at 27.5 eV for the threshold range scans, which is an improvement by a factor of 6 as compared to Schmoranzner *et al* (1993), and to 50 meV at 27.5 eV for the wide range scans (see Ehresmann *et al* 1994).

### 3. Data analysis and uncertainties of the measurements

The absolute Kr 4s-electron photoionization cross section  $\sigma_{4s}(E)$  at exciting-photon energy  $E$  can be evaluated from the measured intensities  $I^{4s}(965 \text{ \AA}; E)$  and  $I^{4s}(917 \text{ \AA}; E)$  of the fluorescent decays of the 4s hole into the two Kr II ground states  $4s^1 4p^6 \ ^2S_{1/2} \rightarrow 4s^2 4p^5 \ ^2P_{1/2,3/2}$  at wavelengths of 964.8 \AA and 917.4 \AA, respectively, according to

$$\sigma_{4s}(E) = \frac{(I^{4s}(965 \text{ \AA}; E)\eta_{965} + I^{4s}(917 \text{ \AA}; E)\eta_{917})k}{\Phi^{4s}(E)} \quad (1)$$

where  $\Phi^{4s}(E)$  is the primary flux of the exciting photons at energy  $E$ ,  $\eta_{965}$  and  $\eta_{917}$  are the absolute quantum efficiencies of the monochromator-detector combination at 964.8 \AA and 917.4 \AA, respectively, and  $k$  is a factor which depends only on the experimental geometry and the target density and is independent of the fluorescence photons' wavelength. In analogy to equation (1) the Ar 3s-electron photoionization cross section  $\sigma_{3s}(E)$  can be determined from the intensities  $I^{3s}(932 \text{ \AA}; E)$  and  $I^{3s}(920 \text{ \AA}; E)$  of the transitions  $3s^1 3p^6 \ ^2S_{1/2} \rightarrow 3s^2 3p^5 \ ^2P_{1/2,3/2}$  at 932.1 \AA and 919.8 \AA, respectively. Since the factor  $k$  is unknown the known absolute Ar 3s-electron photoionization cross section (Möbus *et al* 1993) has been used to eliminate this factor, yielding for  $\sigma_{4s}(E)$

$$\sigma_{4s}(E) = \frac{(I^{4s}(965 \text{ \AA}; E)\eta_{965} + I^{4s}(917 \text{ \AA}; E)\eta_{917})\Phi^{3s}(E)\sigma_{3s}(E)}{(I^{3s}(932 \text{ \AA}; E)\eta_{932} + I^{3s}(920 \text{ \AA}; E)\eta_{920})\Phi^{4s}(E)} \quad (2)$$

Thus the absolute Kr 4s-electron photoionization cross section was determined using equation (2) from the measured intensities of the relevant fluorescence lines recorded in different scans for Kr and Ar. In equation (2) the absolute quantum efficiencies can be replaced by relative ones. These have been determined experimentally by replacing *in situ* the monochromatized synchrotron radiation beam by a 2 keV electron beam for electron impact excitation. The measured fluorescence intensities were compared with accurately (within an uncertainty of 8%) determined absolute fluorescence emission cross sections for 2 keV electron impact (Jans *et al* 1993). In this way the relative quantum efficiencies were evaluated within an uncertainty of 12%.

The measured fluorescence and exciting-photon intensities were corrected for higher orders of the primary monochromator (Bradshaw *et al* 1984) and the background, which together yield an uncertainty of <3%. The pressure in the target cell was kept

constant by an automatic control system within an uncertainty of  $<0.1\%$ . The pressure was determined by an absolute pressure sensor (MKS Baratron 1 Torr) which avoids the necessity of corrections to the measured pressure due to the changing of the target gas.

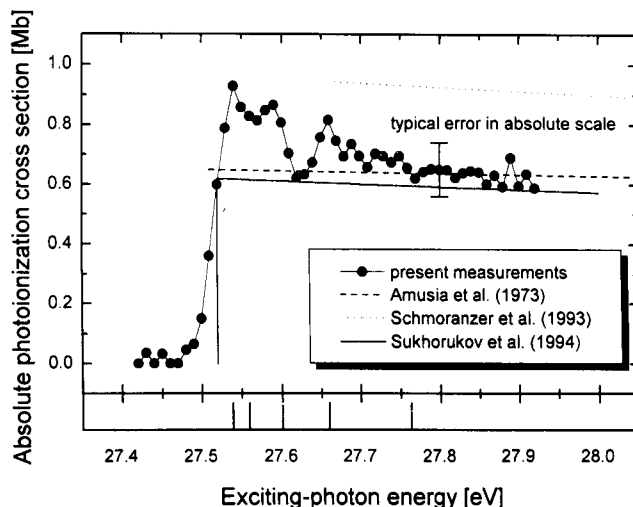
The absolute Ar 3s-electron photoionization cross sections were taken from Möbus *et al* (1993). Cross sections for energies which have not been quoted by Möbus *et al* (1993) were obtained by linearly interpolating the data of these authors. Below the Ar 3s-electron ionization threshold relative cross sections were determined as before (see Schmoranzer *et al* 1993) and calibrated to the absolute values above the 3s ionization threshold.

The presented cross sections have not been corrected for cascade effects which are assumed to be small (Schmoranzer *et al* 1993). The influence of cascades will be discussed in detail for the different energy ranges in section 4.

#### 4. Absolute Kr 4s-electron photoionization cross section

##### 4.1. Threshold range

Figure 2 shows the absolute 4s-electron ionization cross section in the exciting-photon energy range between 27.5 eV and 27.9 eV. Energy positions of atomic resonances (Codling and Madden 1971) have been indicated as bars at the bottom of the figure. The full line represents the most recent intermediate-coupling  $CI$  results of Sukhorukov *et al* (1993), the dotted line the previous  $LS$ -coupling  $CI$  results of the same group (Schmoranzer *et al* 1994) and the broken line the early  $RPAE$  results of Amusia *et al* (1973). There are no cascades in this exciting-photon energy range because the threshold of the lowest Kr II satellite level is at 27.99 eV. From figure 2 it is evident that the first four resonances affect the cross section (resulting probably in an increase of the cross



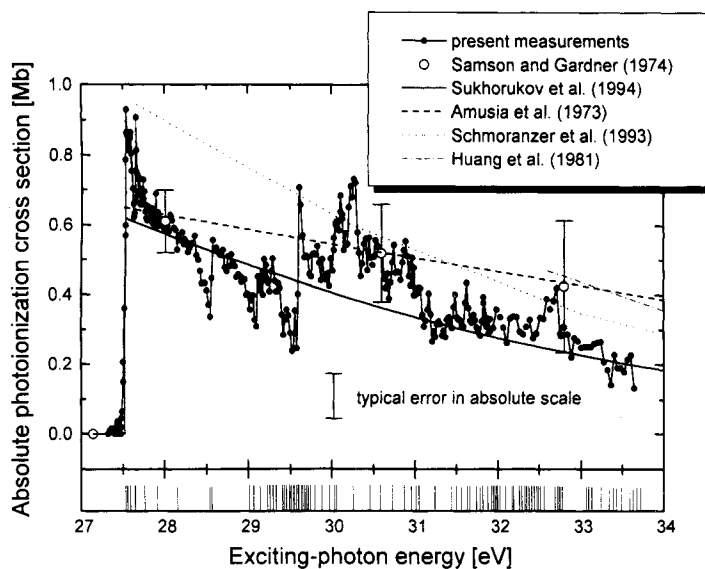
**Figure 2.** Absolute Kr II 4s-electron photoionization cross section in the near-threshold range. Measured positions of atomic resonances from Codling and Madden (1971) have been indicated as bars at the bottom of the figure.

section at the positions of the resonances) whereas the fifth resonance shows no significant effect.

In contrast to the 2s photoionization in Ne (Schartner *et al* 1992) and the 3s photoionization in Ar (Möbus *et al* 1993) where there is evidence for a plateau in the cross sections immediately above threshold, this conclusion can definitely not be drawn from the present data for Kr because of the occurrence of resonances at these energies (see figure 2). Since the theoretical results (Sukhorukov *et al* 1993, Schmoranzer *et al* 1993, Amusia *et al* 1973) have been obtained without including these resonance effects, a comparison between theory and experiment can only be drawn at exciting-photon energies where there are no atomic resonances. At these energies the configuration interaction (CI) calculations in intermediate coupling of Sukhorukov *et al* (1994) agree with the experimental values within the uncertainties of the measurements whereas the theoretical results of Schmoranzer *et al* (1993) obtained in the frame of *LS*-coupling CI technique are too high. The reason for this discrepancy is the explicit neglect of the so-called Coulomb interaction correlational decrease (cf Schmoranzer *et al* 1993, Sukhorukov *et al* 1994). In our preceding paper (Sukhorukov *et al* 1994) it was discussed in detail that correlation effects lead to a significant decrease of the Coulomb interaction which has influences on the energies of the atomic and ionic states and on the corresponding transition amplitudes, too. The intermediate configurations which influence most strongly the Coulomb matrix elements and the correlational corrections to the Coulomb matrix elements have been calculated in second-order perturbation theory and higher-order corrections have been estimated.

It is remarkable that the calculations by Amusia *et al* (1973) using the random-phase approximation with exchange (RPAE) also agree with the measurements in this energy range close to threshold.

Figure 3 shows the cross section in an extended energy range up to 33.5 eV. The present measurements are in agreement with earlier ones of Samson and Gardner (1974)



**Figure 3.** Absolute Kr II 4s-electron photoionization cross section in the exciting-photon energy range between threshold and 33.6 eV. Measured positions of atomic resonances from Codling and Madden (1971) have been indicated as bars at the bottom of the figure.

and Schmoranzer *et al* (1993). The latter experimental values have not been shown in figure 3 for simplicity. For the measurements of Samson and Gardner (1974) the error bars have been included in figure 3 for comparison with the present ones which are considerably smaller. The cross section is strongly modulated by resonance structures. The energetic positions of the atomic resonances have been measured early by Codling and Madden (1971) and were included as bars at the bottom of figure 3. The corresponding assignments of these resonances, however, are still lacking. These resonances lead to both increase or decrease of the 4s-electron ionization cross section, which is understandable in the framework of the theory of Fano (1961) if for different resonances different values for the  $q$ -parameter are assumed. At excitation energies where there are no resonances (e.g. at 28.7 eV) the calculations of Sukhorukov *et al* (1994) show the best agreement with the experimental data. Since all other calculations (Amusia *et al* 1973, Huang *et al* 1981, Schmoranzer *et al* 1993) yielded higher cross sections in this extended near-threshold range, this finding demonstrates very well the importance of including the Coulomb interaction correlational decrease (Schmoranzer *et al* 1993, Sukhorukov *et al* 1994) in the calculations.

Cascade effects are assumed to be small in this energy range since cascades may only occur via population of the  $4s^2 4p^4 4d/5d$  configurations which are mixed with the  $4s^1 4p^6 \ ^2S_{1/2}$  state. This assumption was corroborated at an exciting-photon energy of 31.5 eV where the PES measurements of Langer (1993) agree very well with the present results (see figure 4).

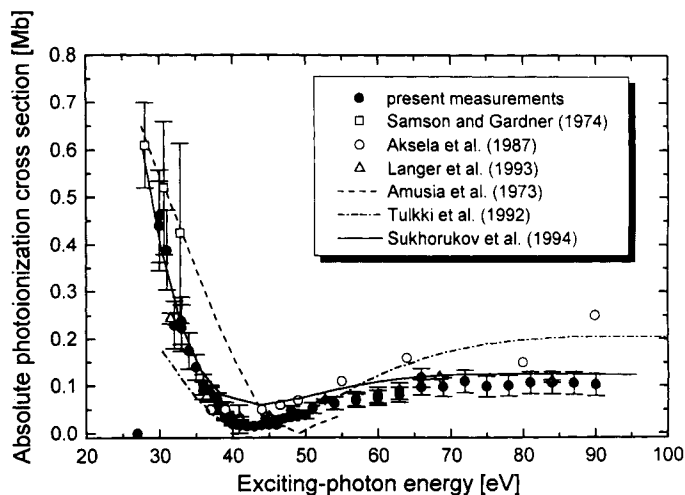


Figure 4. Absolute Kr 4s-electron photoionization cross section in the exciting-photon energy range between threshold and 90 eV.

#### 4.2. 30 eV to 90 eV range

Figure 4 shows the present absolute Kr 4s-electron ionization cross sections between 30 eV and 90 eV together with selected other experimental results (Samson and Gardner 1974, Aksela *et al* 1987, Langer 1993) and selected calculations (Amusia *et al* 1973, Tulkki *et al* 1992, Sukhorukov *et al* 1994). Cascade effects are assumed to be small in this energy range, too, as the comparison with PES cross sections from Langer (1993)

at several exciting-photon energies shows (figure 4). The previous experimental data of Ehresmann *et al* (1993) were completed particularly in the region of the minimum of

**Table 1.** Measured Kr 4s-electron photoionization cross section as a function of the exciting-photon energy.

$E_{\text{exc}}$ (eV)	$\sigma_{4s}$ (Mb)	$E_{\text{exc}}$ (eV)	$\sigma_{4s}$ (Mb)
29.97	0.400 (95)	46.97	0.031 (6)
30.97	0.388 (85)	47.98	0.041 (9)
31.97	0.230 (50)	48.98	0.041 (9)
32.97	0.243 (50)	50.98	0.054 (11)
33.97	0.175 (39)	53.98	0.064 (13)
34.97	0.140 (27)	56.98	0.071 (14)
35.97	0.097 (20)	59.98	0.078 (15)
36.97	0.083 (16)	62.98	0.086 (16)
37.97	0.061 (13)	65.99	0.107 (18)
38.97	0.043 (9)	68.99	0.098 (20)
39.97	0.023 (8)	71.99	0.110 (24)
40.97	0.019 (5)	74.99	0.099 (22)
41.97	0.015 (5)	77.99	0.100 (22)
42.97	0.015 (4)	80.99	0.107 (24)
43.97	0.017 (4)	84.00	0.106 (24)
44.97	0.023 (5)	87.00	0.106 (24)
45.97	0.023 (5)	90.00	0.102 (23)

the cross section function by the present results. Table 1 lists the experimental cross sections for this range of exciting-photon energy. The uncertainties quoted in table 1 are the calculated standard deviations for each individual cross section. It is worthwhile to note that the minimum of the Kr 4s-electron photoionization cross section is not as deep as the corresponding one in the Ar 3s-photoionization cross section. The comparison between theory and experiment reveals the following important facts.

(i) For the theoretical description of the direct Kr 4s-electron photoionization cross section it is necessary to include the Coulomb interaction correlational decrease to obtain agreement between experiment and theory in the near-threshold range (cf in figure 3 the curves from Schmoranzler *et al* (1993) and Sukhorukov *et al* (1994)).

(ii) The Coulomb interaction correlational decrease leads to a lower energy for the calculated minimum of the cross section which is in better agreement with the experiment (cf in figure 4 the curves from Sukhorukov *et al* (1994) and Amusia *et al* (1973)).

(iii) By including the configuration interaction of the  $4s^1 4p^6 \ ^2S_{1/2}$  state with the correlation satellites (cf Schmoranzler *et al* (1993) and Sukhorukov *et al* (1994)) in the theoretical consideration, significantly lower cross sections at exciting-photon energies above the energy of the cross section minimum were calculated. These improved theoretical results are found to agree very well with the present experimental data.

(iv) The best agreement between experiment and theory at energies around the minimum of the cross section was obtained by the calculations of Tulkki *et al* (1992). However, at higher as well as at lower energies with respect to the minimum these calculations compare less favourably with the measurements.

## 5. Summary

In the present paper the absolute Kr 4s-electron photoionization cross section was measured by PIFS for the first time with a narrow exciting-photon energy width of

25 meV from the threshold up to 33.5 eV. Cross sections up to an exciting-photon energy of 90 eV were measured with a bandwidth of 590 meV. A comparison with other available experimental data yielded agreement in the threshold range (Samson and Gardner 1974). In the energy range between 45 eV and 90 eV, however, the present cross sections are significantly lower than those of Aksela *et al* (1987) but agree very well with the most recent measurements of Langer (1993). Furthermore, the minimum of the Kr 4s-photoionization cross section as a function of exciting-photon energy was found at lower energies than predicted by Amusia *et al* (1973).

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### References

- Aksela S, Aksela H, Levasalmi M, Tan K H and Bancroft G M 1987 *Phys. Rev. A* **36** 3449–50
- Amusia M Ya, Ivanov V K, Cherepkov N A and Chernysheva L V 1973 *Proc. 8th Int. Conf. on Physics of Electronic and Atomic Collisions (Belgrade)* (Amsterdam: North-Holland) p 581 Abstracts
- Bradshaw A M, Braun W, Buchholz W, Fröhling R, Gaupp A, Peatman W, Petersen H, Pflüger J, Schäfers F and Wolf F P 1984 *BESSY Annual Report* pp 43–76
- Codling K and Madden R P 1971 *J. Res. NBS* **76A** 1–12
- Ehresmann A, Schmoranzer H, Vollweiler F, Mentzel G and Schartner K H 1994 *Phys. Lett. A* **184** 432–4
- Fano U 1961 *Phys. Rev.* **124** 1866–78
- Huang K N, Johnson W R and Cheng K T 1981 *At. Data Nucl. Data Tables* **26** 33–45
- Jans W, Kühne M, Ulm G, Anton M, Möbus B and Schartner K H 1993 *BESSY Annual Report* and private communication
- Kennedy D J and Manson S T 1972 *Phys. Rev. A* **5** 227–247
- Langer B 1993 private communication
- Möbus B, Magel B, Schartner K H, Langer B, Becker U, Wildberger M and Schmoranzer H 1993 *Phys. Rev. A* **47** 3888
- Parpia F A, Johnson W R and Radojević V 1984 *Phys. Rev. A* **29** 3173–80
- Samson J A R and Gardner J L 1974 *Phys. Rev. Lett.* **33** 671–3
- Schartner K H, Lenz P, Möbus B, Magel B, Schmoranzer H and Wildberger M 1990 *Phys. Scr.* **41** 853–6
- Schartner K H, Lenz P, Möbus B, Schmoranzer H and Wildberger M 1988a *Phys. Lett.* **128A** 374–7
- 1988b *Phys. Rev. Lett.* **61** 2744–7
- Schartner K H, Möbus B, Mentzel G, Ehresmann A, Vollweiler F and Schmoranzer H 1992 *Phys. Lett.* **169A** 393–5
- Schmoranzer H, Ehresmann A, Vollweiler F, Sukhorukov V L, Lagutin B M, Petrov I D, Schartner K H and Möbus B 1993 *J. Phys. B: At. Mol. Opt. Phys.* **26** 2795–810 (Corrigendum 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 377–9)
- Schmoranzer H, Wildberger M, Schartner K H, Möbus B and Magel B 1990 *Phys. Lett.* **150A** 281–5
- Sukhorukov V L, Lagutin B M, Petrov I D, Schmoranzer H, Ehresmann A and Schartner K H 1994 *J. Phys. B: At. Mol. Opt. Phys.* **27** 241–56
- Tulkki J, Aksela S, Aksela H, Shigemasa E, Yagishita A and Furusawa Y 1992 *Phys. Rev. A* **45** 4640–5