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Charge distribution of Kr ions produced upon photoionization around the 2s edge

A.C.F. Santos[a,*], S. Pilling[b], D.P. Almeida[c]

[a] Instituto de Física, Universidade Federal do Rio de Janeiro, Rio de Janeiro 21991-972, Brazil
[b] Laboratório Nacional de Luz Síncroton, Campinas 13084-971, Brazil
[c] Departamento de Física, Universidade Federal de Santa Catarina, Florianópolis 88040-979, Brazil

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ABSTRACT

Charge state spectra of krypton ions generated after ionization (by a single photon) of the *L* shell have been measured by using the PEPICO technique. Relative abundances of Kr^{q+} ions in charge state up to 8+ were obtained using monochromatized synchrotron radiation. A comparison with other experimental and theoretical data is presented.

1. Introduction

When penetrating X-rays pass through matter, they disperse energy by interaction with the target. In the photoelectric effect, the X-ray photons are totally absorbed in a single encounter and their energy is transferred to the target electrons, causing their ejection. The specific ionization, i.e., the number of primary and secondary ion pairs produced per unit length of the particle's path, of X-rays is several times smaller than a projectile electron of the same energy. The vacancy production after inner-shell excitation or ionization in an atom is accompanied by subsequent radiative, i.e., photon emissions due to the shift of the vacancy to outer shells and/or non-radiative decay (Auger or Coster–Kronig decay).

The connection between photoionization and the ionization by impact of fast charged particles has been long discussed for both atomic [1–6] and molecular [7] systems. For charged particle impact, the energy transfer to the target addresses an extensive range, from zero to the full energy of the incident beam. Consequently, both valence and inner-shell ionization and excitation processes are open. On the other hand, synchrotron radiation allows separate studies of valence and inner-shell ionization processes when the wavelength is selected accordingly. The yield of multiply charged ions by charged particle impact generally results from two separate processes: inner-shell ionization concomitant with secondary electronic ejection, or direct outer-shell ionization with simultaneous emission of *q* electrons. The detection of all products of the reaction in coincidence in order to identify unequivocally the reaction channel is a very complicated task. Up to date, only (e,2e), (e,3e) [8] and COLTRIMS [9] experiments are possible, where two or three emerging electrons can be collected after the collision. Therefore, the photoionization data can be helpful to investigate the multiple ionization channels by electron impact.

Alcantara et al. [7] carried out measurements of the branching ratios for the fast proton and VUV photons collisions on CH_2Cl_2. They pointed out that as the proton energy increased, the fragmentation pattern resembled more and more the corresponding photon impact spectra at lower energies. Their results show that the fragmentation pattern of the CH_2Cl_2 molecule by fast protons is dominated by distant collisions, and that the contribution of distant collisions in proton impact ionization is defined by the behavior of the photoionization near threshold. It was shown that the partial ion yield of the charged products in the proton impact spectra could be directly compared to the corresponding fragmentation pattern for photoionization through the momentum transfer, *Q*. For high projectile velocities, *Q* depends on the projectile velocity as v^{-1}.

In a previous paper, Santos, Homem, and Almeida [2] presented neon photoion charge state distributions as a function of photon energy after *K*-shell vacancy. By combining various theoretical and experimental data, they demonstrated that multiple ionization of Ne atoms by 2000 eV electron impact occurs mainly via

* Corresponding author. Tel.: +55 2139387947; fax: +55 2139387368.
E-mail address: toni@if.ufrj.br (A.C.F. Santos).

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outer-shell ionization. They anticipated that their methodology could be applied to more complex systems as well obtaining further knowledge on the ionization pathways resulting in multiple electron removal in charged-particle interactions.

Krypton represents a still more formidable challenge to analyze the observed multiple ionization in terms of production mechanisms. In this paper, we report on experiments on the production of multiply charged krypton ions created in the reorganization cascades subsequent to the production of single L edge vacancies in Kr, from the Laboratório Nacional de Luz Síncrotron (LNLS) SXS beam line. This paper is divided as follows: In Section 2 the experiment is briefly described and in Section 3 the results are presented.

2. Experiment

The experimental apparatus has been described in detail elsewhere [2,6]. Synchrotron radiation from the SXS beam line is dispersed to obtain monochromatic X-rays from 1900 eV to 1925 eV at the Brazilian Synchrotron Light Laboratory (LNLS). A Willey and McLaren time-of-flight mass spectrometer (TOF) is used for charge analysis of the resulting ionic fragments. The TOF is designed to have a maximized efficiency for ions with energies up to 30 eV. The Photo-Electron-PhotoIon COincidence (PEPICO) technique is used to acquire the mass spectra. The vacuum chamber base pressure is usually maintained in the order of 10^{-8} Torr. Typical gas pressure during the measurement was 1×10^{-5} Torr. The partial ion yields (PIY) are obtained by recording the ions produced after interaction of the linearly polarized light with krypton atoms at given X-ray energies. During the experiment, in order to avoid undesirable charge transfer between the recoil ion and the background, the pressure is kept below 10^{-5} Torr. The ejected electrons are accelerated in the direction opposite to that of ions; they are detected without energy analysis by two micro-channel plates detectors, and this switches on a 'start detecting ions' signal.

3. Results and discussions

Using the procedures outlined, relative yields for single and multiple ionization of krypton atoms were determined. A typical raw PEPICO spectrum is shown in Fig. 1. Generally speaking, the spectrum in Fig. 1 can be understood in terms of vacancy cascade. As it develops, the number of electrons in the shells decreases while the corresponding binding energies increase. The decay time of the core excited hole is given approximately by $\tau \sim \hbar/\Gamma$, where Γ = 215 meV is the Kr 2s lifetime broadening $\tau \sim 3$ fs [10].

From the PEPICO spectra, the partial ion yield of Kr ions can be obtained by integration of the corresponding peaks and normalization of the net areas to the total number of events. The relative fractions PIY^{q+} of ions (partial ion yield) in charge state i are given by

$$PIY^{q+} = \frac{I^{q+}/\varepsilon^{q+}}{\sum_i I^{i+}/\varepsilon^{i+}} \quad (1)$$

where ε^{q+} is the efficiency of the MCP for detecting a ion of charge state q. For the present spectrometer, the ion kinematics inside the TOF tube as well as the efficiency of the MCPs is essentially independent of the nature of the ions.

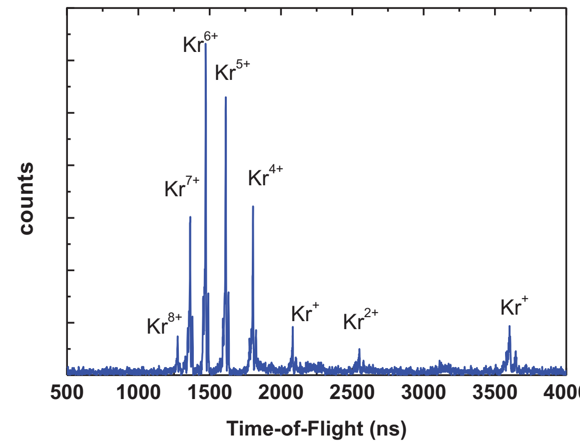

Fig. 1. Raw krypton TOF spectrum at 1920.5 eV corresponding to $2s^{-1}$ (L_1 shell) excitation process. The present mass resolution is not enough to allow getting a clear Kr isotope identification.

Fig. 2 and Table 1 show the partial ion yield of Kr^{q+} ions following L_1-shell vacancy production as a function of the X-ray energy. For energies above the L_1 edge, the photoionization cross sections are larger than the corresponding L_2 ones. The L_1L_2M and L_1L_3N Coster–Kronig transitions are the most probable processes for the de-excitation of L_1 vacancies [11]. In the present energy range, the PIY fractions exhibit a weak dependence on the photon energy. For instance, the yield of Kr^{7+} ions varies from 31.6% at 1900 eV to 50.7% at 1924 eV, which is greater than in some previously measured data as 40% from Ref. [10] obtained using low-resolution photons from filtered X-ray tube radiation and with poor control on the pressure at the target region; this is also greater than 35% from Ref. [11]. Short et al. [12] have analyzed this fact, and found higher low-q abundances than Carlson et al. [10]. These discrepancies may be associated with: (i) difficulties in discrimination of ionic charge state; (ii) detection efficiencies; (iii) difficulties in defining the optics due to stray photons in the incident beam that can introduce bad alignment of the optical system; (iv) when the pressure in the gas cell is too high, there are losses in the PIY counting rate due to the charge-transfer between the ions and the

Table 1
Partial Kr^{q+} ions following L_1-shell vacancy.

X-ray energy (eV)	Kr^{+} (±10%)	Kr^{2+} (±10%)	Kr^{3+} (<±10%)	Kr^{4+} (±8–10%)	Kr^{5+} (±5%)	Kr^{6+} (±1%)	Kr^{6+} (±2%)	Kr^{6+} (±5%)
1900	0.33	0.22	1.4	2.1	11.2	40.2	31.6	12.8
1916	0.053	0.10	1.1	1.5	10.6	46.2	32.2	8.3
1917	0.18	0.091	0.45	1.1	6.3	33.2	33.5	25.2
1918	0.12	0.14	1.2	1.4	10.0	42.1	28.7	16.3
1919	0.24	0.079	0.35	1.0	6.1	34.6	36.9	20.7
1920	0.067	0.081	1.0	1.3	10.0	42.8	30.5	14.1
1920.5	0.45	0.14	0.60	1.2	6.2	30.8	37.4	23.2
1921	0.16	0.065	0.35	0.61	5.3	29.4	30.7	33.3
1922	0.17	0.055	0.38	0.71	5.8	34.6	36.6	21.7
1922.5	0.39	0.16	0.44	1.2	5.8	31.4	37.6	23.1
1923	0.17	0.043	0.34	0.72	5.5	33.3	34.3	25.6
1924	0.37	0.11	0.33	1.1	4.8	35.5	44.7	13.1