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Photoionization with Excitation of the 4s and 4p Subshells in Krypton

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PHOTOIONIZATION WITH EXCITATION OF THE 4s AND 4p SUBSHELLS IN KRYPTON

by

Ahmad Farhat

A Thesis
Submitted to the
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in partial fulfillment of the
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In spite of all of their contributions, no one bears any responsibility of any error or omission whatsoever. The burden solely rests on the shoulders of the writer.

Ahmad Farhat
PHOTOIONIZATION WITH EXCITATION OF THE
4s AND 4p SUBSHELLS IN KRYPTON

Ahmad Farhat, M.A.
Western Michigan University, 1994

High-resolution photoelectron spectra of the nd, Rydberg satellites in krypton have been measured for the first time for six different angles and for photon energies between 68.5 and 250 eV. The relative intensities of these satellites with respect to the 4s and 4p main lines are in very good accord with the most recent measurements of Krause et al. (1992) at 68.5 eV. They also confirm that the theoretical predictions for the relative intensity of these satellites are too high. We determine the ratios for nd satellites with respect to the 4s and 4p main lines, and extract the angular distribution parameter $B$. Our results generally confirm the predictions taken from the latest theoretical interpretations.
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CHAPTER I

INTRODUCTION

Photoionization is a process where a photon of frequency $v$ and energy $hv$ interacts with an atom or molecule to produce an ion and one or more electrons. The interaction can be represented by the equation

$$hv + M \rightarrow M^{m*} + me,$$

where $m \geq 1$.

Since ionization energies (the minimum energy necessary to remove an outer-shell electron from the system) lie in the range $5 - 25$ eV, the corresponding minimum frequency of the photon to produce ionization is about $1.3 \times 10^{15} - 6 \times 10^{15}$ s$^{-1}$. Not all photon absorption processes produce ionization. In molecules, for example, dissociation occurs with high probability at low photon energies. In this process neutral or excited fragments are produced. As the photon energy increase above threshold it eventually exceeds the binding energy of the electrons in inner shells of the atom or molecule, (these shells are labeled K, L, M, etc., counting from the most tightly bound electrons). Thus a very energetic photon, e.g. an X-ray, can remove any electron whose binding energy is less than the photon energy.
Since that time the interest has focused from "main line" phenomena, associated predominantly with "one electron" effects, to multielectron process governed by electron correlations. The latter manifest themselves by producing satellites lines near the main lines. This development was driven by the progress in instrumentation, particularly with synchrotron radiation, in contrast to the initial studies in this field. Today we see that the improvement in theory has led to sophisticated computational methods to meet the challenge of new experimental results, opening a period of fruitful interplay between theory and experiment. This introduction summarizes some aspects of this development, in particular on the behavior of satellite lines in photoelectron spectra.

Since the discovery of the photoeffect the variation of the photoionization cross section in the vicinity of the ionization threshold has attracted continuing interest. This is not surprising because the nature of the interaction between photons and atoms or molecules is most sensitively exhibited in the low energy, where the exited electron is still around or slowly leaves the ionized system. However, before the advent of synchrotron radiation as a tunable light source in the VUV and soft X-ray region, most threshold and resonance studies were restricted to theoretical calculations. Crude comparisons far from threshold were done with the data points available from
discharge lamps or X-ray tubes. Consequently, the first decade of synchrotron radiation work on atoms and molecules was concentrated on cross-section measurements to assess the different theoretical predictions particularly near threshold.

**Photoionization Cross Section**

The photoionization cross section is the quantity that reflects the electronic properties of an atom. The variation of the cross section with photon frequency is a window through which one can study the electronic nature of an atom subshell by subshell. In the case of krypton, a 36-electron system, the atom we will center our work on, our detailed knowledge begins to diminish significantly.

Several people contributed in this area, for example from threshold to 59.4 eV, Samson’s results (Samson, Lee, 1966) are accepted to the same degree of reliability as they were for the lighter noble gases. The X-ray absorption data of Lukiskii et al. (1969) effectively cover the region from about 49.6 to 539.8 eV. From 90 to 400.5 eV, the DESY synchrotron results (Haensel et al., 1969) complement those values and provide evidence for structure undetected in the X-ray line source experiments. Samson quotes values of Ederer and Tomboulian at 73 and 67.1 eV. Henke’s recent tabulation (1972) carries our knowledge from 392.9 to 1488.6 eV, with the same reliability. Below 1448.6 eV, the
data of Wuilleumier (1972) have been utilized out to 8277 eV, in order to estimate the oscillator strength contribution of the 2s-2p shell. The calculated values of Veigele et al. (1971) have been used between 14336.6 and 1677.7 eV. As will be noted, these shells contribute very slightly to the oscillator strength.

As with argon, krypton also manifests autoionization resulting from excitation of an electron from the outer orbital to ns and nd Rydberg levels. They converge upon the higher energy member \( ^2P_{1/2} \) of spin-orbit pair and decay to lower energy member \( ^2P_{3/2} \).

Since the spin-orbit splitting is significantly larger for krypton, it is easier to resolve the individual resonances, and in fact to absorb sharp and diffuse components corresponding to s-like and d-like quasi-discrete states where the complete wave function for the system is a linear combination of discrete and continuum states.

Below about 17.7 eV, the partial cross section due to ejection of a 4p electron diminishes (as with the 3p of argon) and approaches a cooper minimum at about 71 eV. The so-called cooper minimum is displayed by the absolute photoionization cross section at photoelectron energies \( \epsilon \) where the amplitude of the main transition \( l \rightarrow l+1 \) passes through a zero. The excitation of 4s electrons to Rydberg levels again gives rise to resonances beginning at about 24.8 eV and extending to the threshold for 4s ejection at
about 27 eV. (Samson, 1964) and (Madden and Colling, 1964)

At about 92 eV we encounter the first example of ionization from a d-like orbital. It will be recalled that the larger l-value of such an orbital gives rise to an angular momentum barrier separating inner well states and outer well states.

The data of West and Marr (West et al., 1976) provides a welcomed addition to the otherwise sparsely studied region around the cooper minimum (49.6-92 eV). Their values around 177.3 eV also alter the shape of the d absorption.

More specifically, Schmidt (1980) summarized the main aspects of our understanding of resonance and threshold effects in atomic and molecular photoionization at the beginning of the 80's. Cooper minima, shaped resonances and the effect of interchannel coupling in the cross sectional behavior of main lines were among the more prominent issues studied both experimentally and theoretically.

Advances in the experimental (Samson et al., 1982) and theoretical (Starace et al., 1982) methods revealed the importance of electron correlations and collective phenomena upon the partial cross section and the angular distribution of the emitted photoelectron.
Partial Cross Section

In the interval between \(^2\)P\(_{3/2}\) and its spin-orbit partner \(^2\)P\(_{1/2}\) (14.00-14.66 eV), typical autoionization features are exhibited by Rydberg states converging to \(^2\)P\(_{1/2}\). One notes sharp peaks identified as np→ns transitions, and broad ones corresponding to np→nd transitions, as in the case of argon.

Beyond the \(^2\)P\(_{1/2}\) threshold, the ratio of cross section \(\sigma_{3/2}/\sigma_{1/2}\) is less than statistical (2/1) as predicted by the Walker et al. (1973) theory for declining partial cross section but experiments by Samson et al. (1975) have shown a surprisingly constant ratio of 1.77 up to 18 eV above this threshold. The Walker theory would imply a declining value for this ratio as the cooper minimum is approached.

The onset for ejection of an electron from the 4s shell occurs at 27.514 eV (Moore, 1949). The Rydberg series converging to this limit has characteristic window resonances (Madden and Codling, 1964). Samson and Gardner (1974) have measured the \((4s)^{-1}/(4p)^{-1}\) ratio up to approximately 40.8 eV.

In order to place these partial cross sections on an absolute scale, it is necessary to subtract the contribution of Kr\(^{2+}\), which has a threshold at 38.571 eV. Such a correction, using the data of Samson and Haddad was made before plotting the \((4s)^{-1}\) and \((4p)^{-1}\) partial cross section. A steep decline has been observed in both \((4p)^{-1}\) and \((4s)^{-1}\)
as the cooper minimum is approached, indicating a strong correlation between the 4s and the 4p lines. Similar behavior was noted in the case of the Ar 3s and the 3p lines. The data of Holland et al. (1978) indicate that the Kr$^+$ partial cross section remains small above 80 eV, perhaps displaying small oscillations in the vicinity of the (3d)$^{-1}$ threshold. The cross section for double ionization increases more or less linearly from threshold (Samson and Haddad, 1974), as expected. The ratio $\sigma$(Kr$^{2+}$)/$\sigma$(Kr$^+$) has a plateau between 50 eV < $h\nu$ < 60 eV of approximately 0.17, well below the threshold for(3d)$^{-1}$ at 93.83 eV. (Codling and modden, 1964). The double ionization cross section up to this energy must involve electron-electron correlation since shake off and Auger process are energetically forbidden. Before going any further, it is better to clarify what is meant by shake off and Auger process. When an electron is ejected from a multielectron atom, the remaining electrons suddenly find themselves in a different central potential. After the ensuing readjustment, there is probability that a given electron will find itself in a new state. If this is a higher excited state, the process is called "shake up", but if it is a continuum state it is known as "shake off". The Auger effect, on the other hand, is a two-electron process in which an electron makes a discrete transition from a less shell to vacant. There is a gap in our knowledge of partial cross sections until 277
eV, where the X-ray photoelectron spectroscopic studies of Krause (1969) are available.

Krause reports subshell ratios for the (4s)$^{-1}$, (4p)$^{-1}$, (3d)$^{-1}$, (3p)$^{-1}$ and (3s)$^{-1}$ at discrete X-ray lines between 277 and 1487 eV, measured at an angle of 90° with respect to the photon propagation direction. Multiple electron transitions have also been taken into account using the information in Krause's paper (1969).

The consequences of creating a vacancy in the n = 2 to n = 1 shells in Krypton have been investigated by Krause and Carlson (1967) experimentally and rationalized with a model that incorporates shake off and various alternative Auger and radiative decays. For the L-shell study, a titanium anode was used, producing primarily the 4510.84 eV TiK$_{\alpha}$ line the initial vacancy probability distribution was calculated to be 2s = 0.30; 2p = 0.563; 3s = 0.05; 3p = 0.07; 3d = 0.015; and 4s = 0.003, hence predominantly in the n = 2 shell. The experimentally observed change distribution is seen to have major components at Kr$^{4+}$, Kr$^{5+}$, Kr$^{6+}$, with mean charge of 5.22. To induce a primary vacancy in the K-shell, a molybdenum anode was used. It primarily generated the 17.479 KeV Mo K$_{\alpha}$ line, and this initial vacancy distribution was calculated to be 1s = 0.865; 2s = 0.083; 2p = 0.037; 3s = 0.007; 3p = 0.004; 3d = 0.002; and 4s, 4p = 0.002. The experimental and calculated charge distributions have a large contributions
between Kr$^{5+}$ and Kr$^{9+}$, with a mean charge of 6.0

The Angular Distribution

Kennedy and Manson (1972) have calculated the angular distribution parameter called $\theta$ (which is going to be explained in detail later) for Kr 3d and Manson (1973) has calculated it for Hg 5d. The rapid change in $\theta$ near threshold is due principally to the rapid variation in the Coulomb phase shift, as for np orbitals. The change in $\theta$ for the d orbitals is somewhat more rapid than for the p orbitals, a behavior which is attributable to the fact that the ratio of dipole matrix elements varies somewhat more rapidly in d-orbital ionization. The 3d orbitals have no node and hence no cooper minimum. As with 2p orbitals agreement between HS(Hatree-Slater) and HF(Hartree-Fock) calculation is very good. The high-energy asymptotic value of $\theta$ approaches a value slightly greater than 1, but apparently less than 1.5. Threshold and resonance behavior again proved very helpful in helping to verify the predictive power of the different theoretical approaches beyond the one-particle picture.

In this work, my advisor and her collaborators measured the 4s, 4p correlation satellite spectra for krypton with high statistical certainty. The photoelectron spectra induced by photon of energies between 29 and 250 eV were recorded at several angles to allow us to deduce:
1. The ratios of the satellites over the main lines 4s and 4p. These are proportional to the relative cross-section.

2. The photoelectron angular distribution parameter.

We present in this thesis the photoelectron spectra of the correlation satellites in krypton, displaying the relative cross-section data taken at angles of 0°, 54.9° and 90°, and finally the values of the β parameters for the Rydberg nd lines. The differential spectra which were recorded as a set, are corrected for the different responses of the two analyzers at 0° and 90° and have a generally small and nearly flat background subtracted.

The aim of this work is to present high-resolution results of photoionization in the outermost shell of the krypton in both graphical and tabular form. The spectra were acquired for several excitation energies. In these cases, it appeared important to test theoretical prediction and earlier experimental results with the high accuracy inherent in the present experiment. In partial fulfillment to my master's degree, I was responsible for the complete analysis of the presented results.
Synchrotron radiation started about 25 years ago to become an important tool for a wide range of basic and industrial research activities. The expansion of many of these activities was, besides the specific progress in accelerator techniques and synchrotron radiation instrumentation, dependent on new development in vacuum techniques. An illustrative example of such an interaction between two different technological fields are the gas-phase experiments with synchrotron radiation. At the beginning of the growing synchrotron radiation research field gas-phase experiments played only a marginal role. Until 10 years ago only a few groups performed larger gas-phase experiments with synchrotron radiation. This situation has changed dramatically since the construction and opening of a new generation of electron storage rings, specially designed for the purpose of synchrotron radiation generation, the so-called "dedicated light source", in the mid-eighties. Since that time, gas-phase experiments with synchrotron radiation experienced an unexpected growth and are now well established at most synchrotron radiation laboratories. This chapter describes the experimental
apparatus that was used in the gas-phase experiment performed. The obtained data are reported in the present thesis. Briefly, the apparatus consists of a rotating chamber that houses two electron time-of-flight spectrometers and is described below.

Rotating Chamber

The vacuum system consists of two main components, an experimental rotating chamber and a differential pumping section (also called the Window-section), which connects the chamber to the monochromator by means of the vacuum. The latter has to overpass a difference in pressure of $P_{\text{mono}} \leq 10^{-8}$ mbar in the monochromator and $P_{\text{th}} = 5 \times 10^{-5}$ mbar in the chamber during the measuring operation. The process is realized by the differential pumping section, which is made of two chambers connected by a capillary and correspondingly emptied with a turbo-molecular pump. For the chamber, a turbo pump of 1000 l/s was used, whereas for the monochromator a 350 l/s turbo pump was used. Another possibility to connect to the photon beam line is to put a window that separates the two vacua, but the VUV radiation is permeable (at least a part of it). If we insert an aluminum window, only the synchrotron light under the Al-edge of about 70 eV will pass through, while light of higher energy will be stopped. This feature has been used in order to eliminate the higher orders of the monochromators in certain cases.
The connection between the window section and the measuring chamber occurs by means of a differentially pump whirling execution and of an included second capillary. The whirling execution allows the whole measuring chamber together with the two detectors used for our measurement as well as the gas valve to be swung under vacuum conditions around the radiation as shown in Figure 1. The normal range of the angles is usually of 180°. Since the two detectors have an angle of 125.3° between them, they cover a total angle of about 305°. The disposition of the detectors allows that for an ideal linear polarization of the synchrotron radiation, one of the analyzers goes in the direction of the electric field $\vec{E}$, while the other detects the electrons at the magic angle (54.7° to the $\vec{E}$-field). This set-up can use either a gas valve or an oven which are located, because of symmetrical reasons, on the angles bisecting lines between the two analyzers. Normally the chamber is pumped with a turbo pump (1000 l/s). However, during the measuring operation using the oven the latter is replaced with a cryopump (900 l/s), in order to avoid metal depositions on the rotational curls and consequently damages the turbo-pump. For the present experiment, we only used the gas line.

The interior of the chamber is shielded with a cylinder of highly permeable $\mu$-metal, its function being to
Figure 1. Schematic of the Experimental Apparatus.
protect it against magnetic fields, especially the earth's magnetic field.

The Detectors

The main elements of our measuring equipment are the two time-of-flight spectrometers meant to be identical in construction, which will be called TOF (time-of-flight) analyzers. Figure 2 shows a schematic of the TOF used. The kinetic energy of the electrons emitted in the volumes of the interaction region or source as shown in Figure 2 has been established for the 679 mm length TOF (Becker and Wehlitz, 1993). The entry-diaphragm \( a_1 (\phi = 1.8 \text{ mm}) \) is 20 mm away from the source. It is connected, like the whole analyzer except for the stream-section, on mass potential. Immediately after, there is another diaphragm \( a_2 (\phi = 4.0 \text{ mm}) \), which is connected to another electric potential. This diaphragm serves especially to oven experiment, in order to prevent the slow thermic electrons with the help of negative alternating current (usually 1-2 V), to enter the analyzer. The diaphragm \( a_3 (\phi = 10.00 \text{ mm}) \) is the entry to the stream section, being electrically connected with it. A voltage induced therein \( (U_{\text{acc}} > 0 \text{ V}) \) speeds or slows down \( (U_{\text{acc}} < 0 \text{ V}) \) the electrons, which then move freely inside the stream section. Experiments with lens tensions showed that one can focus even electrons of certain kinetic energy and therefore increase their transmission. Unfortunately,
Figure 2. Schematic of Time of Flight Analyzer.
other electrons of different kinetic energy are defocused and their information to the microchannel-plates impeded. This leads to a very irregular transmission function for the analyzer, in which an important part of the flight's time method is lost, namely the "smooth" transmission up to the small \( E_{\text{kin}} = 0.5 \text{ eV} \) kinetic energies. Diaphragm \( a (\phi = 20 \text{ mm}) \) is installed in order to limit the radiation so that electrons could be sent only directly to the microchannel plates (MCPs). Secondary electrons, which are a result of the collisions of the primary electrons with the wall, are consequently selected/eliminated in a gradual manner. The position of this diaphragm is critical. If it is too far from the source point, it would reduce the number of indicated electrons. If it is too close to the source point, there would be 'mountains' in the spectrum, which could seriously encumber a precise evaluation of it.

At the end of the stream section, there is a detector. The electrons with a voltage of about 1000 V are accelerated and detected through a chevron-built-up microchannel-plate MCP. The signal appears as an energy pulse on the anode. The acceleration is necessary, because all electrons including the slow ones at their meeting in the microchannel-plate have a kinetic energy. The MCPs we used have an active surface of 1452 \( \text{mm}^2 \).

Around the whole stream section there is a magnetic field shielding (\( \mu \)-metal). The stream section itself is
also pumped with a 50 l/s turbomolecular pump. This protects on the one hand the MPCs of high pressure, which in turn leads to higher darkness in the spectrum, and reduces on the other hand the number of collision of electrons with the remaining gas.

The choice of accelerating and decelerating electrons, and therefore voltages, depends on the experiment and on the time interval of the synchrotron radiation. At BOSSY, the Berlin synchrotron radiation source, the "Single-Bunch" operation has a bunch-deviation of 208 ns. In our experiment we use the "single-bunch" operation as a timing structure in order to stop our electronics. The timing structure is used to give us a "time window" since we are measuring the time-of-flight of the electrons.

The energy resolution is given by the length of the working section and the size of the interaction volume. The energy resolution of the analyzer \( \Delta E \) involves 1% of the kinetic energy of the electrons \( E_e = E_{kin} - eU_{acc} \). The decomposition possibilities of the analyzer \( \Delta E/E_{kin} \) could be modified with the acceleration voltage \( U_{acc} \):

\[
\frac{\Delta E}{E_{kin}} = \frac{\Delta E}{E_e + eU_{acc}}
\]

giving: \( \frac{\Delta E}{E_e} = 1\% \)
The Measuring Electronics

Both micro-channel plates at the end of the detector are built in a tandem fashion. If an accelerated electron hits the first MCP, an avalanche results, that is strengthened by the second MCP. The current amplification on one MCP is of the order of \((1-5) \times 10^4\). The electron’s avalanche is disconnected by a capacitor and produces a voltage impulse of about 10-30 mV and 2 ns in length. Figure 3 shows the associated electronics to our detection system.

The voltage pulse is amplified through a two-stage rapid preamplifier to about a factor of 50-100, before being sent through a discriminator (constant fraction discriminator, CFD). The MCP pulses spread in a strong way in their size but remain similar in form. A simple discriminator that would react only to the impulse’s height, would not reach the necessary time precision (<100 ps). In the CFD the coming impulse is added to an inverse impulse, delayed by \(\Delta t=0.6\) ns, which has a constant amplitude relation to the original impulse. The norm impulse starts then at the zero passage of the combined signal. This signal from the CFD is sent to the start of a Time-to-Amplitude-Converter (TAC). The stop signal of the TAC is provided by the bunch-marker signal from the synchrotron radiation ring. The TAC discharges an impulse, whose height is proportional to the time between the starting and the stopping impulse. The latter is then transformed from
Figure 3. Schematic of the Detection and Associated Electronics.
the analogs to digital convertor in a number corresponding to the impulse's height and transmitted to the computer together with the values of the synchrotron radiation current, and the gas pressure.

The spectra contain also a background noise that originate from the MCP detectors, which are not produced by the electrons issued in the source volume. This noise is not related to the synchrotron impulse and spreads all over the whole spectrum. We found that lowering this noise depends on the voltage of the micro-channel plates and on the gas pressure in the detector.

**The Radiation Monitor**

The radiation monitor's function, as shown in Figure 1, is to measure the number of photons and the radiation current, respectively. This occurs because the synchrotron light behind the interaction volume are intercepted by a steel diode, which releases electrons through the photoelectric effect. The latter could be observed as a current, which is proportional to the number of photons and integrated during the whole measuring operation of the spectrum on the radiated photon energy. Unfortunately, the radiation monitor can not distinguish between photons of the first order and photons of the higher orders. It is therefore necessary to know the higher orders of the beamline, if one wants to calibrate the spectra with the
current monitor. For the kind of measurement we made for noble gasses, the radiation monitor was used only as a control mean, not as an absolute mean.
CHAPTER III

DATA ANALYSIS AND RESULTS

In this thesis we report on the first systematic high resolution photoelectron spectrometry study for the nd(2S_{1/2}) (n = 4-9) satellites in Kr covering the photon energy from 68.5 to 250 eV. The experiment was performed at the Hamburger Synchrotronstrahlungsabor (HASYLAB) on the new undulator beam line (BW3) utilizing a high-resolution SX-700 monochromator (Møller 1993). The measurements were made during the single-bunch and double-bunch operation of the electron storage ring. Spectra were recorded using two time-of-flight (TOF) electron spectrometers with a nominal resolving power of 100. Both analyzers are mounted on a rotatable chamber perpendicular to the axis of the incoming synchrotron radiation to allow for the determination of angular distributions as described in Chapters I and II.

The high flux, high-resolution combination of this beam line allowed us to record the highest resolution spectra yet of the Kr valence satellite region using monochromatized synchrotron radiation. Representative spectra, recorded at 68.5 eV are shown in Figure 4, for three different angles of the electron analyzer. As the resolution of the time of flight spectrometer varies
Figure 4. High Resolution Photoelectron Spectrum of the Kr Correlation Satellites as Recorded at 68.5 eV for Three Different Angles.
linearly as a function of kinetic energy, the full width half maximum (FWHM) for every line in this spectrum is different. All spectra have been completely corrected for transmission effects, differences in detector efficiencies and source volume anisotropies. The middle panel of this figure corresponds to the so-called "magic angle" where the intensities of the features are independent of angular distribution effects. Satellites in this panel are designated according to the scheme used by Krause et al (1992). The assignments of these lines are based on the semi-empirical calculations of Hansen and Persson (1987) whose binding energies were derived from optical data. The \( nd(2S_{1/2}) \) satellites, which are the main interest of this thesis, result entirely from electron-electron correlation. They are indicated in the 0° spectrum.

Representative spectra, recorded at 0° for three different energies are shown in Figure 5. We find that, with increasing energy the intensity of the 4p main line remains the same while the intensity for the 4s main line decreases. The \( nd(2S_{1/2}) \) satellites intensities, like 4s main line, decrease with increasing energies.

One of the ratios for the \( nd(2S_{1/2}) \) with respect to the 4p main line is displayed as a function of angular distribution in Figure 6. The value of \( \beta \) of the 4d satellite with respect to the 4p line is obtained and is close to 2. The \( \beta \) reference indicate the \( \beta \) for the 4p main line used in
Figure 5. High Resolution Photoelectron Spectrum of the Kr Correlation Satellites as Recorded at Three Different Energies for 0° Angle.
Figure 6. Schematic of the Ratio for the Satellite 4d over 4p as a Function of the Angle.
order to extract the one for the 4d. The value of the ratios correspond to the partial cross section of the satellite 4d with respect to the 4p main line. It is obtained by dividing the fitted area of satellite 4d by the fitted area of the 4p main line. According to the relation, giving in the dipole approximation (Krause et al., 1992).

\[ \frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} \left[ 1 + \frac{\beta}{4} (1 + 3pcos\theta) \right] \]

the relative partial cross section \( \sigma \) for the photoionization process can be obtained from the intensity of the photoelectrons observed at the magic angle (\( \theta = 54.7^\circ \)), and the dipole angular distribution parameter \( \beta \) for the photoelectron can be derived from a measurement made at two different angles, preferably \( \theta = 0^\circ \) and \( \theta = 90^\circ \). The degree of polarization \( p \) was determined preferentially from the He 1s and the Ne 2s photolines, recorded at \( \theta = 0^\circ \) and \( \theta = 90^\circ \), with \( \beta = 2 \) for these s photoelectrons. Optimum accuracy could then be achieved, and in our experiment we extract \( p = 97\% \) for the linear polarization of the synchrotron radiation light.

Our results for the nd(\( ^2S_{1/2} \)) satellites intensities for \( n = 4 \) to 6 are displayed in Figures 7,8 and 9 respectively. We notice two important trends in the experimental results of Figures 7,8 and 9 when compared to theoretical predictions. The first is, according to the configuration
Figure 7. Schematic of $\beta$ and Satellite 4d with Respect to 4s and 4p as a Function of Photon Energy.
Figure 8. Schematic of $\beta$ and Satellite 5d With Respect to 4s and 4p as a Function of Photon Energy.
Figure 9. Schematic of $\beta$ and Satellite 6d with Respect to 4s and 4p as a Function of Photon Energy.
interaction picture, these satellites should have $\beta$ values identically equal to two for all photon energies. Our measurement show in Figures 7,8 and 9, that $\beta$ is almost equal to 2. Secondly, the ratios of the satellites nd($^2S_{1/2}$), ($n = 4-6$) over the 4s main line should be independent of photon energy. Careful depiction of Figures 7,8 and 9 lead us, however, to infer that the ratios might be slightly correlated to the photon energy. This should require further experimental investigations. However the present data confirm the latest predictions for all members of the series at all energies (Tulkki et al. 1992). Figures 7,8 and 9 also agree with earlier experimental data at 68.5 eV by Krause (Krause et al. 1992).

Numerical data from our analysis are summarized in Tables 1 to 10. Assignment and corresponding binding energies are taken from Minnhagen et al. (1968) and are tabulated in the first 3 columns. The $\beta$ average in Table 2, is calculated by averaging the $\beta$ for each satellite with respect to 4s and 4p main lines. Our binding energies are in excellent agreement with krause et al. (1992) at 68.5 eV. Values of $\beta$ close to two are obtained for the 4d and 5d lines, indicating small contributions to these members from overlapping satellite lines with small $\beta$ values. The ratios and the $\beta$ parameters are generally decreasing with increasing binding energy. In comparing our results with calculations of relative intensities for the 4d and the 5d
lines we find that they are overestimated by an extensive configuration interaction calculation (Smid and Hansen, 1983). This is in contrast to the findings for analogous transitions in argon, where good agreement is noted.

The error of our results is approximately 7% on the ratios and 15% on $\beta$ parameter due to:

1. Different transmission function for different kinetic energies although we did correct for transmission functions of the spectrometer.

2. Error in determining the polarization which subsequently effects the value of $\beta$ parameter.

3. The fitting for all the peaks may not be perfect, although we fitted each of them individually with a gaussian function.

Table 1

Kr 4s,4p Correlation Satellites (hv = 68.5 eV)
Ratios/4s

<table>
<thead>
<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4s</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4s</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>2.00</td>
</tr>
<tr>
<td>($^{1}D$)4d($^{2}S_{1/2}$)</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.164</td>
<td>1.87</td>
</tr>
<tr>
<td>($^{1}D$)5d($^{2}S_{1/2}$)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.048</td>
<td>1.88</td>
</tr>
<tr>
<td>($^{1}D$)6d($^{2}S_{1/2}$)</td>
<td>37.82</td>
<td>37.82</td>
<td>0.028</td>
<td>1.51</td>
<td></td>
</tr>
<tr>
<td>($^{1}D$)7d($^{2}S_{1/2}$)</td>
<td>38.58</td>
<td>38.58</td>
<td>0.020</td>
<td>0.52</td>
<td></td>
</tr>
<tr>
<td>($^{1}D$)8d($^{2}S_{1/2}$)</td>
<td>39.04</td>
<td>39.04</td>
<td>0.010</td>
<td>0.61</td>
<td></td>
</tr>
<tr>
<td>($^{1}D$)9d($^{2}S_{1/2}$)</td>
<td>39.33</td>
<td>39.33</td>
<td>39.30</td>
<td>0.005</td>
<td>0.41</td>
</tr>
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</table>
Table 2

**Kr 4s,4p Correlation Satellites (hv = 68.5 eV)**

<table>
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<tr>
<th>Assignment</th>
<th>This work</th>
<th>Minn-hagan</th>
<th>Ratios/4p</th>
<th>β</th>
<th>βav.</th>
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<tbody>
<tr>
<td>4p</td>
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<td>14.22</td>
<td>1.000</td>
<td>0.34</td>
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<tr>
<td>(1D) 4d(2S\textsubscript{1/2})</td>
<td>33.96</td>
<td>33.94</td>
<td>0.042</td>
<td>1.98</td>
<td>1.92</td>
</tr>
<tr>
<td>(1D) 5d(2S\textsubscript{1/2})</td>
<td>36.47</td>
<td>36.47</td>
<td>0.017</td>
<td>1.98</td>
<td>1.93</td>
</tr>
<tr>
<td>(1D) 6d(2S\textsubscript{1/2})</td>
<td>37.82</td>
<td>0.009</td>
<td>1.75</td>
<td>1.63</td>
<td></td>
</tr>
<tr>
<td>(1D) 7d(2S\textsubscript{1/2})</td>
<td>38.58</td>
<td>0.066</td>
<td>0.93</td>
<td>0.73</td>
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</tr>
<tr>
<td>(1D) 8d(2S\textsubscript{1/2})</td>
<td>39.04</td>
<td>0.034</td>
<td>1.02</td>
<td>0.81</td>
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</tr>
<tr>
<td>(1D) 9d(2S\textsubscript{1/2})</td>
<td>39.33</td>
<td>39.30</td>
<td>0.019</td>
<td>0.84</td>
<td>0.62</td>
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</table>

Table 3

**Kr 4s,4p Correlation Satellites (hv = 84.8 eV)**

<table>
<thead>
<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4s</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4s</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D) 4d(2S\textsubscript{1/2})</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.191</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D) 5d(2S\textsubscript{1/2})</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.079</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D) 6d(2S\textsubscript{1/2})</td>
<td>37.82</td>
<td>37.82</td>
<td>0.034</td>
<td>1.91</td>
<td></td>
</tr>
<tr>
<td>(1D) 7d(2S\textsubscript{1/2})</td>
<td>38.58</td>
<td>38.58</td>
<td>0.020</td>
<td>1.89</td>
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<tr>
<td>(1D) 8d(2S\textsubscript{1/2})</td>
<td>39.04</td>
<td>39.04</td>
<td>0.010</td>
<td>1.88</td>
<td></td>
</tr>
<tr>
<td>(1D) 9d(2S\textsubscript{1/2})</td>
<td>39.33</td>
<td>39.33</td>
<td>39.30</td>
<td>0.005</td>
<td>2.00</td>
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</table>
Table 4
Kr 4s,4p Correlation Satellites (hv = 84.8 eV)
Ratios/4p

<table>
<thead>
<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4p</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4p</td>
<td>14.22</td>
<td>14.22</td>
<td>14.22</td>
<td>1.000</td>
<td>4.110^{-3}</td>
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<tr>
<td>(1D)4d(^2S_{1/2})</td>
<td>33.96</td>
<td>33.9</td>
<td>33.94</td>
<td>0.072</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D)5d(^2S_{1/2})</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.030</td>
<td>2.00</td>
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<tr>
<td>(1D)6d(^2S_{1/2})</td>
<td>37.82</td>
<td>37.82</td>
<td>0.013</td>
<td>1.87</td>
<td></td>
</tr>
<tr>
<td>(1D)7d(^2S_{1/2})</td>
<td>38.58</td>
<td>38.58</td>
<td>0.007</td>
<td>1.84</td>
<td></td>
</tr>
<tr>
<td>(1D)8d(^2S_{1/2})</td>
<td>39.04</td>
<td>39.04</td>
<td>0.004</td>
<td>1.85</td>
<td></td>
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<tr>
<td>(1D)9d(^2S_{1/2})</td>
<td>39.33</td>
<td>39.33</td>
<td>0.002</td>
<td>2.00</td>
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</table>

Table 5
Kr 4s,4p Correlation Satellites (hv = 98.0 eV)
Ratios/4s

<table>
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<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4s</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4s</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>2.00</td>
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<tr>
<td>(1D)4d(^2S_{1/2})</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.216</td>
<td>1.98</td>
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<tr>
<td>(1D)5d(^2S_{1/2})</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.097</td>
<td>1.96</td>
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<tr>
<td>(1D)6d(^2S_{1/2})</td>
<td>37.82</td>
<td>37.82</td>
<td>0.041</td>
<td>1.75</td>
<td></td>
</tr>
<tr>
<td>(1D)7d(^2S_{1/2})</td>
<td>38.58</td>
<td>38.58</td>
<td>0.020</td>
<td>1.99</td>
<td></td>
</tr>
<tr>
<td>(1D)8d(^2S_{1/2})</td>
<td>39.04</td>
<td>39.04</td>
<td>0.009</td>
<td>2.00</td>
<td></td>
</tr>
<tr>
<td>(1D)9d(^2S_{1/2})</td>
<td>39.33</td>
<td>39.33</td>
<td>0.006</td>
<td>1.56</td>
<td></td>
</tr>
</tbody>
</table>
### Table 6

Kr 4s,4p Correlation Satellites (hv = 98.0 eV)
Ratios/4p

<table>
<thead>
<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4p</th>
<th>β</th>
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</thead>
<tbody>
<tr>
<td>4p</td>
<td>14.22</td>
<td>14.22</td>
<td>14.22</td>
<td>1.000</td>
<td>0.21</td>
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<td>(^1D) 4d(2^2S_1/2)</td>
<td>33.96</td>
<td>33.9</td>
<td>33.94</td>
<td>0.077</td>
<td>1.97</td>
</tr>
<tr>
<td>(^1D) 5d(2^2S_1/2)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.034</td>
<td>1.97</td>
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<td>(^1D) 6d(2^2S_1/2)</td>
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<td>37.82</td>
<td>37.82</td>
<td>0.014</td>
<td>1.80</td>
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<td>(^1D) 7d(2^2S_1/2)</td>
<td>38.58</td>
<td>38.58</td>
<td>38.58</td>
<td>0.007</td>
<td>2.00</td>
</tr>
<tr>
<td>(^1D) 8d(2^2S_1/2)</td>
<td>39.04</td>
<td>39.04</td>
<td>39.00</td>
<td>0.003</td>
<td>2.00</td>
</tr>
<tr>
<td>(^1D) 9d(2^2S_1/2)</td>
<td>39.33</td>
<td>39.33</td>
<td>39.30</td>
<td>0.002</td>
<td>0.94</td>
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</table>

### Table 7

Kr 4s,4p Correlation Satellites (hv = 150.0 eV)
Ratios/4s

<table>
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<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4s</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4s</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>2.00</td>
</tr>
<tr>
<td>(^1D) 4d(2^2S_1/2)</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.185</td>
<td>1.93</td>
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<tr>
<td>(^1D) 5d(2^2S_1/2)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.069</td>
<td>1.96</td>
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<tr>
<td>(^1D) 6d(2^2S_1/2)</td>
<td>37.82</td>
<td>37.82</td>
<td>37.82</td>
<td>0.026</td>
<td>1.88</td>
</tr>
<tr>
<td>(^1D) 7d(2^2S_1/2)</td>
<td>38.58</td>
<td>38.58</td>
<td>38.58</td>
<td>0.014</td>
<td>1.90</td>
</tr>
<tr>
<td>(^1D) 8d(2^2S_1/2)</td>
<td>39.04</td>
<td>39.04</td>
<td>39.00</td>
<td>0.007</td>
<td>2.00</td>
</tr>
<tr>
<td>(^1D) 9d(2^2S_1/2)</td>
<td>39.33</td>
<td>39.33</td>
<td>39.30</td>
<td>0.006</td>
<td>2.00</td>
</tr>
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</table>
### Table 8
Kr 4s,4p Correlation Satellites (hv = 150.0 eV)
Ratios/4p

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<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4p</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4p</td>
<td>14.22</td>
<td>14.22</td>
<td>14.22</td>
<td>1.000</td>
<td>0.94</td>
</tr>
<tr>
<td>(1D)4d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>33.96</td>
<td>33.9</td>
<td>33.94</td>
<td>0.078</td>
<td>1.85</td>
</tr>
<tr>
<td>(1D)5d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.029</td>
<td>1.89</td>
</tr>
<tr>
<td>(1D)6d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>37.82</td>
<td>37.82</td>
<td>37.82</td>
<td>0.013</td>
<td>1.79</td>
</tr>
<tr>
<td>(1D)7d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>38.58</td>
<td>38.58</td>
<td>38.58</td>
<td>0.006</td>
<td>1.84</td>
</tr>
<tr>
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<td>39.04</td>
<td>39.04</td>
<td>39.04</td>
<td>0.003</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D)9d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
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<td>39.33</td>
<td>39.30</td>
<td>0.002</td>
<td>2.00</td>
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</tbody>
</table>

### Table 9
Kr 4s,4p Correlation Satellites (hv = 250 eV)
Ratios/4s

<table>
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<tr>
<th>Assignment</th>
<th>This work</th>
<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4s</th>
<th>β</th>
</tr>
</thead>
<tbody>
<tr>
<td>4s</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D)4d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.220</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D)5d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.468</td>
<td>2.00</td>
</tr>
<tr>
<td>(1D)6d(2S&lt;sub&gt;1/2&lt;/sub&gt;)</td>
<td>37.82</td>
<td>37.82</td>
<td>0.997</td>
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</tbody>
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### Table 10

Kr 4s,4p Correlation Satellites (hv = 250 eV)

<table>
<thead>
<tr>
<th>Assignment</th>
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<th>Krause</th>
<th>Minn-hagan</th>
<th>Ratios/4p</th>
<th>$\beta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>4p</td>
<td>27.51</td>
<td>27.51</td>
<td>27.51</td>
<td>1.000</td>
<td>1.27</td>
</tr>
<tr>
<td>($^1D$)4d($^2S_{1/2}$)</td>
<td>33.96</td>
<td>33.96</td>
<td>33.94</td>
<td>0.062</td>
<td>2.00</td>
</tr>
<tr>
<td>($^1D$)5d($^2S_{1/2}$)</td>
<td>36.47</td>
<td>36.47</td>
<td>36.47</td>
<td>0.023</td>
<td>2.00</td>
</tr>
<tr>
<td>($^1D$)6d($^2S_{1/2}$)</td>
<td>37.82</td>
<td>37.82</td>
<td></td>
<td>0.005</td>
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</table>
CHAPTER IV

CONCLUSION

Electron correlation satellites, virtually unknown 25 years ago and poorly resolved only 5 years ago, can now be delineated in great detail. This is achieved using photoelectron spectrometry coupled to an undulator high-resolution monochromator photon source. The spectra presented in this work give a clear overview of the satellite production in krypton, photoionized in the outer principal shell. Ratios and $\beta$ parameter data derived from the high-resolution spectra resolved several open questions and generally confirmed previous interpretations of the nature of the satellites. The data also indicate a need for more theoretical work to fully account for the observations and to fully understand the various configuration and channel interactions at work. Although the resolution we achieved in this study is unprecedented, it is neither the ultimate limit that can be reached nor is it high enough to resolve all features appearing in a correlation satellite spectrum.

These results indicate the need for additional, more sophisticated calculations, as well as better resolved satellite spectra in the region of high energy limit.
To approach the limit imposed by the natural width of atomic levels, advances on the side of the photon source must be complemented by improvements of the users' apparatus, namely, the electron spectrometer used photoemission studies.


Madden, R. P. & Codling, K. (1964). Recently discovered


