Argon-Ion Charge Distributions Following Near-Threshold Ionization

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When an atom is photoionized in an inner shell, there are two mechanisms by which the remaining electron cortege relaxes to fill the vacancy: x-ray emission and radiationless Auger and Coster-Kronig transitions. In the former, the inner-shell hole moves to a less tightly bound orbital without increasing the number of atomic vacancies. In Auger processes, however, the energy liberated by transfer of a less-tightly-bound electron to the inner-shell vacancy is transferred to another electron which is ejected into the continuum. In this case, the charge on the residual ion increases by one. Through a series of radiative and non-radiative processes, the initial vacancy bubbles up until all vacancies arrive at the outermost shell. Due to the many possible routes by which this may occur, there can be a broad distribution of residual ion charge states characteristic of the decay of a single inner-shell vacancy.

There have been several measurements of ion charge distributions following inner-shell photoionization. Using photons from x-ray tubes to produce K vacancies in argon well above absorption edges, Carlson and Krause measured the resultant charge distributions with a magnetic analyzer.[1] More recently, Tonuma et al have used time-of-flight techniques to measure charge distributions of xenon ions resulting from synchrotron-radiation photoionization of L_{123} electrons.[2] The results showed a clear dependence on photon energy and L-subshell ionization thresholds.

Both Carlson and Krause and Tonuma et al made comparisons of their measurements with the results of Monte-Carlo simulations of charge distributions. The former authors used published radiative and nonradiative transition probabilities and, in addition, included estimates of shakeoff calculated in the sudden limit. The recent simulations of Tonuma et al employ more accurate calculations of radiative[3] and nonradiative[4] transition probabilities and the sudden-approximation estimates of Carlson and Nestor.[5] Both calculations show general agreement with the data.

A number of effects have been omitted from both theoretical treatments, however. As the atom autoionizes, outer-shell electrons see a reduced nuclear screening, resulting in increased binding energy. As a result, some channels may become closed. Decay rates change as the supply of outer-shell electrons is depleted. Well above the photoionization threshold, the sudden-approximation calculations of Carlson and Nestor provide a good description of shakeoff. As the energy of the photoionizing x ray approaches threshold from above, the low-energy photoelectron remains in the vicinity of the atom when the inner-shell hole decays, and in this regime the two-step excitation-decay picture is not valid. The threshold energy dependence of shakeoff has been measured by Armen et al[6] and was found to rise gradually as the high-energy asymptotic limit is approached. These effects all contribute to a complicated ion charge distribution which is strongly photon-energy dependent. The differences between theory and experiment are typically small for low charge states for which only a few processes contribute. The production of very high charge states of, e.g., xenon, can occur by a large variety of channels and for these charge states theory typically overestimates their relative probability.[7]

Because so many processes can contribute to each charge state, it is difficult to determine the effect of each by examining the total ion charge distribution; the total-ion charge distribution represents an average over many effects. To overcome this limitation, we have recently measured argon-ion production as a function of both photon energy and Auger decay channel following photoionization of K-shell electrons with highly monochromatic synchrotron radiation. When measured differential in decay channel, the ion charge distributions are greatly simplified. Analysis, in progress, of these simplified distributions will permit extraction of information about relative decay rates and shakeoff effects that is obscured in the singles spectra.

When normalized to constant photon flux, some interesting threshold effects can be seen in the charge distributions measured coincident with K-$L_{23}L_{23}$ Auger decay. Since virtually all L_{23} vacancies decay by L-MM Auger transitions, argon ions should be produced predominantly in charge state 4^+. Well above threshold there is, in addition, a substantial component of Ar^{5+} resulting from shakeoff and L-MMM double-Auger processes. At energies several hundred eV above the K photoionization threshold, the Ar^{5+} to Ar^{4+} ratio has reached an asymptotic value of ≈ 0.5. A large component of Ar^{3+} is evident in the energy region centered around the argon 4p resonance about 3 eV below the K-shell photoionization threshold in the K-shell absorption spectrum. This is due to capture of the K electron to a bound 4p orbital. There is a suggestion in the Ar^{3+} data of capture of the K electron to bound 5p and 6p states also. Similar data have been obtained at many energies for the other Auger transitions responsible for filling the initial K vacancy and exhibit features similar to those shown for the K-$L_{23}L_{23}$ coincident data. These pronounced shifts in charge state are not evident in spectra not obtained in coincidence.

The experiment required about eight days of beam time, including four shifts of timing-mode operation, and was performed in December, 1989, on NSLS beamline X-24A. Our apparatus consisted of a time-of-flight spectrometer to detect argon ions and a cylindrical mirror analyzer to select particular Auger electrons. Data were collected using the X-24A PDP computer. The experiment was prepared at the Oak Ridge National Laboratory in laboratories occupied by the University of Tennessee accelerator-based atomic physics group and transported to NSLS. Our gas-phase experiment was isolated from ring vacuum by a beryllium window.

We propose to continue experiments very similar to that discussed above using gaseous xenon. The experimental apparatus will remain virtually unchanged. In subsequent experiments, we hope to provide differential pumping to permit removal of the beryllium window and access to photon energies lower that the 3 keV window cutoff. This will permit extension of these experiments to the L edge of krypton and the K edges of neon and helium.

1. T. A. Carlson and M. O. Krause, Phys. Rev. **137**, A1655, 1965.

2. T. Tonuma, A. Yagishita, H. Shibata, T. Koizumi, T. Matsuo, K. Shima, T. Mukoyama, and H. Tawara, J. Phys. B: At. Mol. Phys. **20**, L31-L36, 1987.

3. J. H. Scofield, At. Data Nucl. Data Tables **14**, 121, 1974.

4. M. H. Chen, B. Crasemann, and H. Mark, At. Data Nucl Data Tables **24**, 13, 1979.

5. T. A. Carlson and C. W. Nestor, Jr., Phys. Rev. A **8**, 2887, 1973.

6. G. B. Armen, T. Åberg, K. R. Karim, J. C. Levin, B. Crasemann, G. S. Brown, M. H. Chen, and G. E. Ice, Phys. Rev. Lett. **54**, 182, 1985.

7. T. Mukoyama, Journal of the Physical Society of Japan, Vol. 55, No. 9, 3054, 1986.

ARGON - ION CHARGE DISTRIBUTIONS FOLLOWING NEAR - THRESHOLD PHOTOIONIZATION

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MOTIVATIONS

B. CRASEMANN	:	INNER - SHELL THRESHOLD PHENOMENA
S. MANSON	:	EDGES WELL KNOWN GLOBALLY, NOT LOCALLY
G. WENDIN	:	AUGER PROCESSES, RELAXATION, SHAKE-UP FROM INNER HOLES
M. KRAUSE	:	AUGER SPECTROSCOPY ON 2ND DECAYS - RELAXATION COINCIDENCE BETWEEN X RAYS AND AUGER ELECTRONS
M. KIMURA	:	CHARGE ANALYSIS OF PHOTOIONS COINCIDENCE BETWEEN EJECTED PARTICLES (PHOTONS, IONS, e^-)

ION CHARGE DISTRIBUTIONS

EXPERIMENT:

	Carlson & Krause	(1960's)	Many papers
	Lightner *et al.*	(1971)	Phys. Rev. A 4 Many papers
SR -->	Hastings & Kostroun	(1983)	NIM 208
SR -->	Tonuma *et al.*	(1987)	J. Phys. B 20

THEORY:

Carlson & Krause		Many papers (Shakeoff)
Mukoyama	(1985)	Bull. Inst. Chem. Res., Kyoto U.
Mirakhmedov & Parilis	(1988)	J. Phys. B (Relaxation)

THEORY MUST INCLUDE:

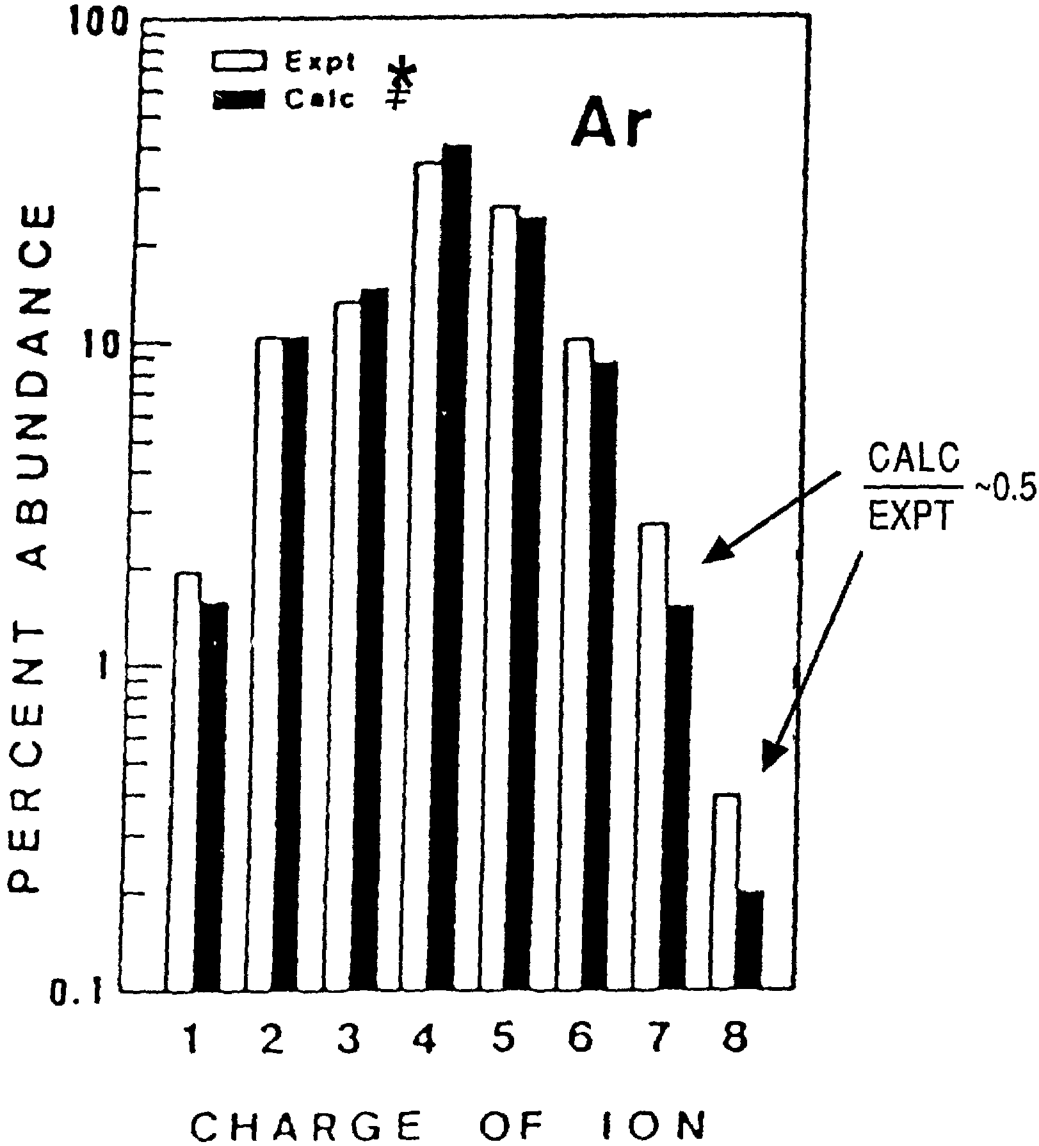
Radiative, Auger, Coster - Kronig Cross Sections

Shakeoff

Relaxation

Change in Auger Rates Due to Depletion of Outer Electrons

Closing of Some Channels

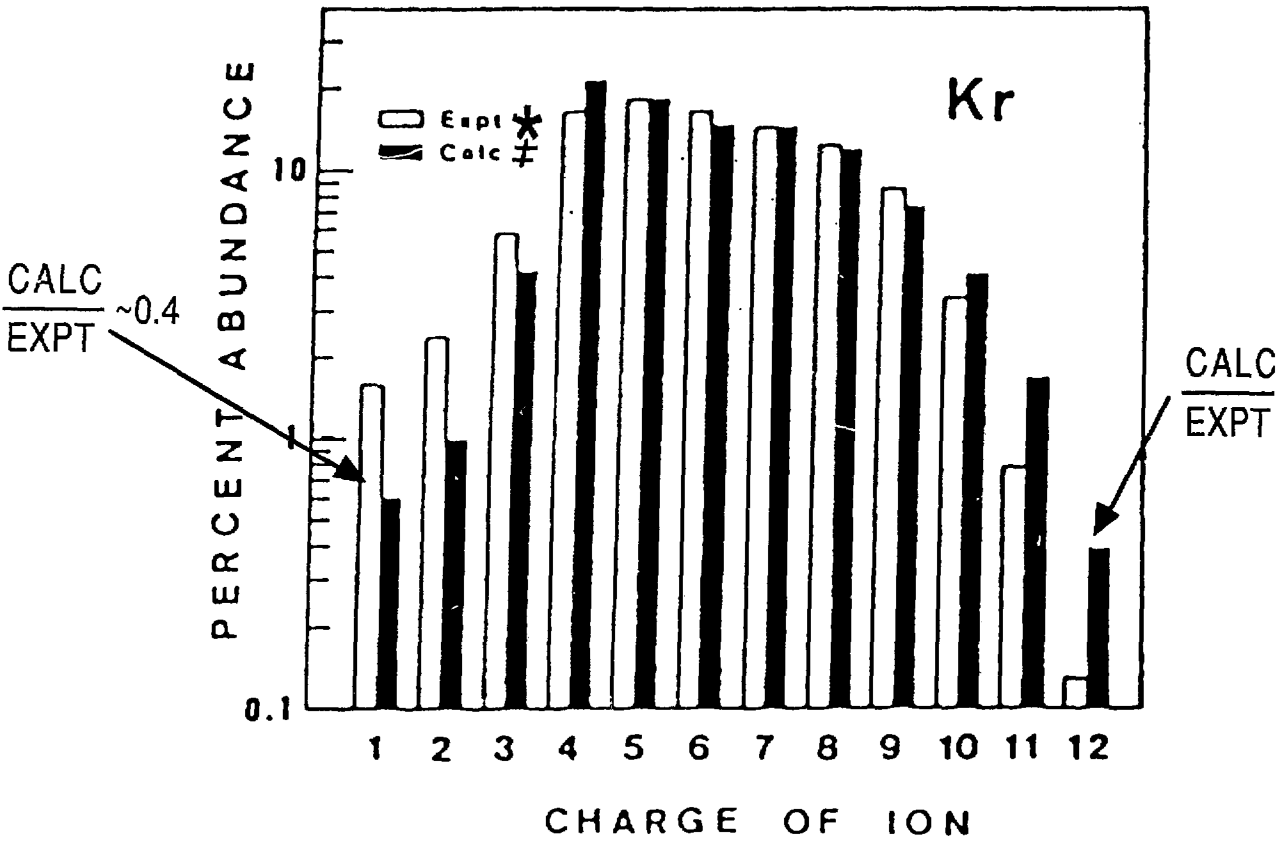


* Carlson & Krause, Phys. Rev. 137 (1965)

≠ Mukoyama, Bull. Inst. Chem. Res., Kyoto U. (1985)

* K vacancies produced by x-ray tubes, filters

≠ Monte Carlo simulation



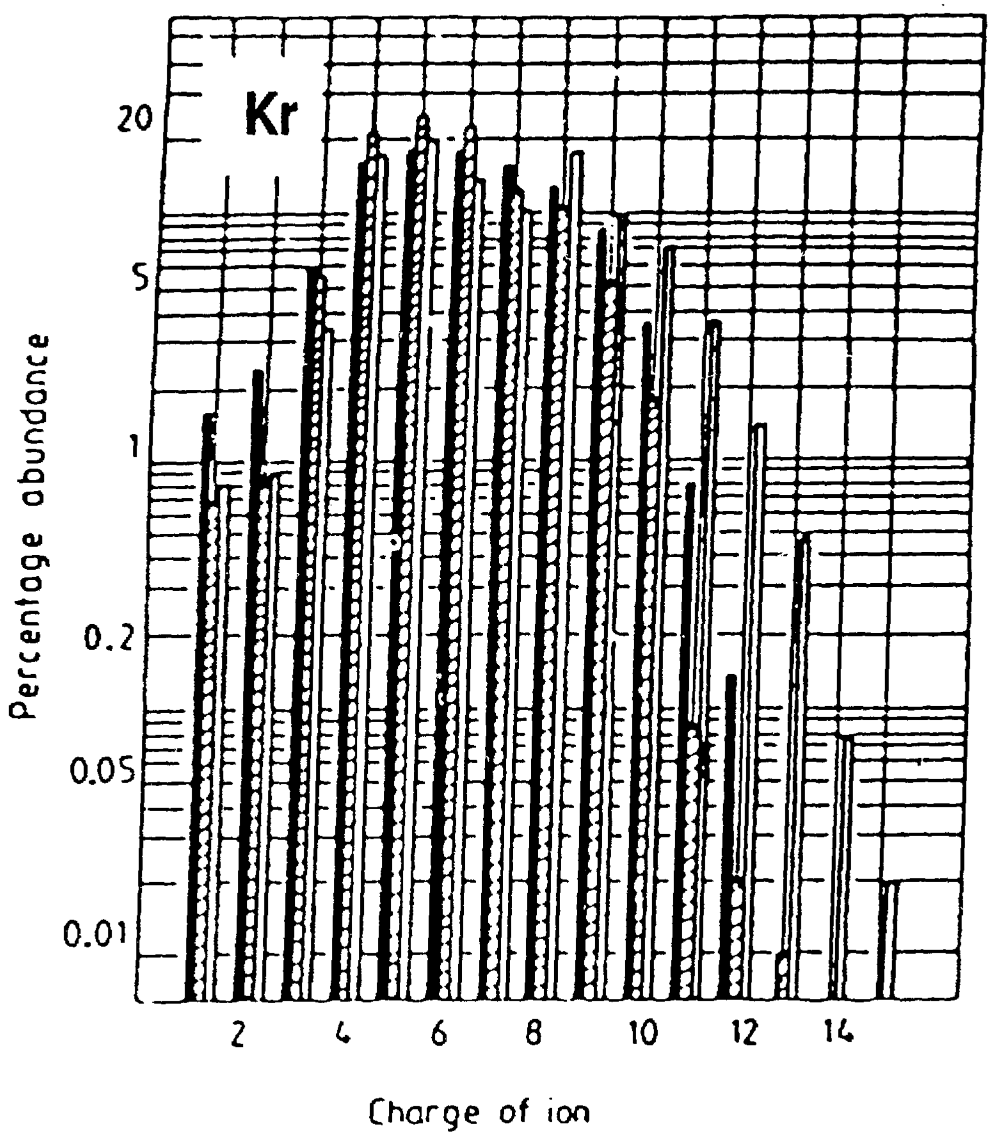
* Krause & Carlson, Phys. Rev. 158 (1967)

≠ Mukoyama, Bull. Inst. Chem. Res., Kyoto U. (1985)

* K vacancies using tubes / filters

≠ Monte Carlo simulation

From Mirakhmedov & Parilis, J. Phys. B 21, 795 (1988)



Krause & Carlson, Phys. Rev. 158 (1967)

Monte Carlo

Monte Carlo with relaxation

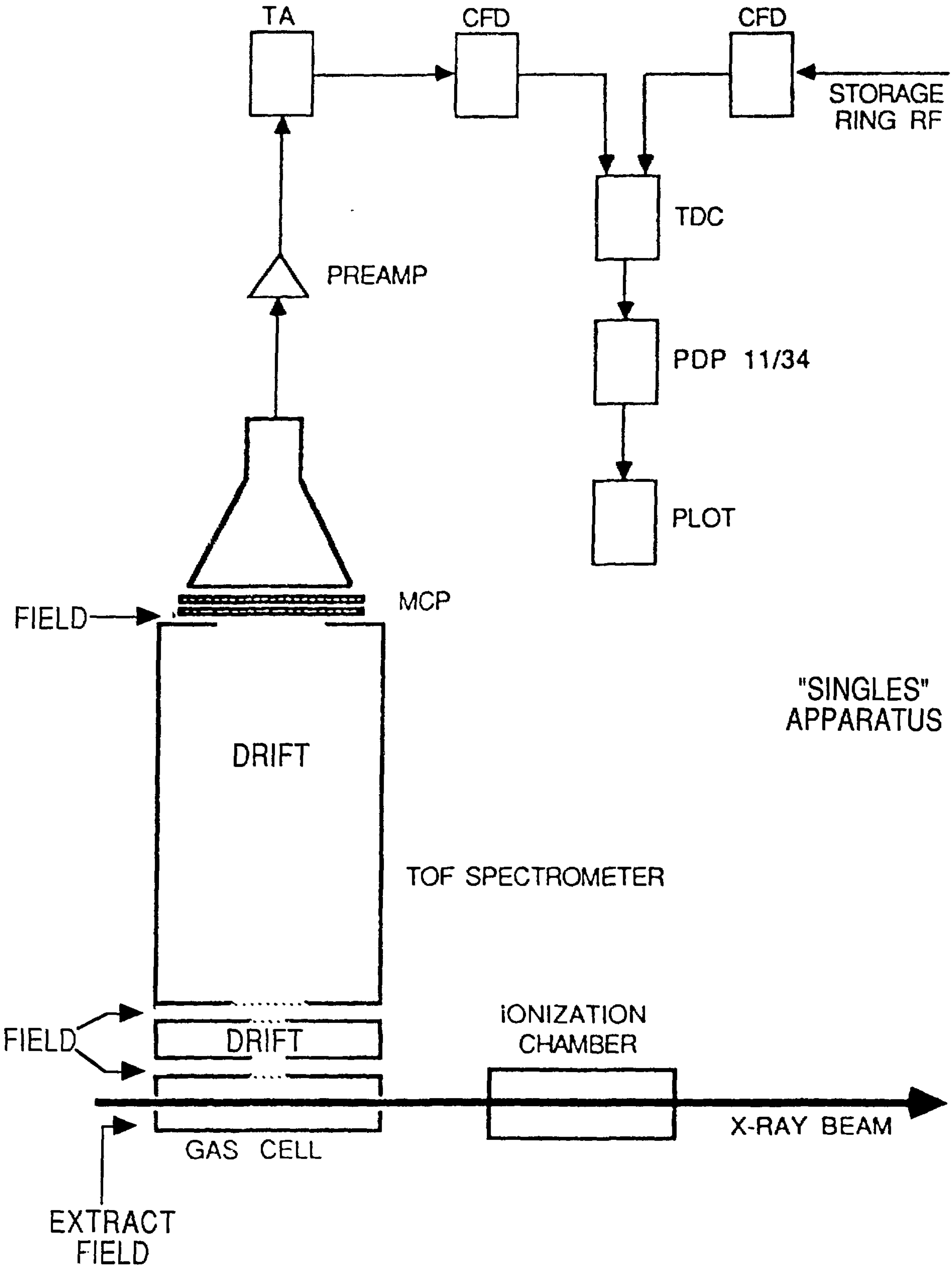
These q distributions average over all processes

==> Not sensitive to details related to individual transitions

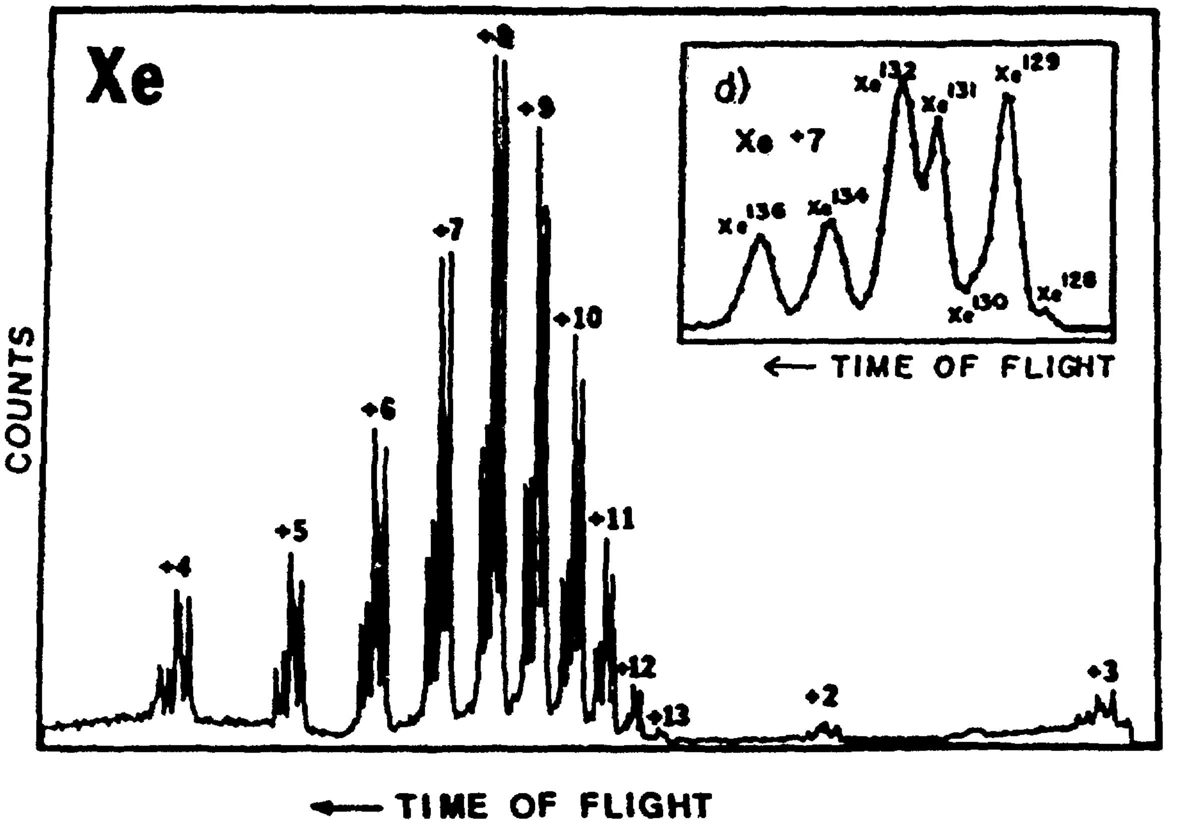
∴ Coincidence experiment with Auger electrons, using very highly monochromatized light

COLLABORATORS

C. Biedmann	UT / ORNL
C. S. O.	"
R. T. Short	"
N. Kellor	"
I. A. Sellin	"
L. Liajeby	MSI
D. Lindle	NIST

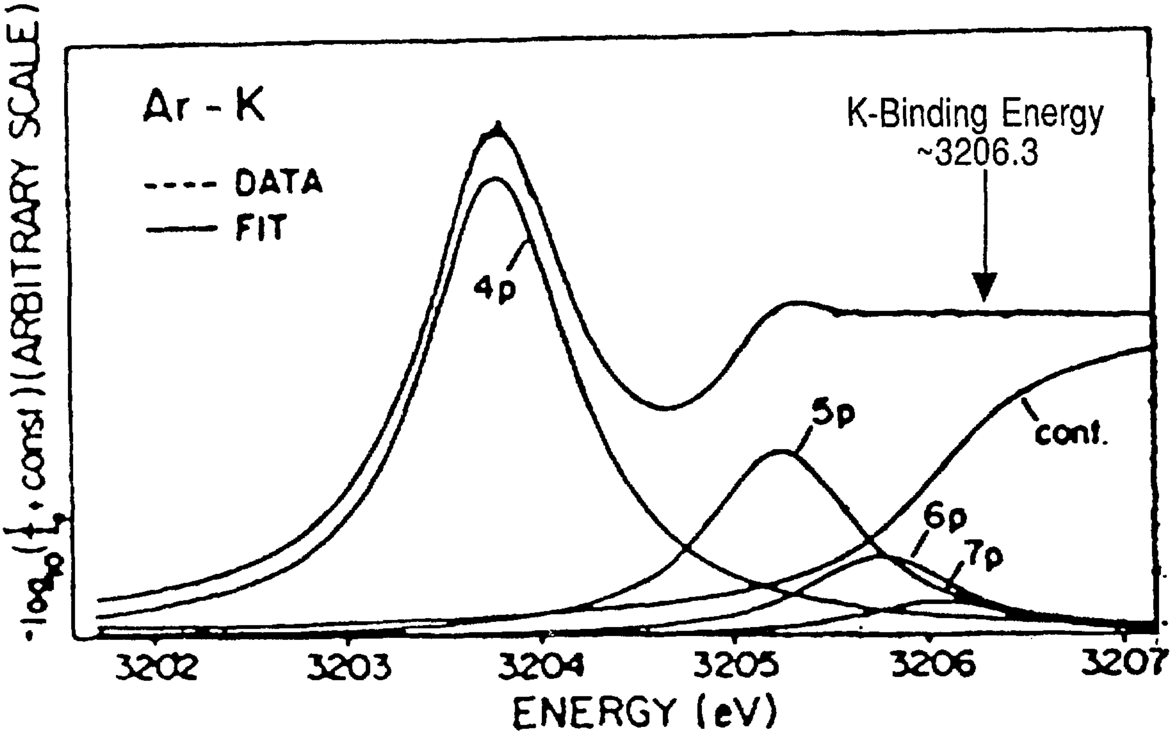

TA
CFD
CFD
STORAGE
RING RF
TDC
PREAMP
PDP 11/34
PLOT
MCP
FIELD
"SINGLES"
APPARATUS
DRIFT
TOF SPECTROMETER
FIELD
DRIFT
IONIZATION
CHAMBER
X-RAY BEAM
GAS CELL
EXTRACT
FIELD

Spectrometer can resolve isotopes of Xe

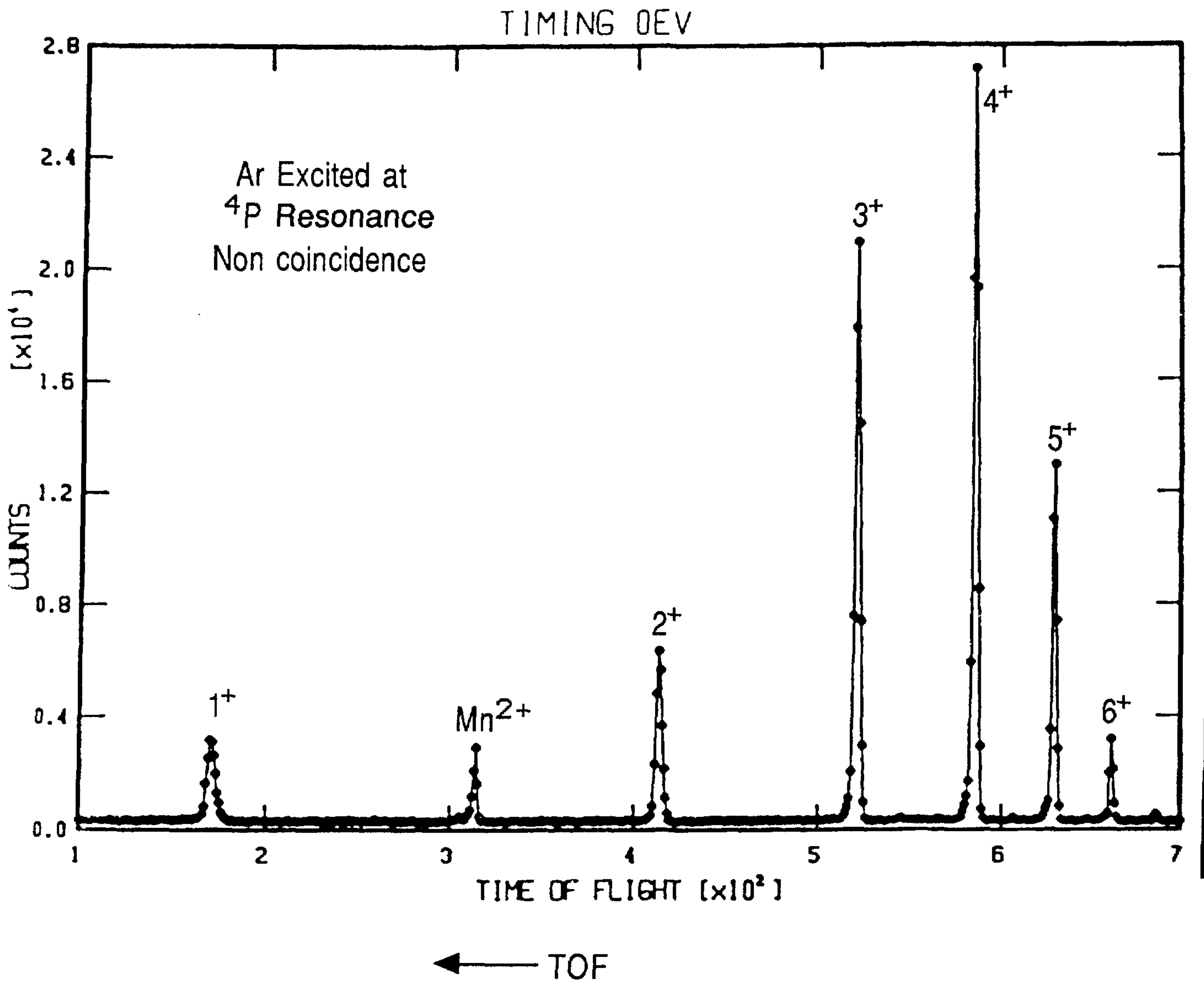


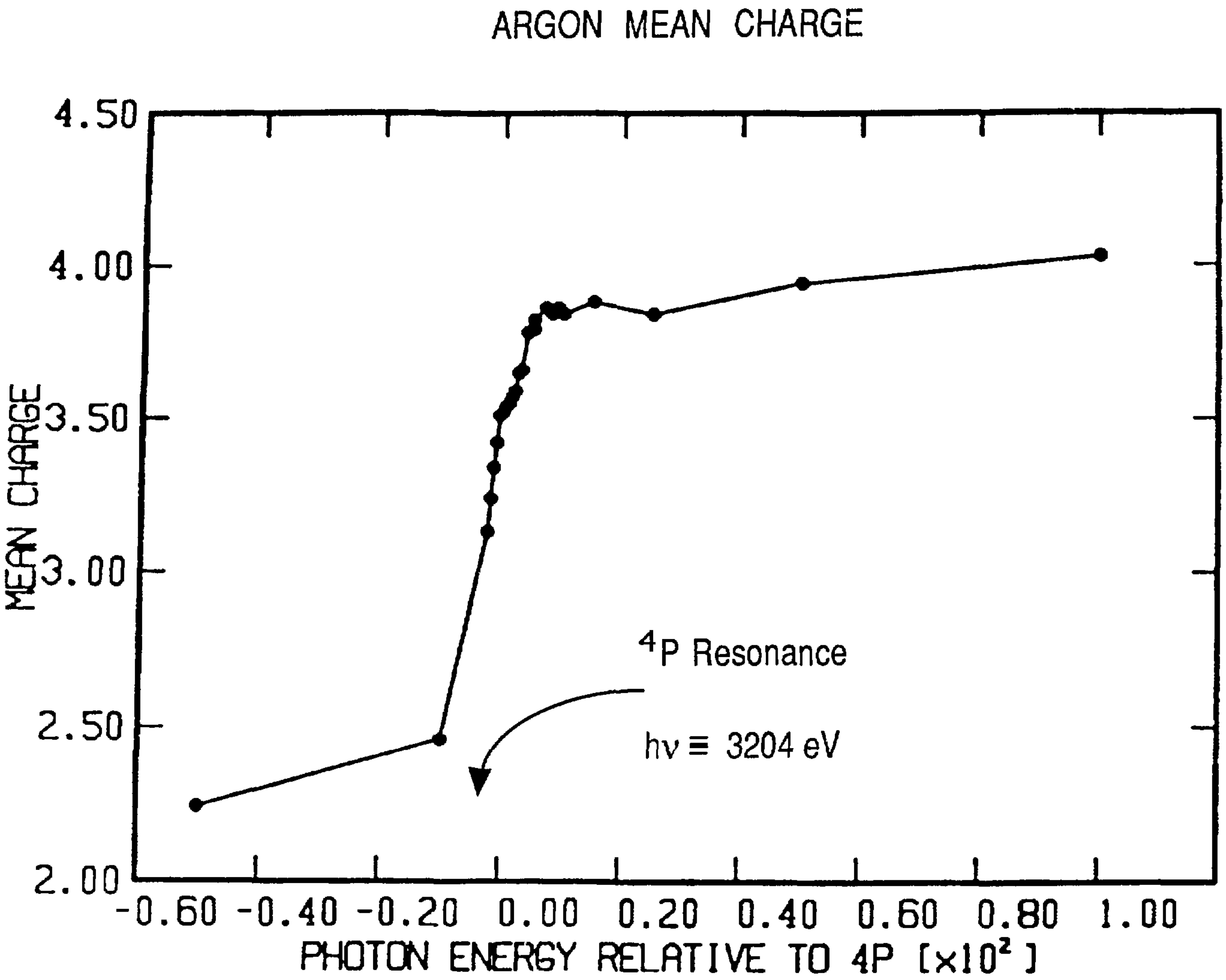
"White" x radiation above ~ 3 keV at SSRL

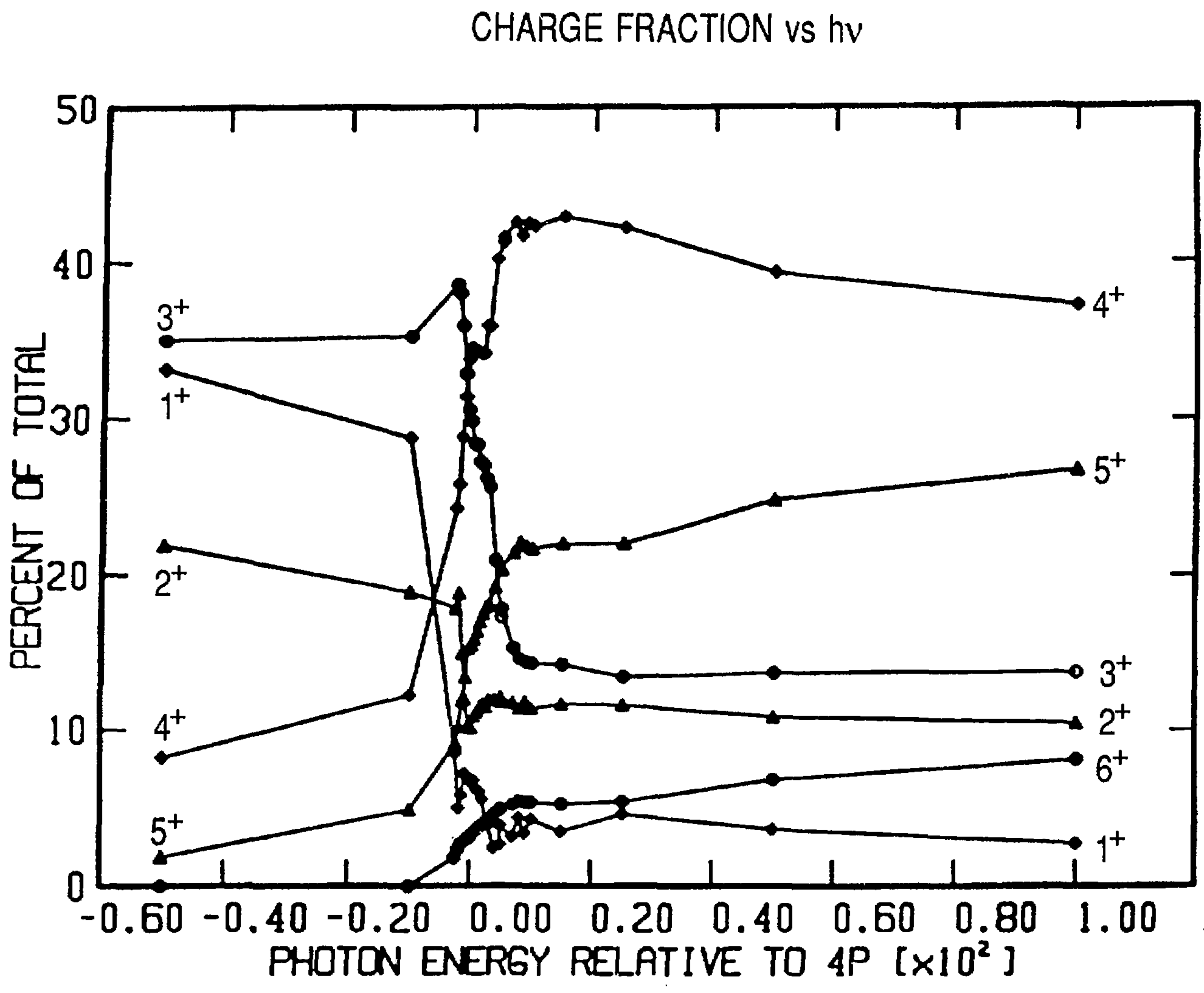
L_{123} vacancies created



Breinig, Chen, Ioe, Parente, Crasemann, Phys. Rev. A 22, 520 (1980)


TIMING 0EV
Ar Excited at
4P Resonance
Non coincidence
1+
Mn2+
2+
3+
4+
5+
6+
COUNTS [x10⁴]
2.8
2.4
2.0
1.6
1.2
0.8
0.4
0.0
1
2
3
4
5
6
7
TIME OF FLIGHT [x10²]
TOF


ARGON MEAN CHARGE
4.50
4.00
3.50
3.00
2.50
2.00
MEAN CHARGE
^{4}P Resonance
hν ≡ 3204 eV
-0.60 -0.40 -0.20 0.00 0.20 0.40 0.60 0.80 1.00
PHOTON ENERGY RELATIVE TO 4P [x10^2]



At the ^{4}p resonance, ~ 2.5 eV below Ar K ionization threshold, q distribution shifts abruptly to higher q.

HOW CAN AN Ar K VACANCY DECAY?

*$K - L_{23}L_{23}$

*$K - L_1L_{23}$

*$K - L_{23}M_{23}$

$K - L_1L_1$

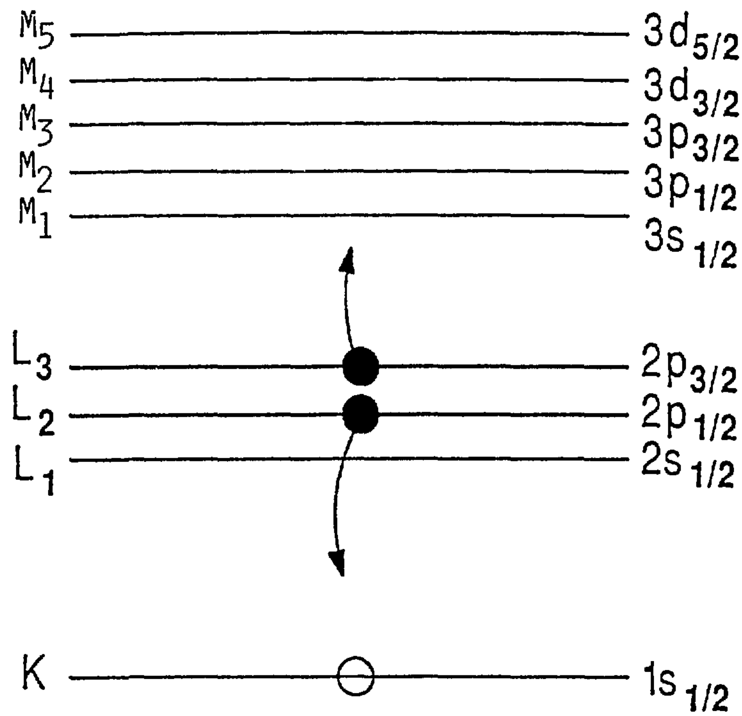
$K - L_1M$

$K - MM$

RADIATIVELY

*MOST IMPORTANT

NOTATION: Ex: $K\text{-}L_2L_3$

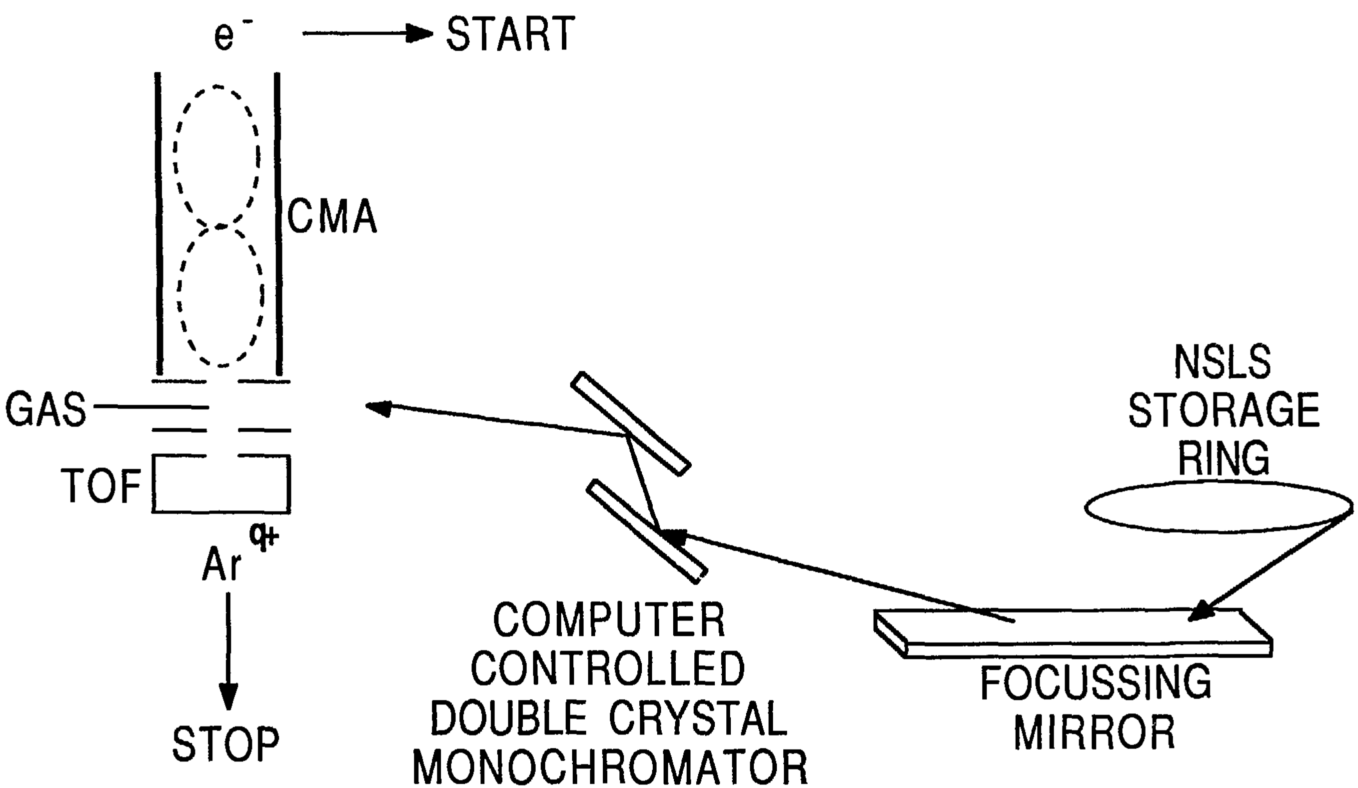


Advantage of requiring Auger coincidence:

1) All initial vacancies are guaranteed to be in K shell.

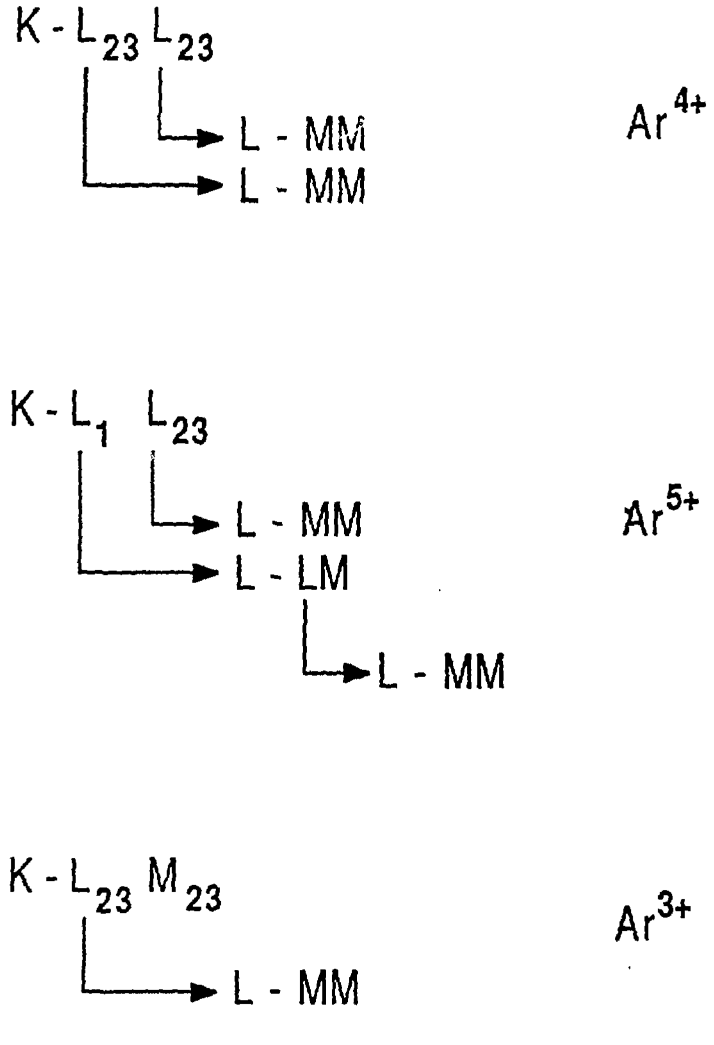
2) First decay is specified.

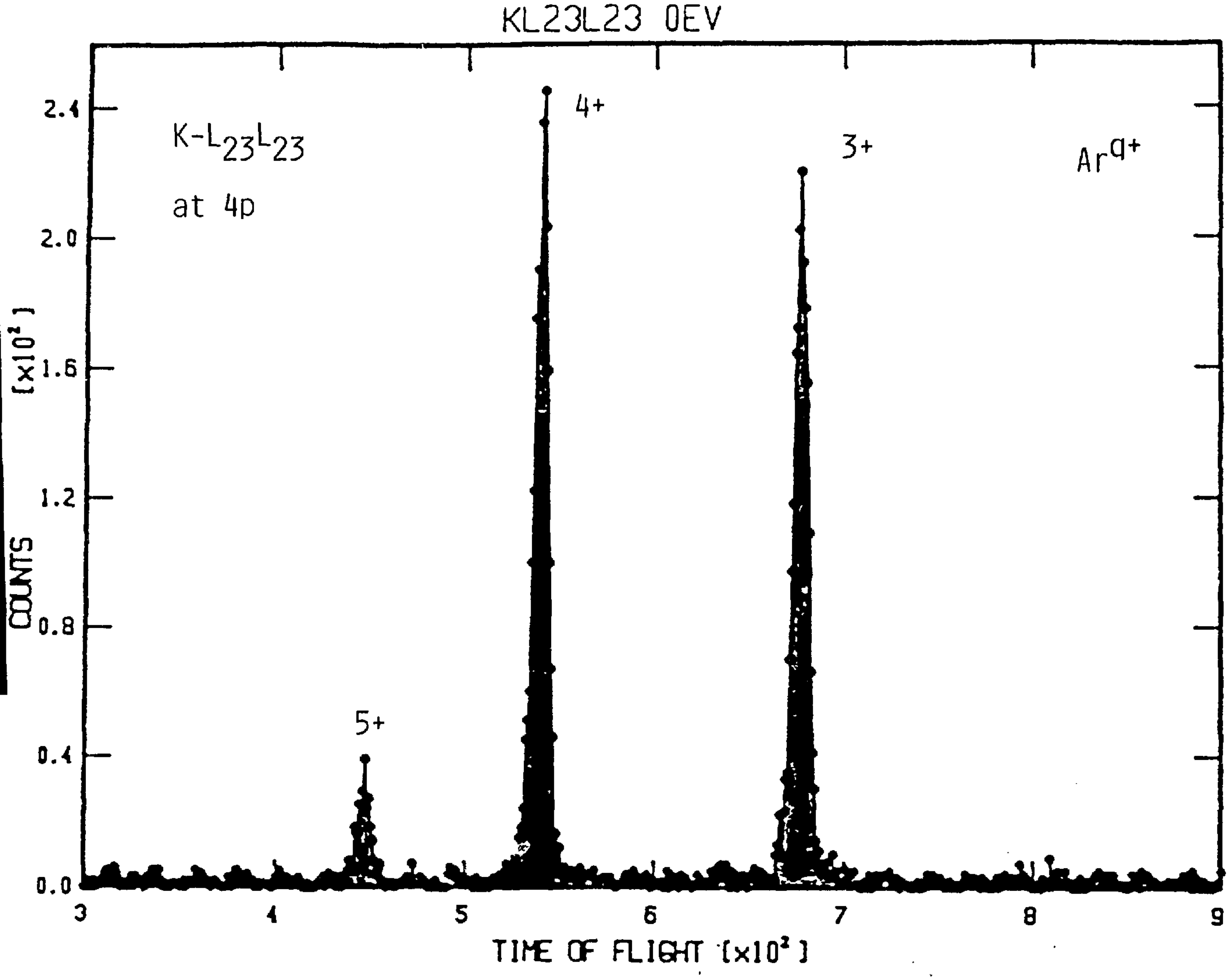
EXPERIMENT



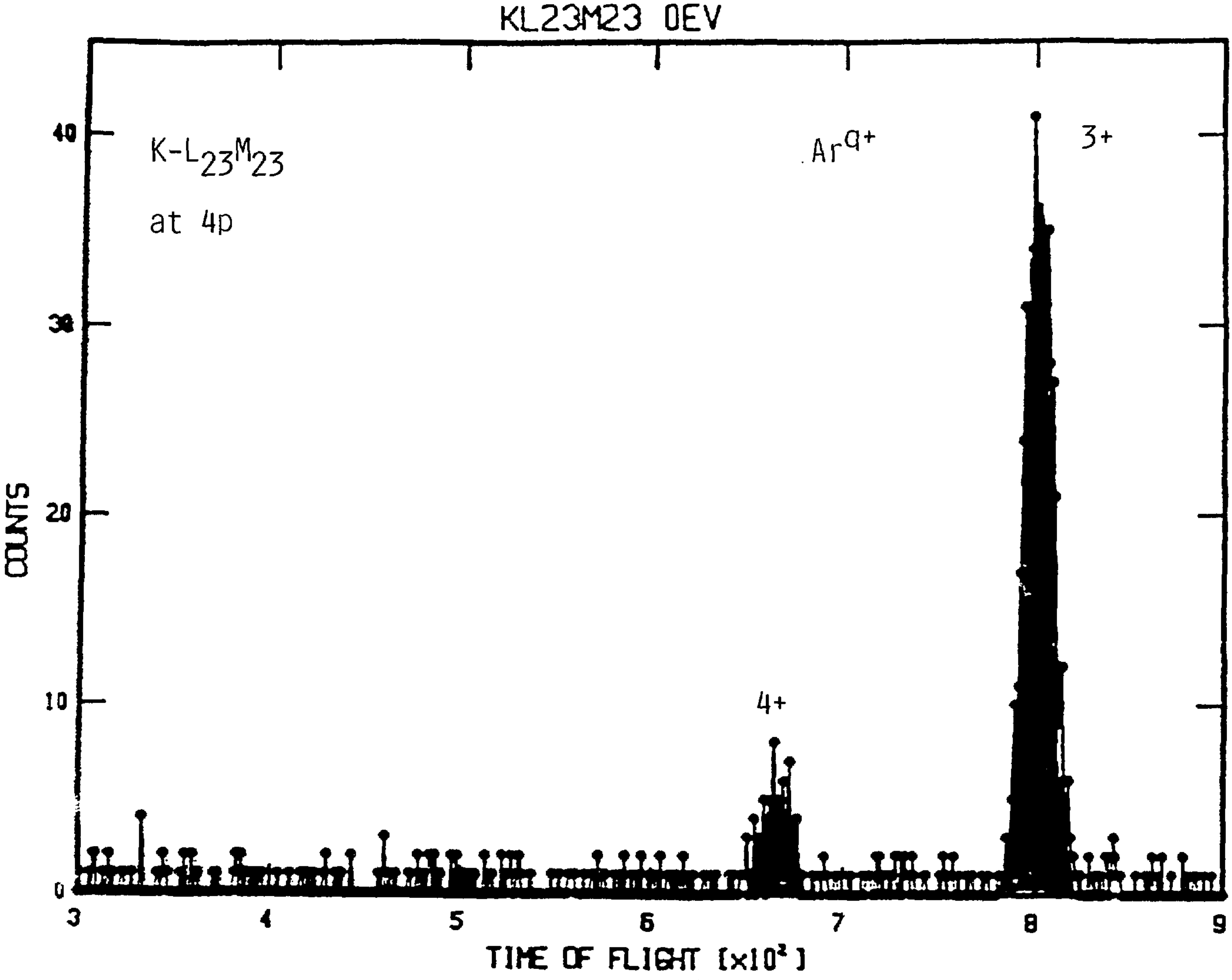
e^-
START
CMA
GAS
TOF
Ar^{q+}
STOP
COMPUTER
CONTROLLED
DOUBLE CRYSTAL
MONOCHROMATOR
FOCUSSING
MIRROR
NSLS
STORAGE
RING

WHAT CHARGE STATES ARE MOST LIKELY FOLLOWING EACH K-AUGER DECAY?

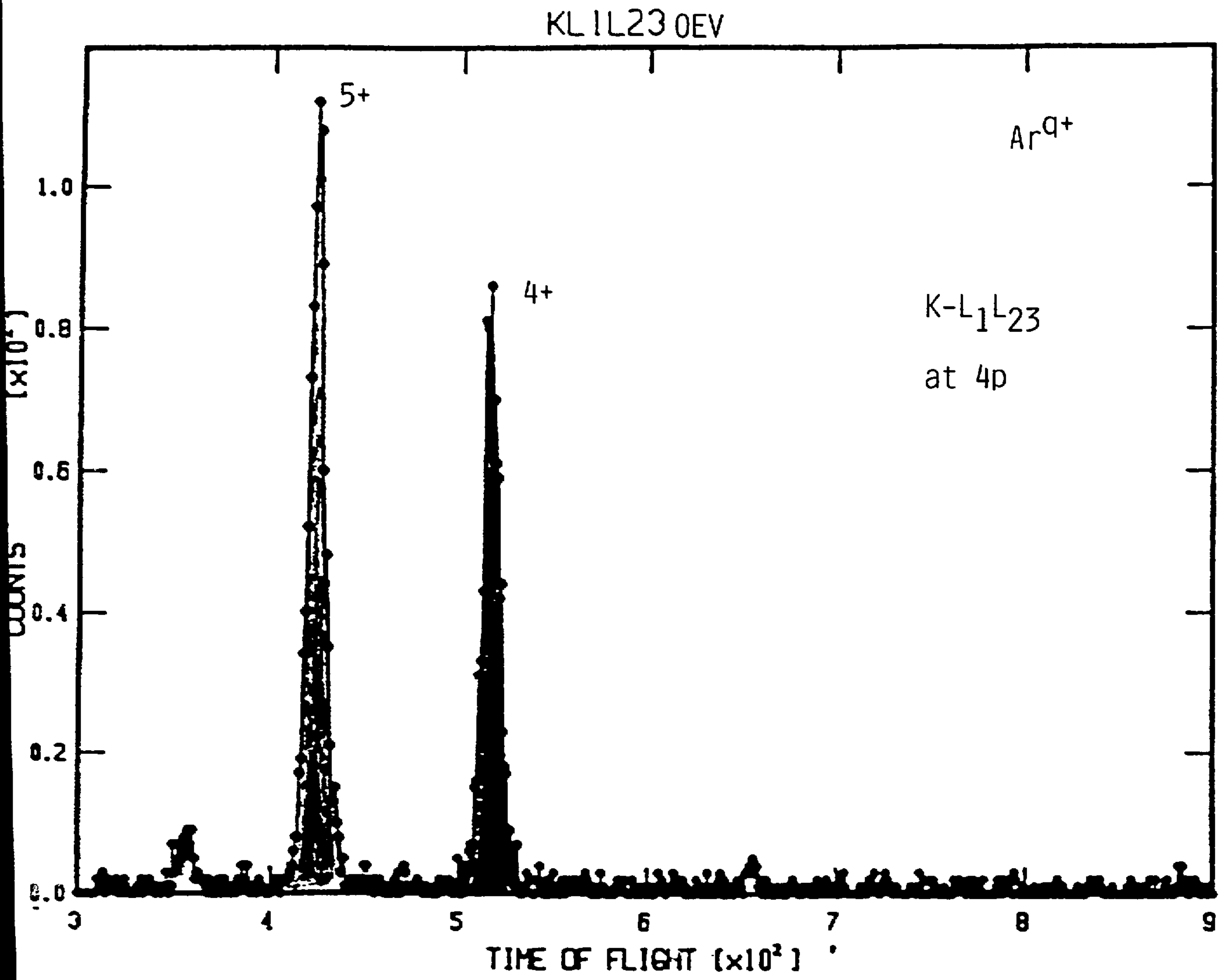




Ar^{q+} in coincidence with $K\text{-}L_{23}L_{23}$ at 2600 eV with $h\nu$ = 3203.8 eV (4p resonance)

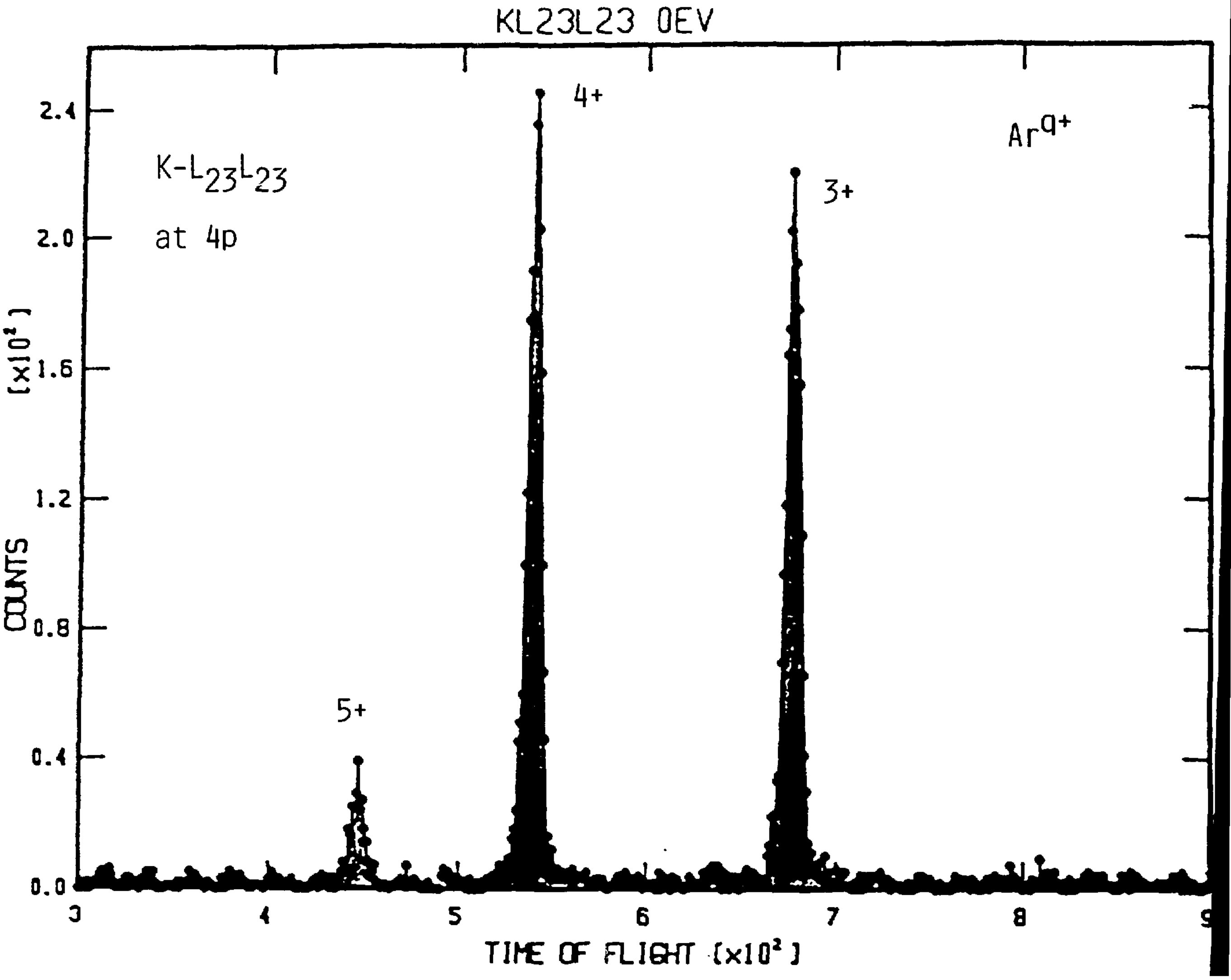


Ar^{q+} in coincidence with $K\text{-}L_{23}M_{23}$ at 2923 eV with $h\nu$ = 3203.8 eV (4p resonance)

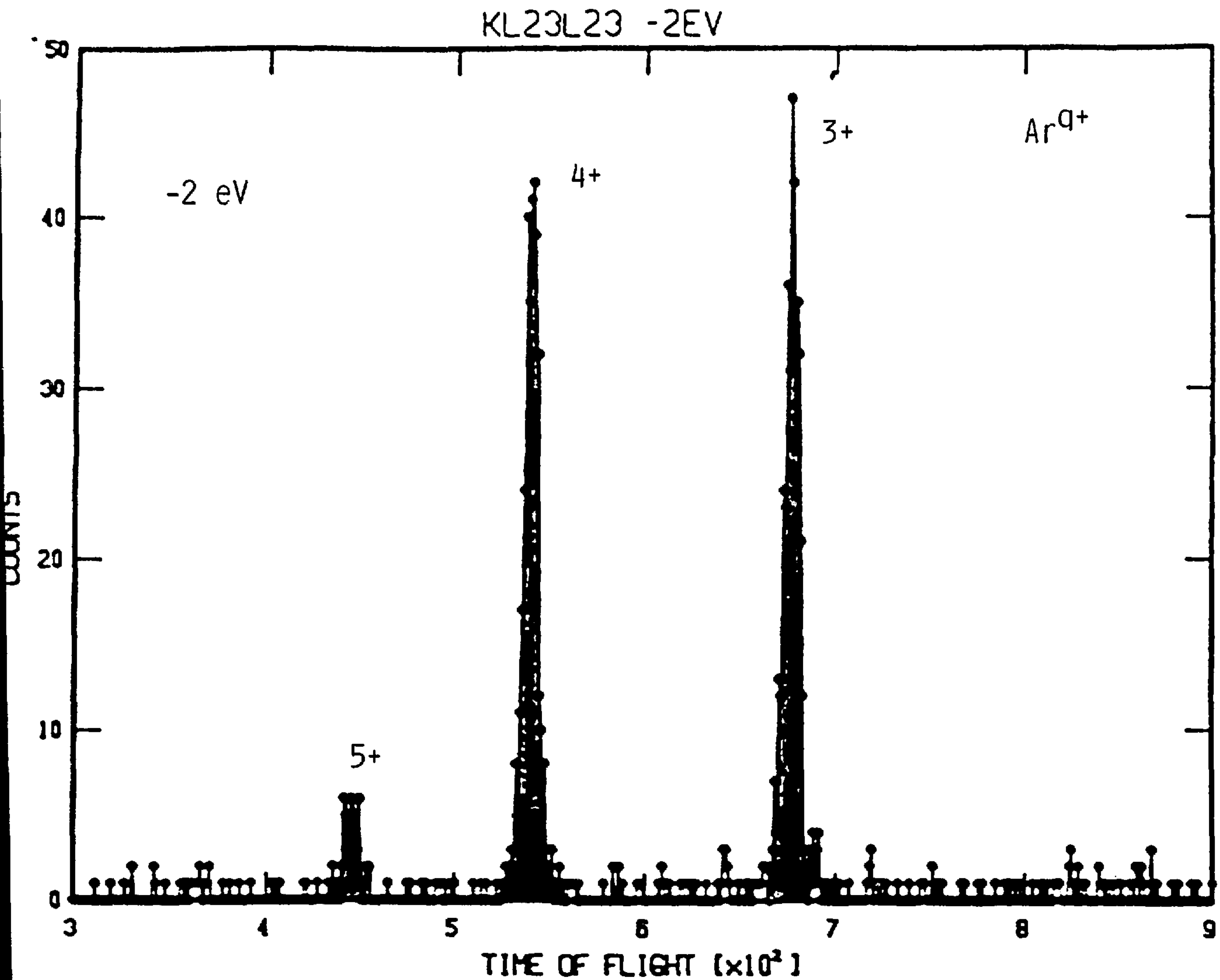


Ar^{q+} in coincidence with K-L_1L_{23} at 2575 eV with

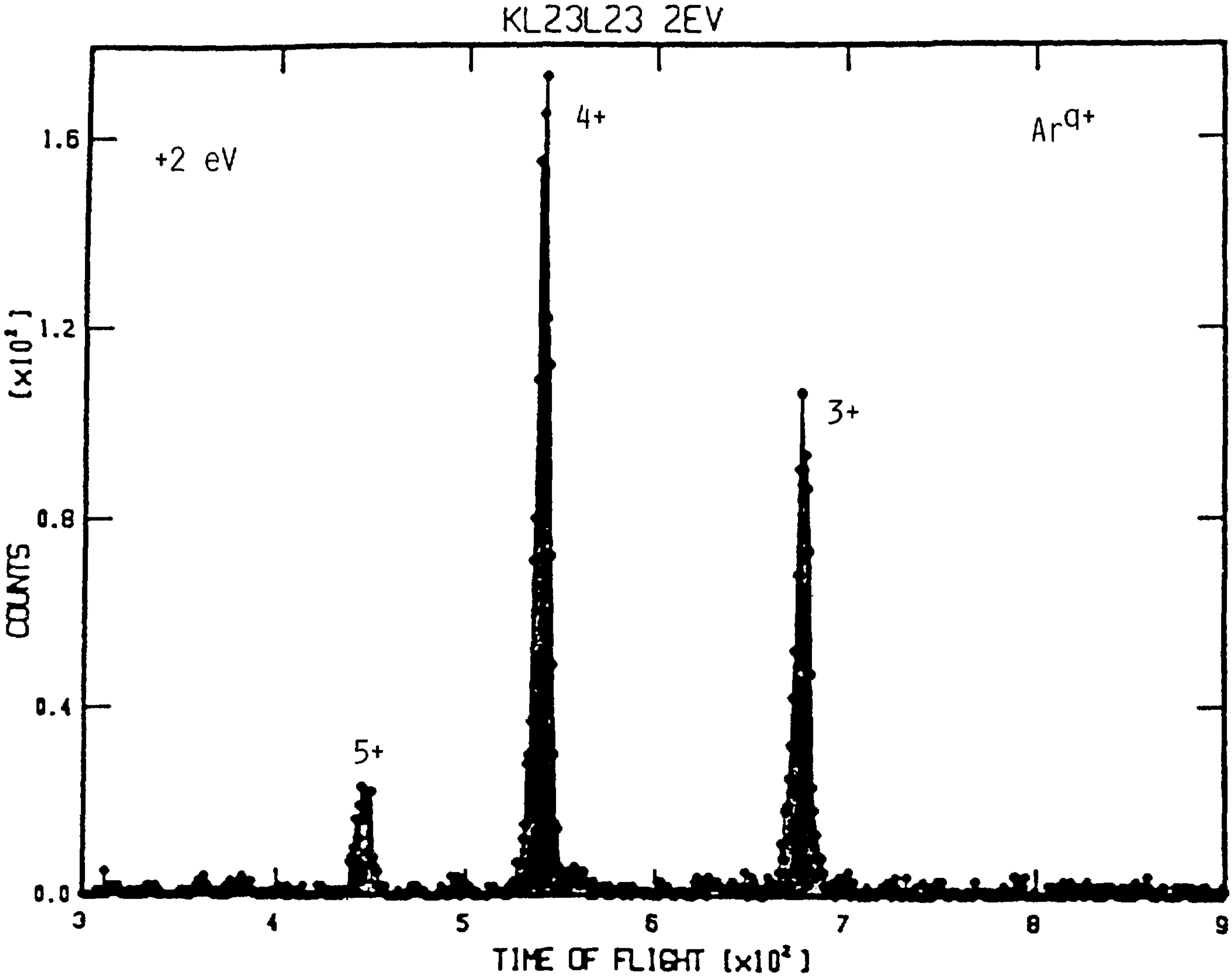
hν = 3203.8 eV (4p resonance)



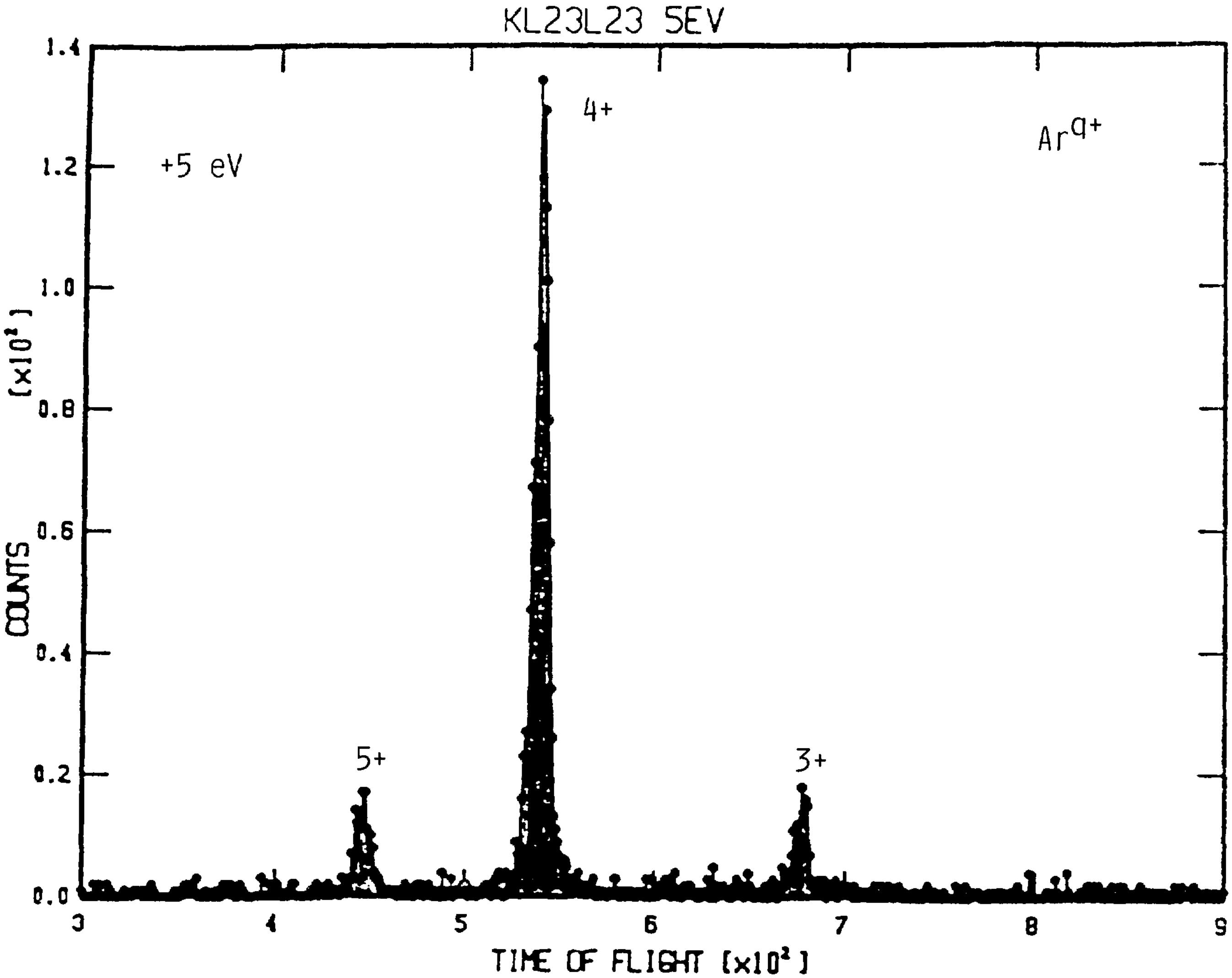
For fixed auger decay, K-$L_{23}L_{23}$, now vary photon energy.



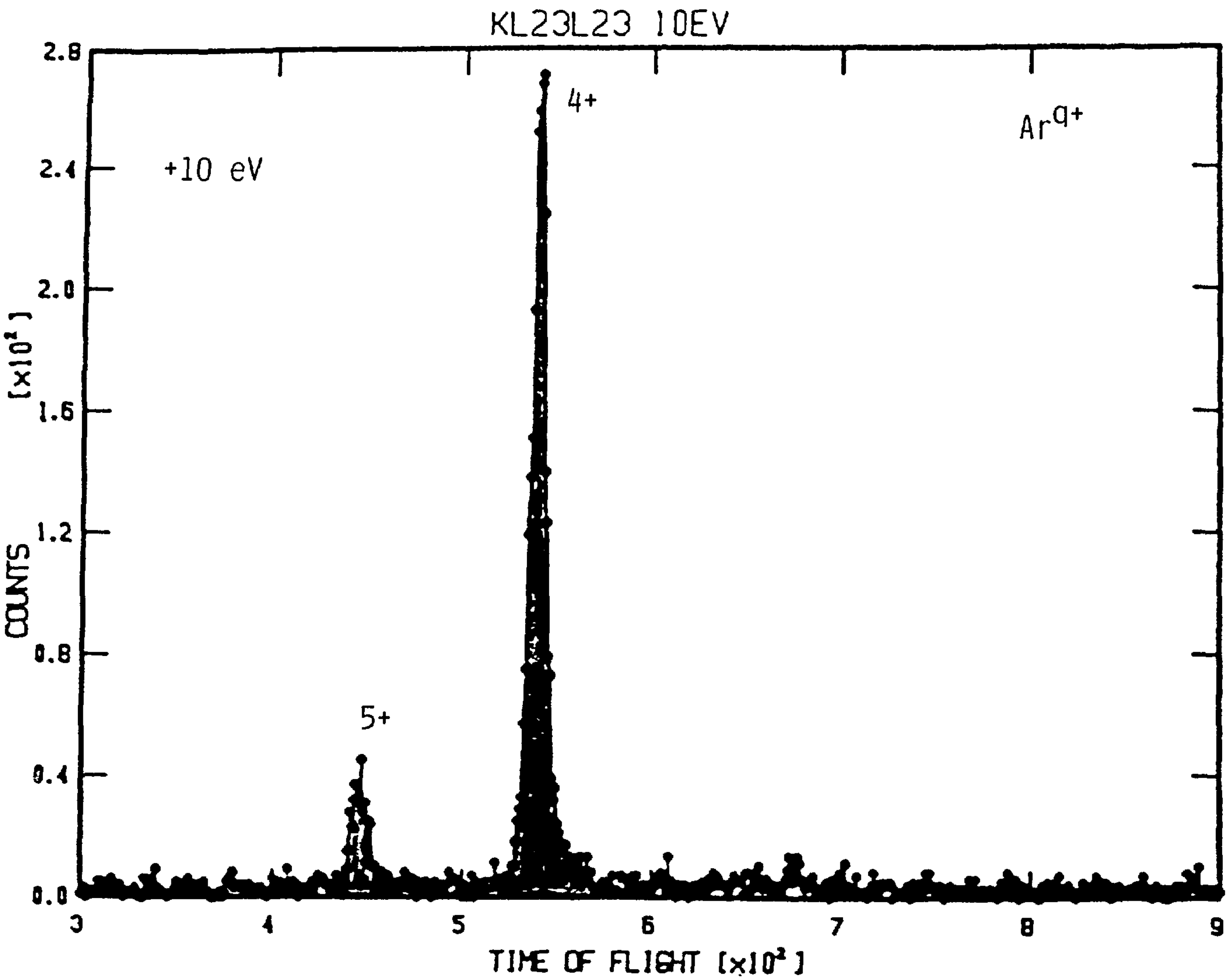
K-$L_{23}L_{23}$ coincidence -2 eV relative to 4p



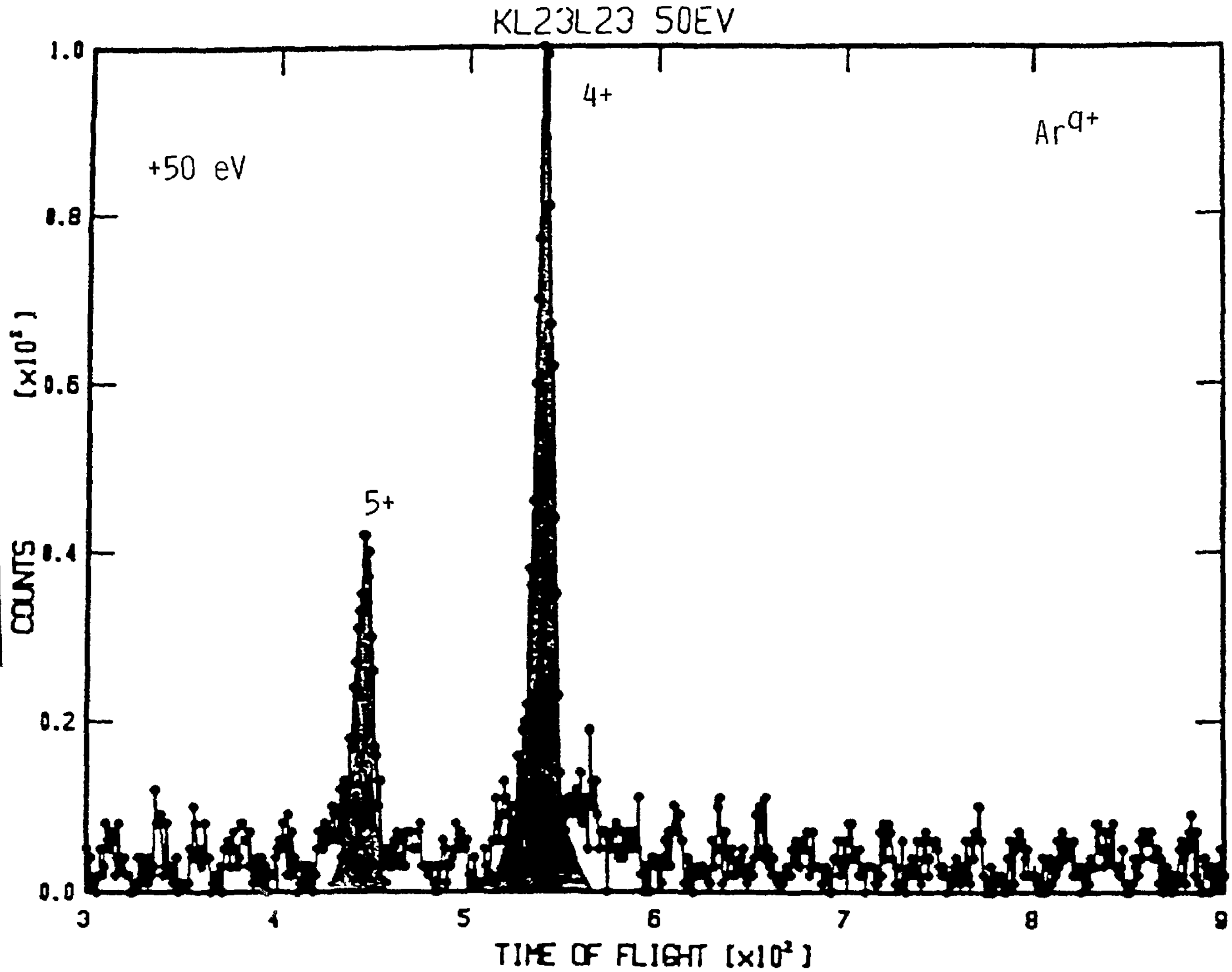
K-$L_{23}L_{23}$ coincidence +2 eV relative to 4p



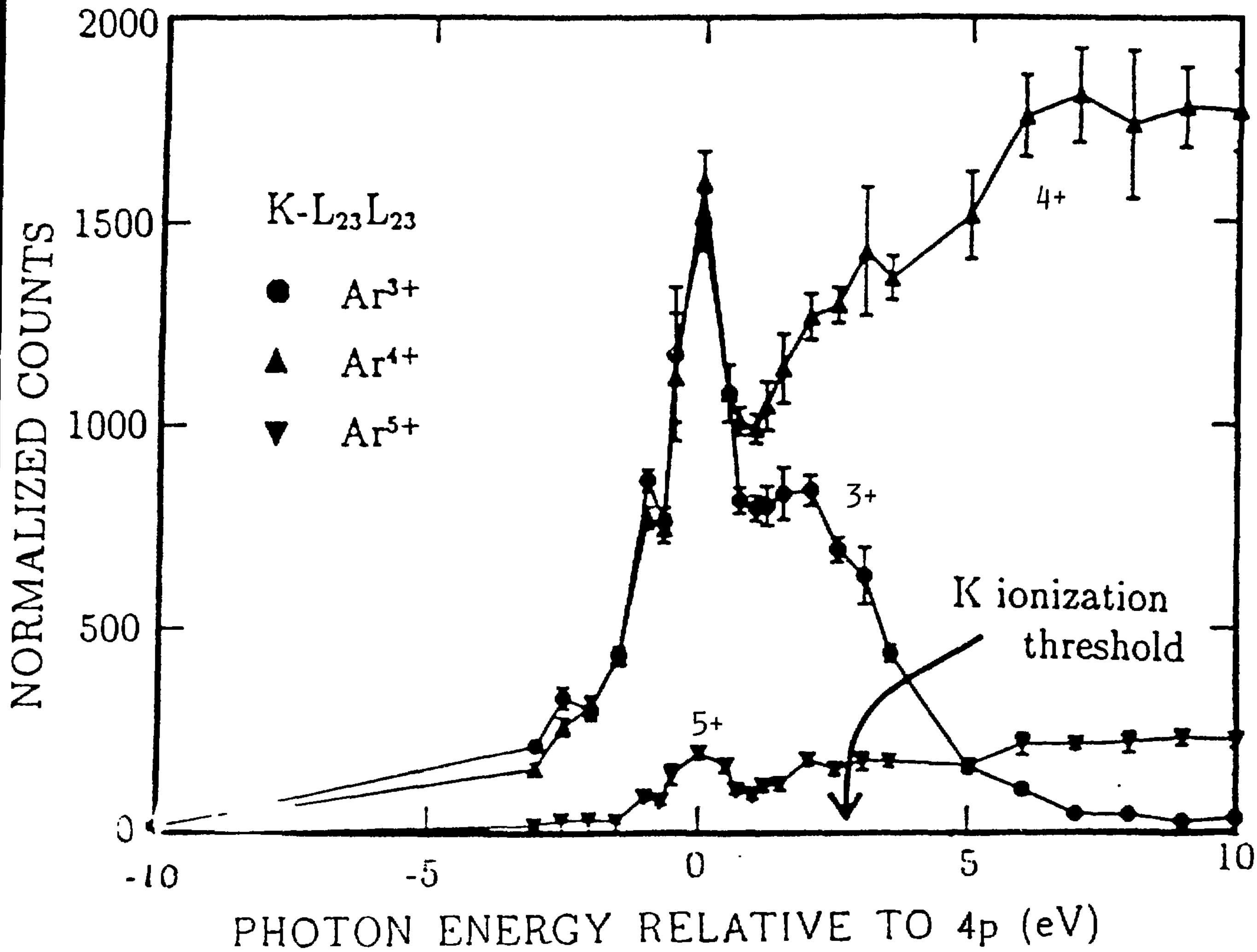
K-$L_{23}L_{23}$ coincidence +5 eV relative to 4p



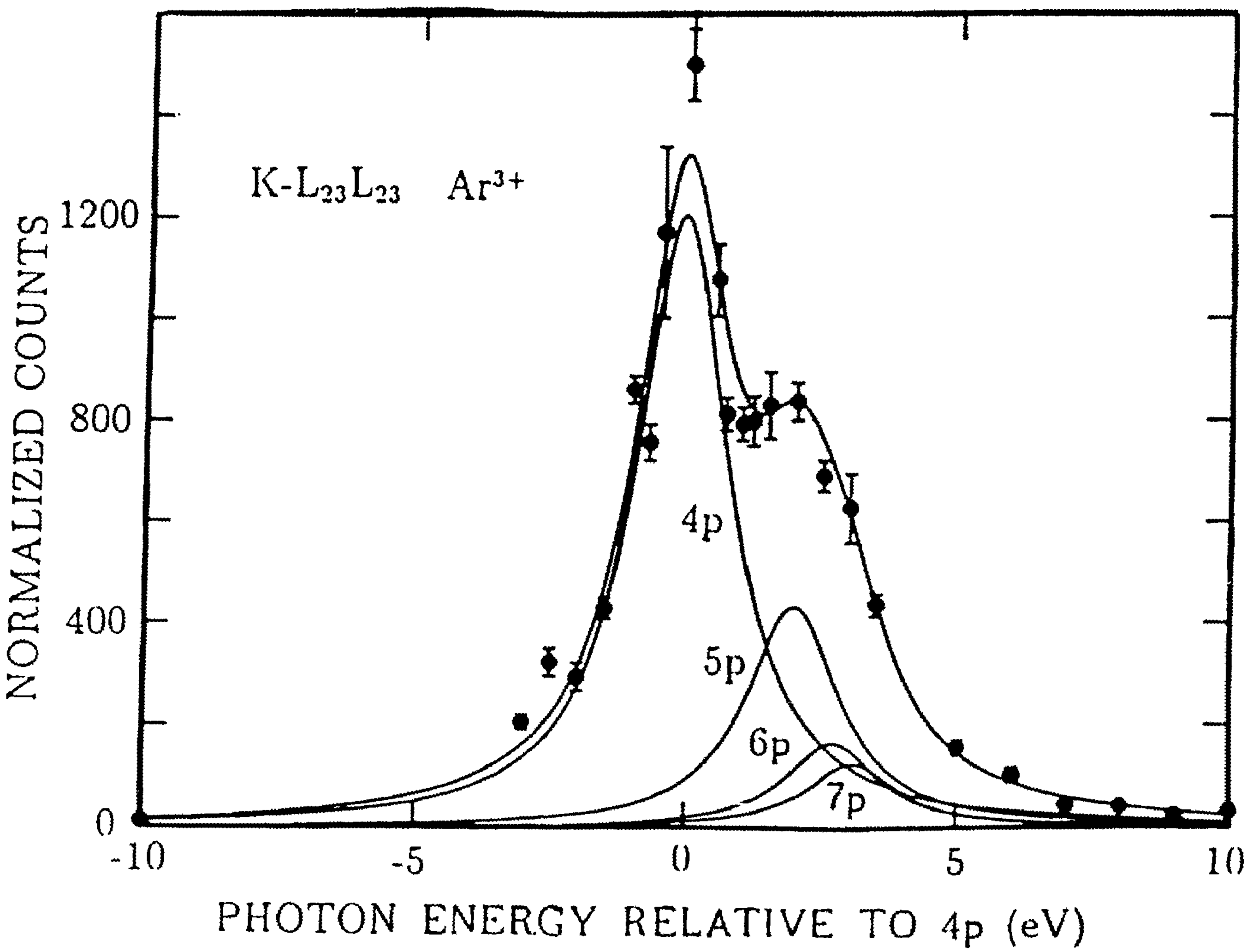
$K\text{-}L_{23}L_{23}$ coincidence +10 eV relative to 4p



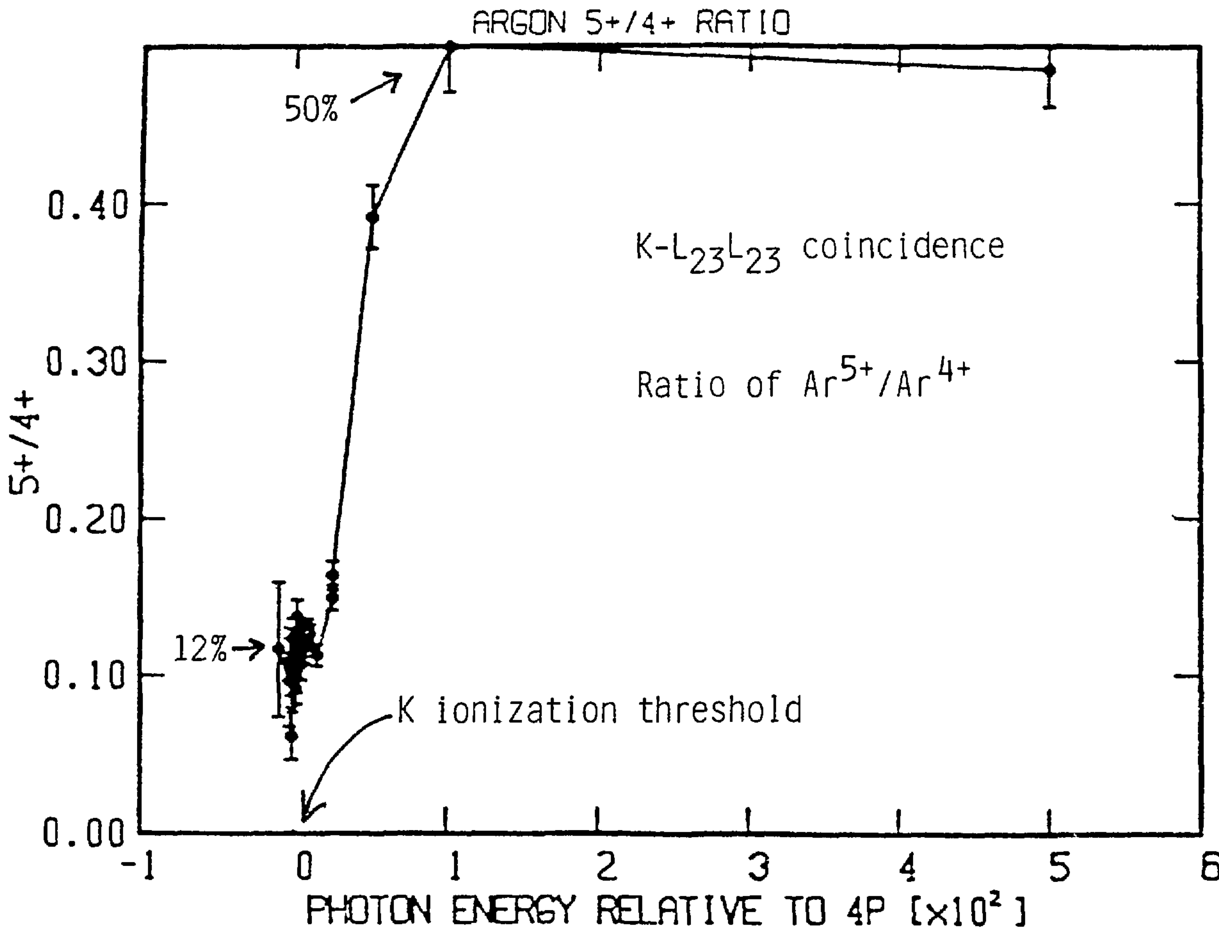
K-$L_{23}L_{23}$ coincidence +50 eV relative to 4p



$Ar^{3+,4+,5+}$ coincident with $K\text{-}L_{23}L_{23}$ auger decay normalized to constant photon flux.



Decomposition of Ar^{3+} coincident with $K\text{-}L_{23}L_{23}$ auger decay into components due to excitation of K electron into bound np levels.



Ar^{5+}/Ar^{4+} ratio depends strongly on $h\nu$

WITH HARDER X-RAYS (APS?)

1) Do coincidence with less probable Auger lines (e.g., K - L_1 L_1).

2) Operate Auger-electron channel at high resolution.

3) Triple coincidence with photon, Auger electron, and photoion.

4) Do spectroscopy of secondary, tertiary, Auger electrons to study relaxation.