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K. W. Jones

Brookhaven National Laboratory, Upton, New York 11973 USA

Invited talk Presented at

XXII International School of Physics

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Experiments in Atomic and Applied Physics Using Synchrotron Radiation*

K. W. Jones
Brookhaven National Laboratory, Upton, New York 11973 USA

ABSTRACT

A diverse program in atomic and applied physics using x rays produced at the X-26 beam line at the Brookhaven National Synchrotron Light Source is in progress. The atomic physics program studies the properties of multiply-ionized atoms using the x rays for photo-excitation and ionization of neutral atoms and ion beams. The applied physics program builds on the techniques and results of the atomic physics work to develop new analytical techniques for elemental and chemical characterization of materials. The results are then used for a general experimental program in biomedical sciences, geo- and cosmochemistry, and materials sciences. The present status of the program is illustrated by describing selected experiments. Prospects for development of new experimental capabilities are discussed in terms of a heavy ion storage ring for atomic physics experiments and the feasibility of photoelectron microscopy for high spatial resolution analytical work.

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Introduction

The National Synchrotron Light Source (NSLS) at Brookhaven National Laboratory (BNL) is a high intensity source of x rays that gives new opportunities for research in atomic and applied physics. This paper describes work in both areas that has been done with x ray beams at the NSLS X-26 beam line that show how new types of experiments are made possible by the use of synchrotron radiation. In atomic physics it is shown that measurements of fluorescent radiation on dilute gas targets can now be done so that it is possible to study the light emitted by atoms in an isolated environment. In applied physics work it is shown that it is now possible to detect the presence of as few as 10^7 atoms in a volume of 10^{-9} cm^3 by the use of synchrotron radiation induced x-ray emission (SRIXE) methods. An example of the application of the SRIXE method is given.

The equipment that is now used can be improved in many different ways in the future. In atomic physics it may be possible to add a cooled heavy ion storage ring (CHISR) in close proximity to the NSLS so that it will be possible to study photoexcitation and ionization of multiply-charged ions with high precision. For applied physics it may now be timely to consider the use of photoelectron microscopy as a complement to the detection of fluorescent x rays. Consideration of the use of such an approach to sub-micron resolution imaging is given.

The combination of descriptions of present experimental results with plans for future developments is intended to show the scope and vitality of the research based on the use of NSLS x ray beams in a succinct manner. A full description of the topic goes beyond the scope of this paper.

(Some references to papers and abstracts on crucial aspects of the work are given in References 1-18).

The entire program that is described here represents the work of scientists from the BNL Department of Applied Science Division of Atomic and Applied Physics: B. M. Johnson, M. Meron, B. M. Gordon, A. L. Hanson, and J. G. Pounds, and from The University of Chicago Department of the Geophysical Sciences: J. V. Smith, M. L. Rivers, and S. R. Sutton. We have also had many collaborators from many different institutions who have left a strong imprint on the program.

It is appropriate to mention specifically a long-term interaction between the Institute of Nuclear Physics at Cracow, the Department of Applied Science Division of Atomic and Applied Physics at Brookhaven, and the Kosciuszko Foundation of New York in this contribution to the Proceedings of the XXI Zakopane Winter School. This interaction has made it possible for visitors from Cracow to spend extended periods of time at Brookhaven. The experiments described here have benefited in many ways from invaluable contributions by A. Z. Hrynkiewicz, J. Kajfosz, M. Cholewa, W. M. Kwiatek, and B. Rajchel during their visits. We hope to see our joint ventures continue and to be able to report further results at subsequent meetings in Zakopane!

Characteristics of the NSLS X-26 Beam Line

The NSLS is an electron storage ring that ran at 2.5 GeV with maximum stored electron currents of ~ 200 mA and average beam lifetimes of around ~ 8 hours. The photon flux produced at the experimental station 20 m from the electron orbit is given in Figure 1. The low-energy cutoff is produced by windows and filters used in the path of the beam.

The actual mechanical layout of the beam line is shown in Figure 2. Beryllium windows are used to separate the beam line vacuum system from the machine vacuum for greater ease in doing experiments. The apparatus in the experimental enclosure (hutch) was arranged so that atomic and applied physics could be done simultaneously. A photograph of the hutch interior is shown to the left in Figure 3. The magnet system at the left sideof the hutch is a Penning-type ion trap for atomic physics work and on the right side is the x-ray fluorescence microprobe system used for trace element measurements.

Measurement of Fluorescent Radiation from Argon Following K-Shell Ionization

Measurements of fluorescent radiation from dilute atomic targets are useful for obtaining atomic data that can be used to provide benchmarks data for improving theoretical atomic structure models and for determining cross sections or rate coefficients for very low energy ion-atom collision processes. With synchrotron radiation from the NSLS the photon flux at the sample position is high enough to make such measurements with target densities corresponding to target pressures in the region of 10^{-10} torr or approximately 3×10^{6} atoms/cm^{3}. Parenthetically, note that the determination of fluorescent radiation is the basis of the SRIXE method, with detection limits now running around 10^{6-7} total atoms. Values quoted for target densities depend on the assumptions made for detection system efficiencies, data accumulation times, and backgrounds. The above values are, therefore, to be considered as approximate.

The first experiment that was done at the X-26 beam line on fluorescent radiation for atomic physics purposes was the study of the radiation produced following the K-shell photoionization of argon (13). This could be done with filtered white radiation since the contribution from the L-shell ionization cross section is less than 10% of the K-shell cross section at the energies considered. The production rate of K vacancies in argon can be easily calculated by calculating the integral of the product of photon flux and cross section over the photon energy distribution. The value obtained is roughly 6×10^3 K vacancies/s for a target density of 10^7 atoms/cm^3, which is adequate for experimental work even though low.

The fluorescent photons produced in the wave length region from 50 to 150 nm were analyzed with a 0.2-m normal incidence spectrometer with a 600 g/mm toroidal grating. This instrument was operated with a resolution of 0.2 nm. A typical spectrum that was obtained for a pressure of 50 mtorr is shown at the bottom of Figure 4. The upper spectra indicate the predicted appearance of lines from individual ionization stages (Ar I to Ar VIII).

Analysis of the data showed that the intensities of the spectral lines were strongly affected by collisions with neutral argon ions. Measurements of the relative intensities of the spectral lines as a function of pressure thus give values for the rate coefficients for charge neutralization in ion-atom collisions at energies around 100 meV. The results of such experiments are now being analyzed.

The mechanisms by which the K vacancy is filled after production results in the further ionization of the atom. For argon a distribution of charge states is produced with values as high as 6 or 7 and a maximum at 4. It is therefore possible to examine a wide range of ions in the course of a single experiment. Figures 5 and 6 show the results of two measurements (16) of the charge-state distribution using a Penning ion trap and a recoil-time-of-flight apparatus on the X-26 beam line.

The results of the first experiments show the feasibility of atomic photoionization studies. Improvements are under way. The photon flux delivered to the target will be increased by several orders of magnitude by the addition of focussing mirrors in the beam line. This will increase the signal/background and make possible measurements at lower target densities. The use of time-resolved spectroscopy to take advantage of the natural bunching of the synchrotron beam will improve the accuracy of the rate coefficient measurements.

X-Ray Microsocpy (SRIXE) Measurements

The use of x-ray fluorescence for analytical measurements has been an established method for many years. The synchrotron light source has catapulted it into a new era because of the high polarization and high brilliance (photons/source area/solid angle/energy interval/s), brightness (photons/solid angle/energy interval/s) and flux (photons/mrad horizontal/energy interval/s) of the x rays emitted. The introductory work described here relies only on the high flux. Further developments of the beam line using mirrors will utilize the high brightness and brilliance of the source.

The SRIXE system that we have used is simple, albeit very powerful. The measurements are carried out after the x-ray beam passes through a window and into the laboratory. Beam intensity is monitored with an ion chamber. Fluorescent x rays are detected at 90 degrees to the incident beam, a position which minimizes the background of Compton-scattered photons. Our measurements used a 30-mm^2 Si(Li) detector with a resolution of around 155 eV for 5.9-keV Mn K-x rays. Sample positions were changed using stepping-motor-driven x-y-θ translators. Beam positions could be set to about 1 micrometer. The beam size was defined by tantalum slits with a spacing that could be adjusted using stepper motor driven micrometers. Minimum beam size was 10 micrometers. A computer was used to control the system so that maps could be easily made of the spatial distribution of specific elements. Thus, what we have is a very versatile system for high energy x-ray microscopy measurements. A photograph of the arrangement is shown in Figure 3. The present system is a refined version of one that was originally used in experiments at the CHESS facility at Cornell University (1).

The detection limits for the system defined by the Currie criterion (ref) were determined by measuring the spectra of various standard reference materials. The results obtained for a home brew of various elements at the 10 parts per million (wet weight) level dispersed in a 20-micrometer slice of gelatin is shown in Figure 3. The conditions for the irradiation are shown in the figure. The minimum detection limits extracted from this data are shown in Figure 7.

The x-ray microscope (XRM) has been used for many investigations in the biomedical and earth sciences. A specific example to illustrate how it can be used is the work on trace element distributions in the rat brain by Kwiatek et al. (19) In this investigation measurements were made of trace elements in the brain for control animals and for animals that had ingested varying amounts of lead. It is known that lead is a toxic element and that it changes the function of the brain. Changes and interactions in the trace element distributions can thus be expected to occur. It is impossible to make measurements at very low concentration levels with good resolution by other techniques than SRIXE or proton-induced x-ray emission (PIXE) so that the XRM should be a truly quantitative improvement in the tools available to the biologist.

The concentration of iron in the rat cerebellar cortex can be taken as a specific example from the above work on the rat brain. Figure 8 shows a conventional photomicrograph of a coronal section of the cerebellar cortex. Structures in the cortex called the molecular and granule cell layers and the fiber tract are indicated. Figure 9 is a photograph taken at higher resolution that shows more details of the three regions. Figure 10 shows a portion of a 30 × 40 pixel map of the iron distribution found in the same region shown in Figure 9. Each pixel was 100 micrometers on a side. White represents highest iron concentrations and black represents the lowest. The three pronged region in white, (iron concentration is high), corresponds to the fiber tract region. This experiment will be part of the Ph.D. thesis to be submitted to the Institute of Nuclear Physics in Cracow by W. M. Kwiatek.

Future Direction in Atomic Physics

Recently there has been much interest in building storage rings for heavy ions. The advantage of the storage ring is that the effective ion current is enhanced because of the high characteristic revolution frequency of the ions in the ring. It seems to be feasible to fill a ring with around 10^{10} ions and to get effective currents close to 10^{16}/s. The interaction rates that are produced will then be many orders of magnitude greater than if experiments were done with ordinary types of ion sources. Rings to date have been proposed for study of electron- ion or ion-ion (or atom) collisions.

We have proposed (11,14) building a storage ring at the NSLS so that it would be possible to do precision photon excitation and ionization experiments using synchrotron radiation. Experiments using electron and ion beams as probes would also be done.

The science that could be done at this facility would cover many crucial areas of atomic physics. A first discussion of the topic has been published (14). Perhaps the most important point to make at this time is to establish the luminosities that are achieved in various types of beam experiments. A tabulation is given in Table I taken from Ref. 14. This shows that the storage ring will substantially extend the range of feasible experiments that can be done in investigating the properties of multiply-charged ions.

The ring requires an elaborate ion source for best operation. At BNL we are fortunate to have a powerful three- stage Tandem facility to use as an injector. These machines produce heavy ions with energies in the region of several hundred MeV and with charge states from fully stripped around Z = 22 to about 50% stripped at lead or gold.

The geographical layout of the proposed ring at BNL is shown in Figure 11 to show how the existing photon and ion facilities can be used to feed the storage ring. A detailed design study for the ring is now in progress.

Future Direction in Applied Physics

A great need in the Applied Physics Program is the development of new methods for characterizing materials at spatial resolutions around 1 micrometer or less. Prospects for doing this by x-ray imaging seem to be limited by the difficulty encountered in the production of focussing x-ray optics of high quality. Thus, it is necessary to consider other types of approach to the problem.

A possible approach seems to be to adopt the methods of electron microscopy to the problem. In this case the x rays would be focussed as tightly as possible so that a spot with as high an x ray flux is formed. Rather than raster a target past this x ray beam in order to form an image it is possible to image the electrons produced in the irradiated area by using electron microscopy techniques. Focussing and imaging of electrons is a well studied problem that can easily be used at sub-micrometer resolutions. The field of photoelectron microscopy has been reviewed by Griffith (20). The high flux of synchrotron photons may now make it feasible to use electron spectroscopy/microscopy methods with high spatial resolution.

Professor A. Z. Hrynkiewicz has worked with the BNL group during several visits on the development of some of these ideas. What we propose to do is adapt (21) a Siemens ELMISCOP 1A transmission electron microscope for use at the synchrotron. In order to do this we discard the first sections of the machine that are used to form the electron beam. The synchrotron beam is then used to irradiate a sample at approximately the normal position. Electrons that are emitted are focussed with an immersion type lens and that produces an object for the remainder of the microscope. The imaged electrons can then be detected with a position sensitive microchannel plate array and the image stored in a computer for final enhancement and analysis.

Additional analysis of the electron optics will be done before going further with the project. The addition of electron energy analysis is desirable and a more refined analysis of the production rates and backgrounds would also be helpful. The results of our first assessment are very encouraging and we have hopes that we will be able to make images at the 100 nm resolution level with this instrument sometime in the future.

Conclusions

It is hoped that the material presented here will give some idea of the scope of work that the BNL Division of Atomic and Applied Physics is carrying out in atomic and applied physics using synchrotron radiation. Most importantly, we hope that the reader will feel the scientific content of the field is large and the questions to be answered important.

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Table II. Luminosities of photon-ion/atom interactions using 0.1% bandwidth synchrotron radiation with a photon current at the target of about 5.10^{15} photons/s, which is based on design parameters of the NSLS for a 5-pole superconducting wiggler, assuming 5 mr horizontal acceptance and full vertical acceptance.

System	Luminosity (cm^{-2} s^{-1})
Gas target (pressure = 10^{-6} torr)	$1.76 \cdot 10^{26}$/(cm length)
Singly-charged ion source (10 keV energy, 10 μA current)	$1.4 \cdot 10^{23}$ (for crossed beams of 1 mm height) $1.4 \cdot 10^{23}$/(cm merging length)(0.1 cm^2 beam area) $1.4 \cdot 10^{22}$/(cm merging length)(1 cm^2 beam area)
Kingdon Ion Trap (PHOBIS, see Ref. 22)	$5 \cdot 10^{22}$/(cm length)
ECRIS and EBIS (100 pnA, 10 keV/amu, for Ar^{+17})	$2.2 \cdot 10^{20}$ (for crossed beams of 1 mm height) $2.2 \cdot 10^{20}$/(cm merging length)(0.1 cm^2 beam area) $2.2 \cdot 10^{19}$/(cm merging length)(1 cm^2 beam area)
CHISR (10 MeV/nucleon)	
For $(\frac{\Delta p}{p}) = 10^{-2}$ (No point in bunching because space charge limit is exceeded)	
$N = 1.2 \cdot 10^{10}$ (Ar^{+17})	$2 \cdot 10^{22}/cm^2$ (1 cm merging length, 1 cm^2 area)
For $(\frac{\Delta p}{p}) = 2 \cdot 10^{-3}$	
$N = (8 \cdot 10^{8})$	$1.3 \cdot 10^{21}$ unbunched $1.6 \cdot 10^{22}$ bunched

Figure Captions

Figure 1. NSLS photon flux at the experimental position 20 m from the electron orbit. Units for the flux are shown at the top of the figure and are in photons/s/mA/eV/mm^2. The electron energy is 2.52 GeV. The curves show the effect of beryllium windows that are incorporated in the beam line and of additional aluminum filters that are used to harden the beam for specific experiments.

Figure 2. Plan and elevation views of the beam line showing the simplicity of the design and the in-line arrangement that makes it possible to do two experiments simultaneously.

Figure 3. Composite figure showing photographs of the experimental area. At the top left an overall view is given of an atomic physics experiment (a Penning ion trap in this case) and the x-ray fluorescence apparatus at the right. The photograph at the lower left shows a view of the details of the x-ray fluorescence apparatus. The figures at the right show x-ray fluorescence spectra obtained for a gelatin standard containing elements at the 10 parts-per-million (wet weight) level and from a thin section of kidney. Additional details are given for each spectrum in the figure.

Figure 4. Fluorescence spectrum recorded in the wave length region from 500 to 1500 A. The radiation was emitted following K-shell ionization using filtered synchrotron radiation (see Figure 1), (Ref. 13).

Figure 5. Distribution of charge states in argon following K-shell ionization using filtered synchrotron radiation. The ions were observed in a Penning ion trap (Ref. 16).

Figure 6. Distribution of charge states in argon following K-shell ionization using filtered synchrotron radiation. The ions were observed using a time-of-flight technique (Ref. 16).

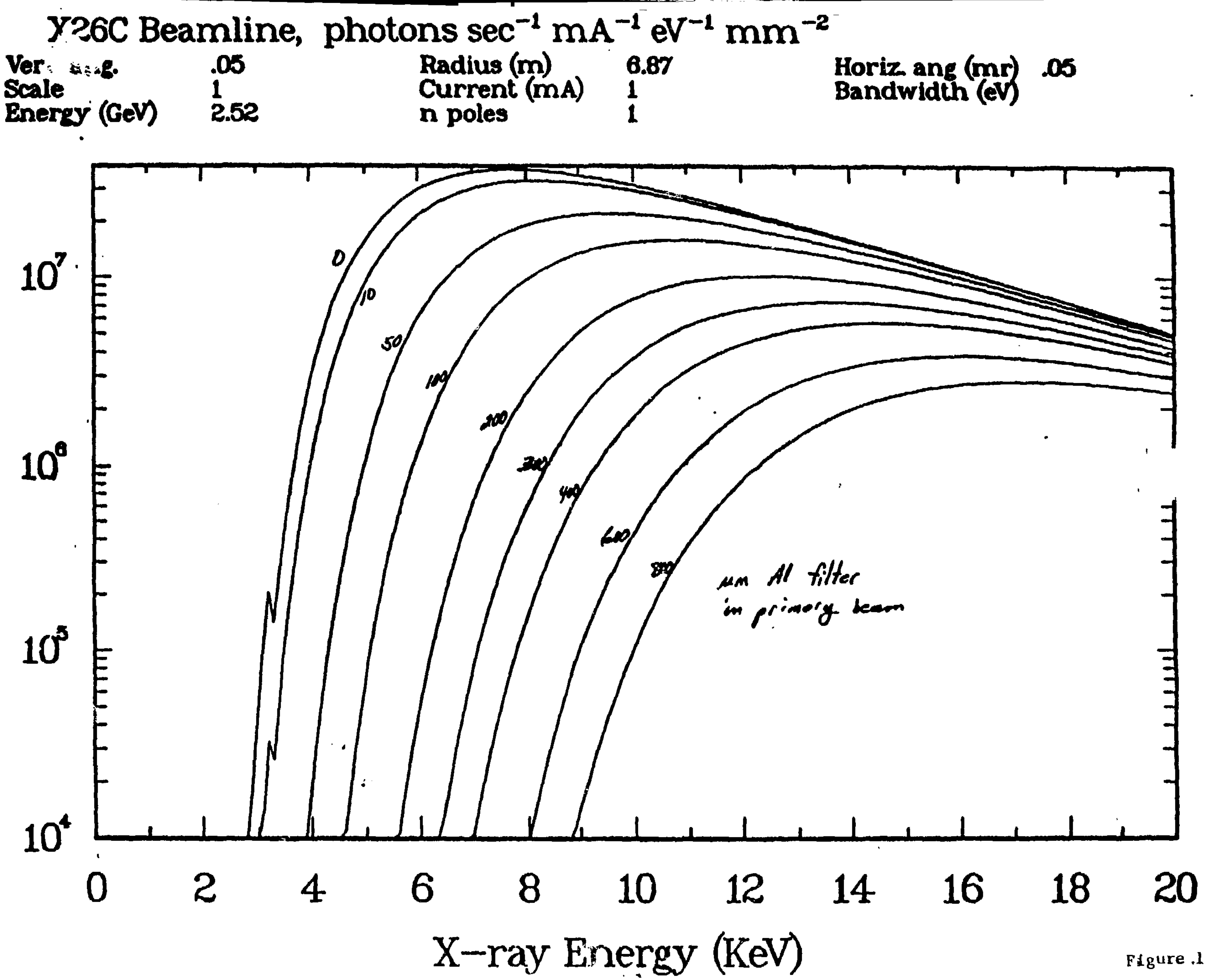
Figure 7. Minimum detection limits determined for trace elements in a 20-micrometer thick gelatin standard. Values are normalized to an electron current of 77 mA, target area of 2.5×10^{-5} cm^2, and counting time of 300 s.

Figure 8. Photomicrograph of a coronal section of the rat cerebellar cortex. Features of interest and a scale are indicated in the photograph.

Figure 9. Photomicrograph of a small section of the coronals ection of rat cerebellar cortex shown in Figure 8. The size and features are shown on the figure.

Figure 10. Distribution of iron in the cerebellar cortex in the region approximately the same as that shown in the photomicrograph of Figure 9. The light pixels show a higher concentration in the region of the fiber tract.

Figure 11. Map of Brookhaven National Laboratory showing the location of the National Synchrotron Light Source, Tandem Accelerator Laboratory, and the proposed heavy-ion storage ring. The length of the tunnel from the Tandem Laboratory to the storage ring is about 140 m.



Y26C Beamline, photons sec^{-1} mA^{-1} eV^{-1} mm^{-2}
.05
Scale 1
Energy (GeV) 2.52
Radius (m) 6.87
Current (mA) 1
n poles 1
Horiz. ang (mr) .05
Bandwidth (eV)
10^7
10^6
10^5
10^4
0
10
50
100
200
300
400
600
800
μm Al filter
in primary beam
0 2 4 6 8 10 12 14 16 18 20
X–ray Energy (KeV)

Figure .1

NSLS BEAM LINE X-26C FEBRUARY 1986

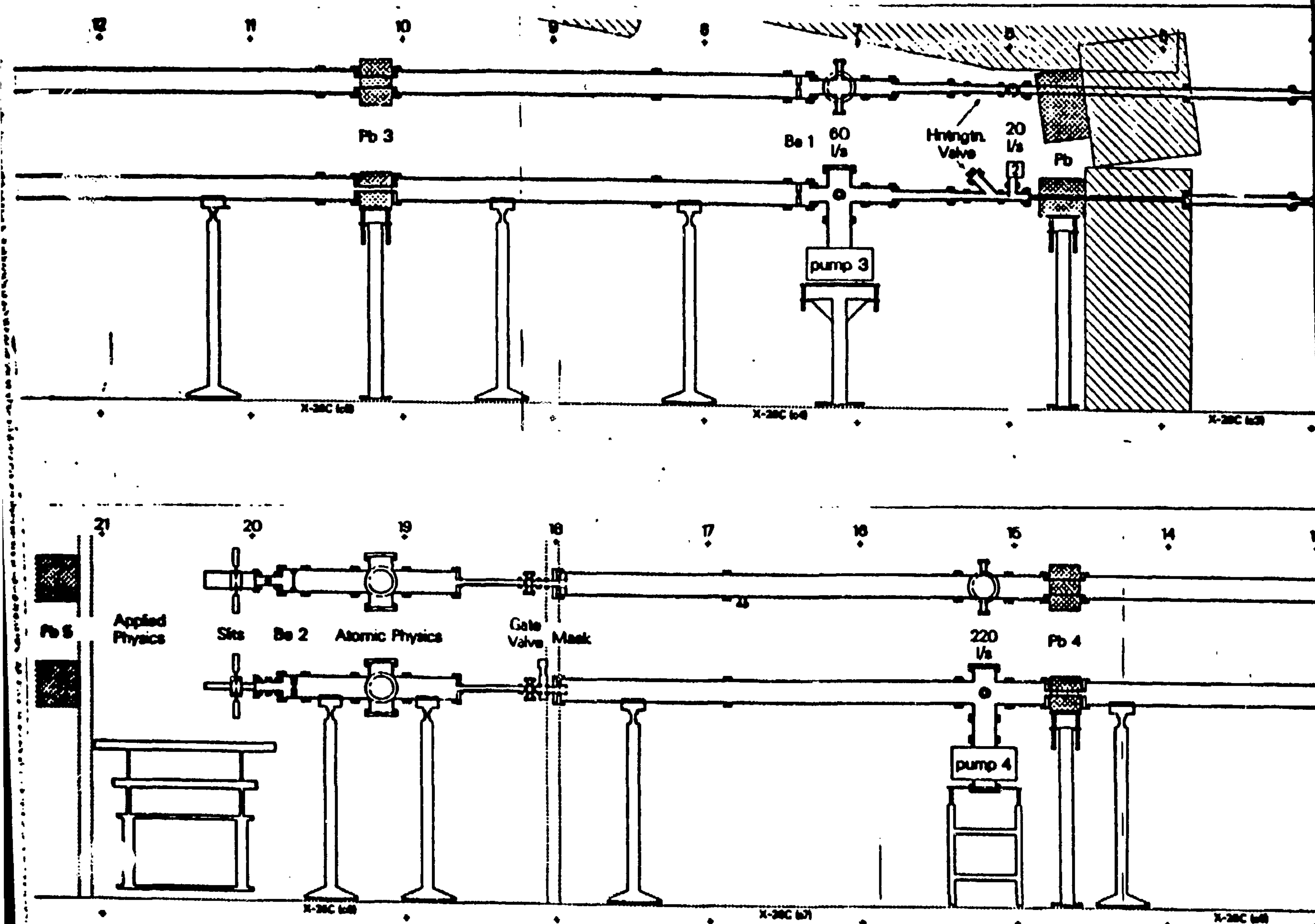


Figure 2

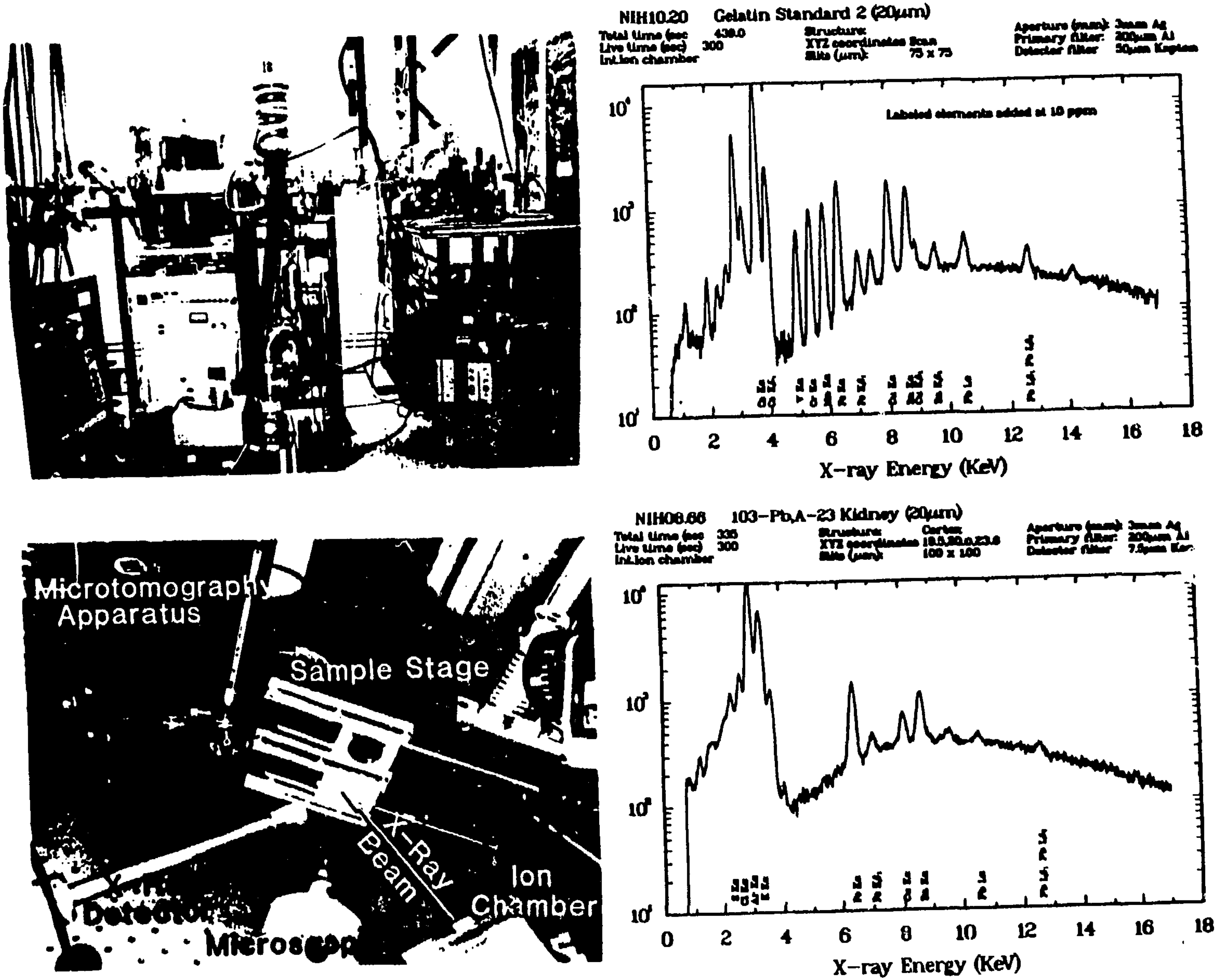
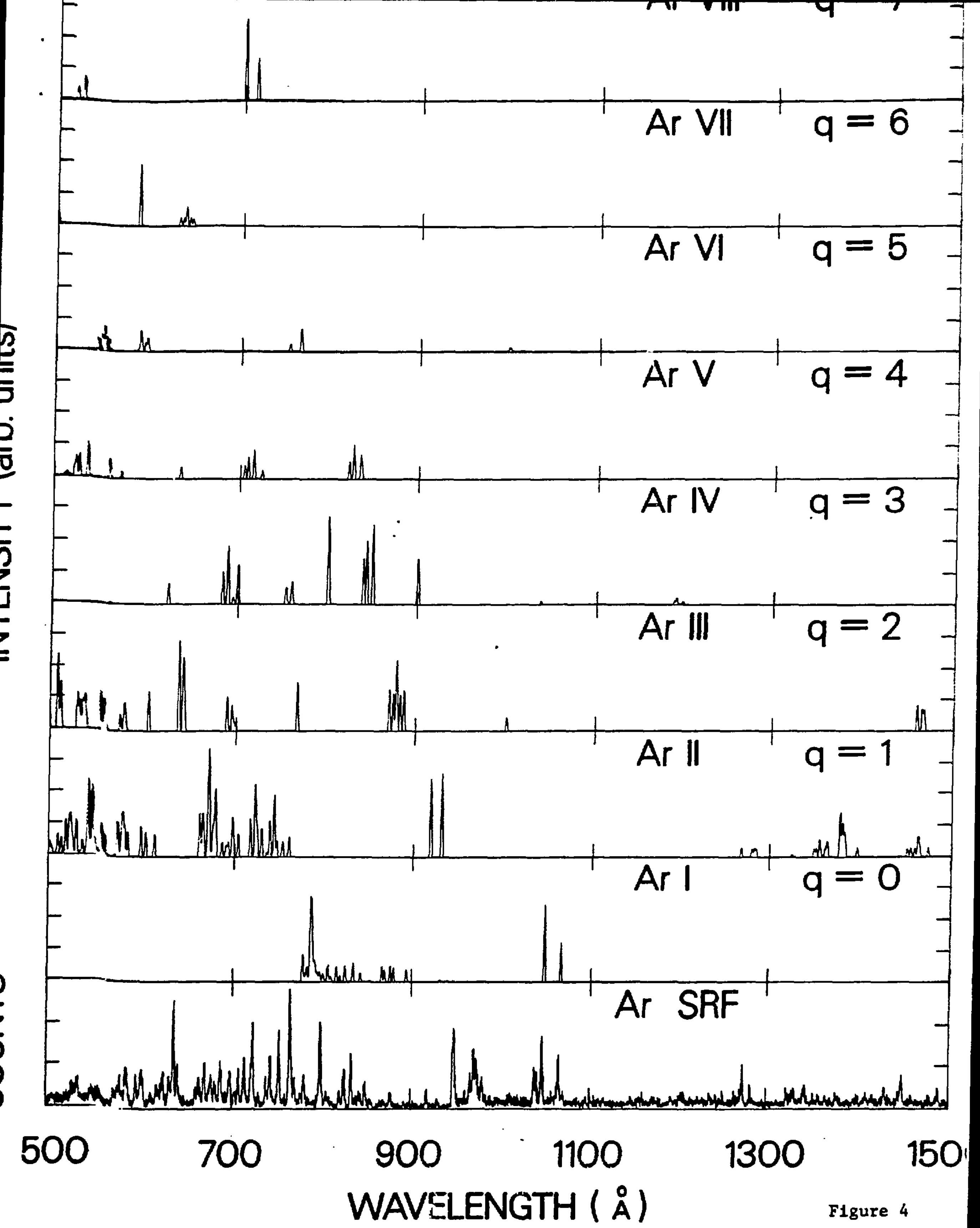

NIH10.20 Gelatin Standard 2 (20μm)
Labeled elements added at 10 ppm
X-ray Energy (KeV)
NIH08.66 103-Pb,A-23 Kidney (20μm)
X-ray Energy (KeV)
Microtomography Apparatus
Sample Stage
X-Ray Beam
Ion Chamber

Figure 3



Ar VII
q = 6
Ar VI
q = 5
Ar V
q = 4
Ar IV
q = 3
Ar III
q = 2
Ar II
q = 1
Ar I
q = 0
Ar SRF
500
700
900
1100
1300
WAVELENGTH (Å)

Figure 4

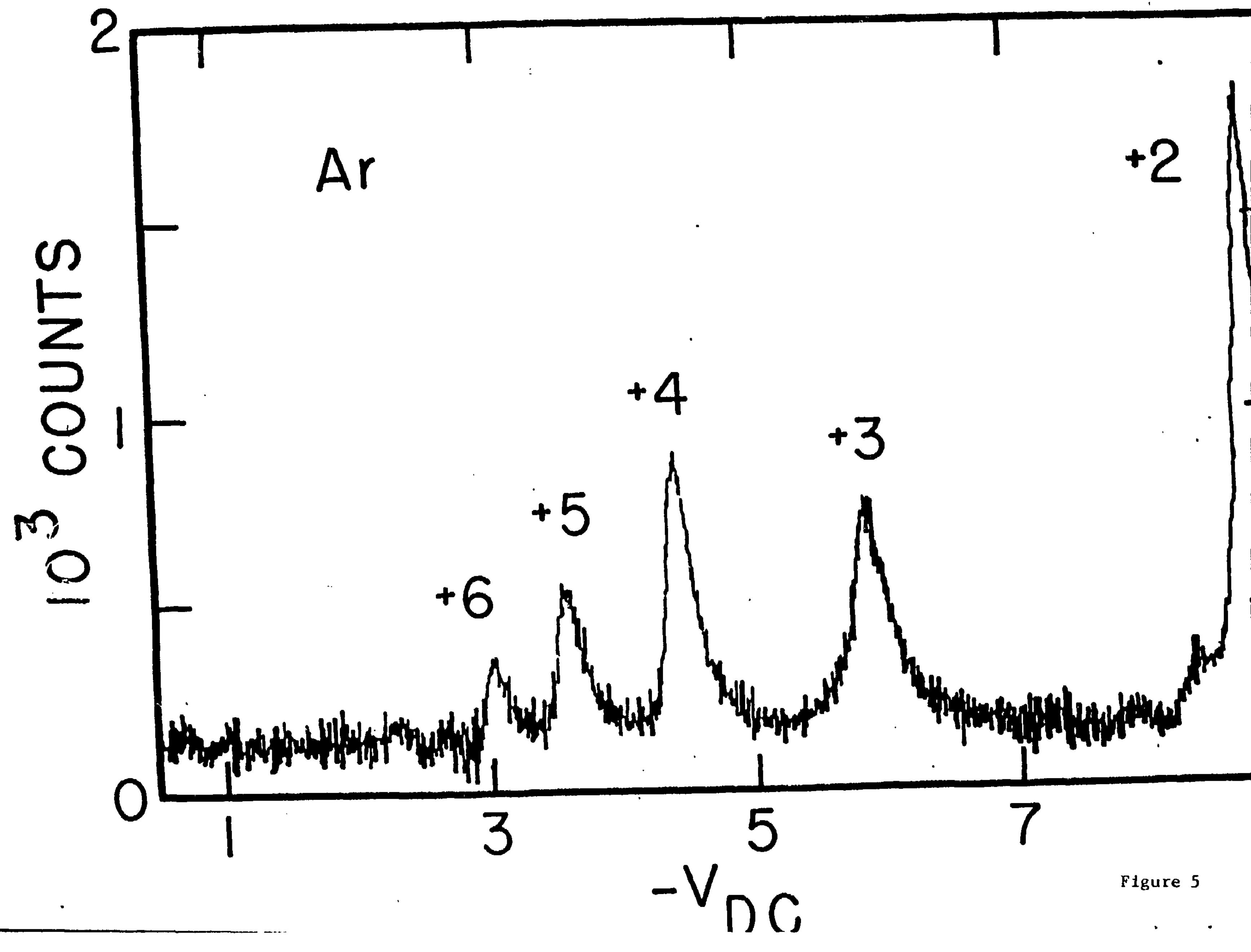
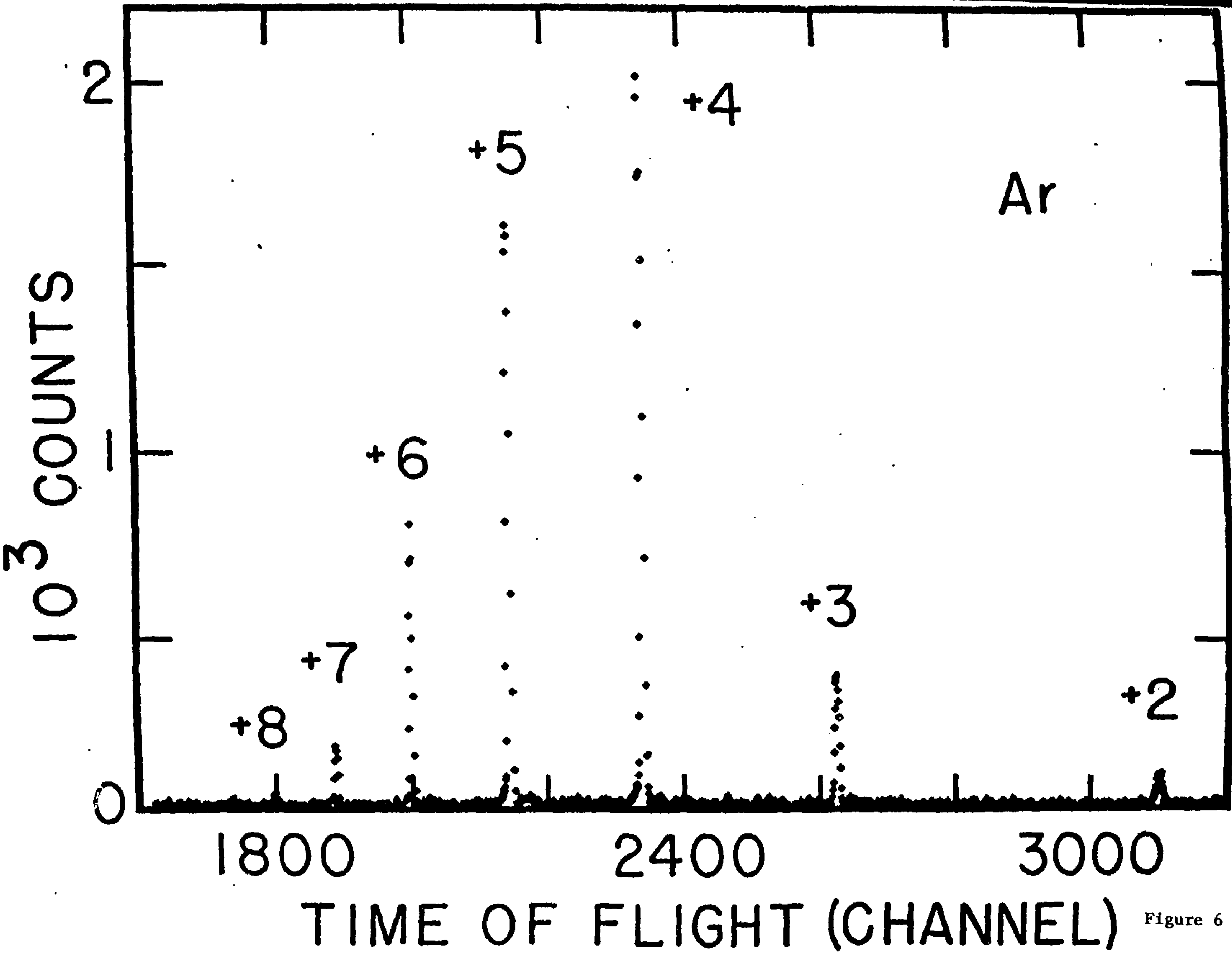


Figure 5



Ar
+2
+3
+4
+5
+6
+7
+8
10³ COUNTS
0
1
2
1800
2400
3000
TIME OF FLIGHT (CHANNEL)

Figure 6

MDL Curve for X-rays

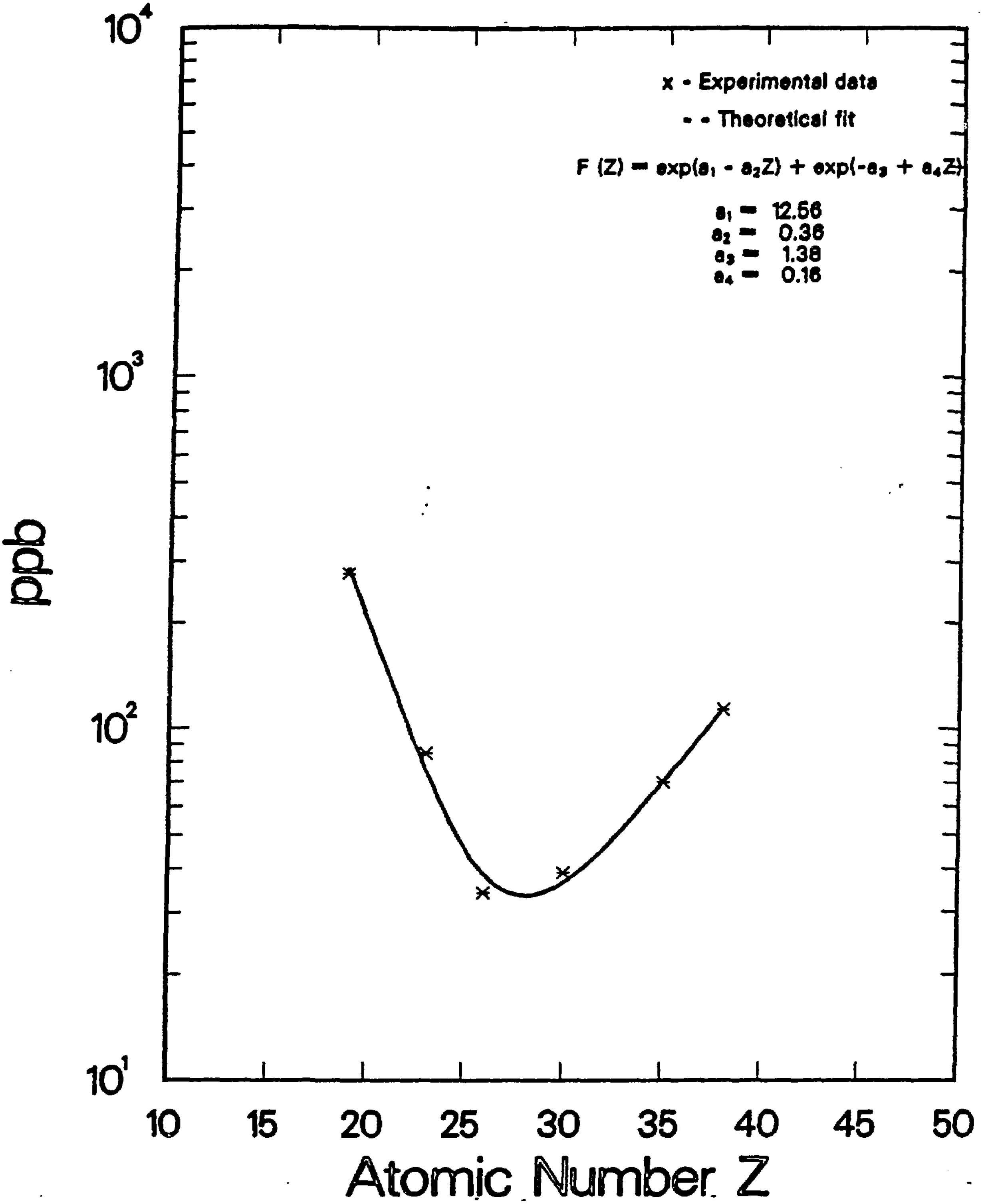


Figure 7

Figure 8

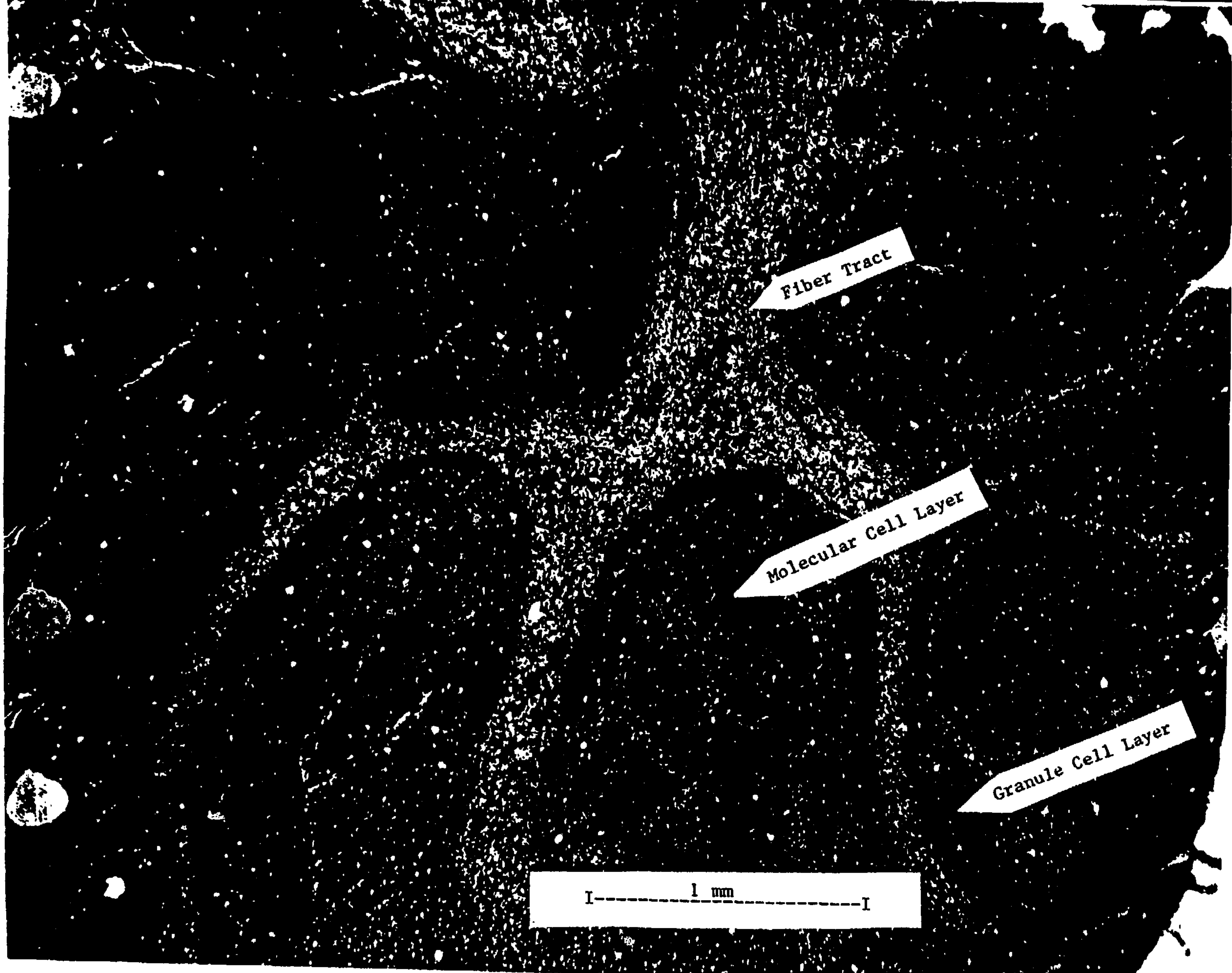


Figure 9

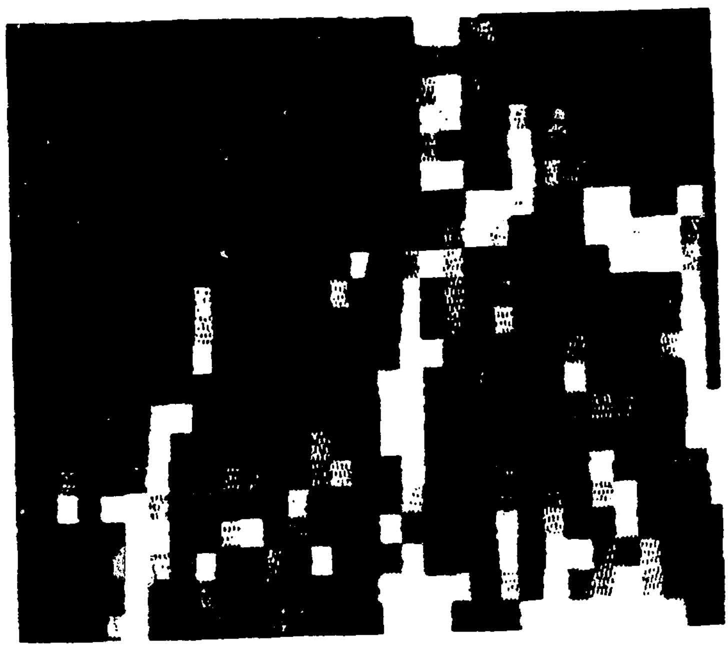


Figure 10

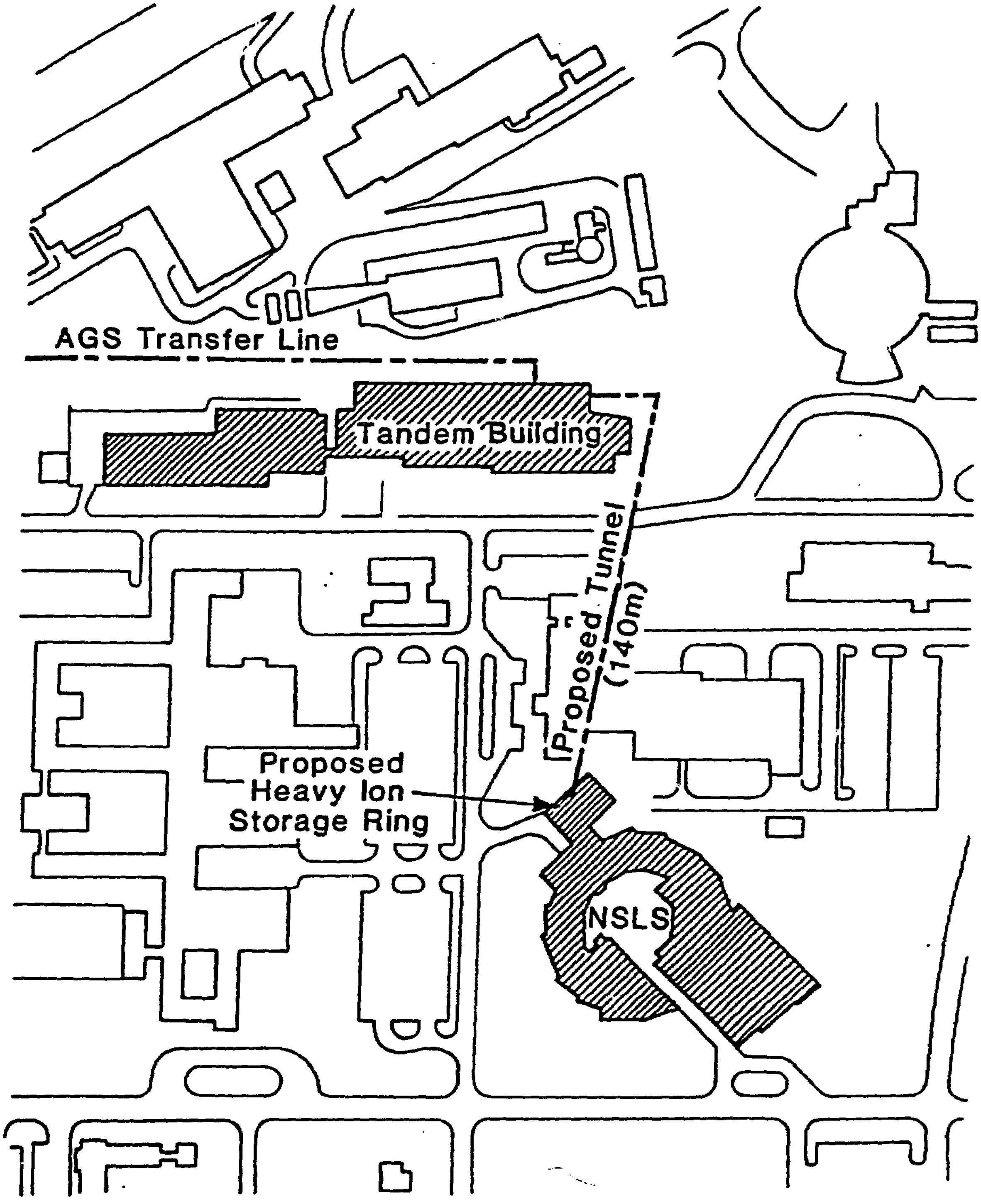

AGS Transfer Line
Tandem Building
Proposed Tunnel (140m)
Proposed Heavy Ion Storage Ring
NSLS

Figure 11