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The Production and Accumulation of Highly Charged Ions

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ATOMIC PHYSICS AND SYNCHROTRON RADIATION:
THE PRODUCTION AND ACCUMULATION OF HIGHLY-CHARGED IONS

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ABSTRACT

Synchrotron radiation can be used to produce highly-charged ions, and to study photoexcitation and photoionization for ions of virtually any element in the periodic table. To date, with few exceptions, atomic physics studies have been limited to rare gases and a few metal vapors, and to photoexcitation energies in the VUV region of the electromagnetic spectrum. These limitations can now be overcome using photons produced by high-brightness synchrotron storage rings, such as the x-ray ring at the National Synchrotron Light Source (NSLS) at Brookhaven. Furthermore, calculations indicate that irradiation of an ion trap with an intense energetic photon beam will result in a viable source of highly-charged ions that can be given the name PHOBIS: the PHOton Beam Ion Source. Promising results, which encourage the wider systematic use of synchrotron radiation in atomic physics research, have been obtained in recent experiments on VUV photoemission and the production and storage of multiply-charged ions.

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1. Historical Perspective

Atomic-physics research with accelerators can be classified into subfields under four main headings: (1) High Charge-State Collisions (including ion-solid, ion-gas, ion-ion, and ion-electron interactions), (2) Beam-Foil (or Fast-Ion) Spectroscopy, (3) Highly-Charged Ion Source Development, and (4) Synchrotron Radiation Research. Although theoretical considerations of atomic structure, as well as excitation and deexcitation of atomic and ionic systems, are common to all of these subfields, the experimental methods, accelerator facilities employed, and foci of attention were previously disparate with distinct practitioners. Recent developments in synchrotron radiation research, however, are paving the way for the convergence of these subfields into a single, unified, and vital experimental research area, which might ultimately be investigated at a single Atomic Physics Facility[1]. This concept is illustrated graphically in fig. 1. Before discussing the illuminating recent accomplishments and the bright future of synchrotron radiation research, let us first look back at the diverse inceptions of the aforementioned subfields of accelerator-based atomic physics.

1.1 High Charge-State Collisions

Prior to the first "small" accelerator conference at Oak Ridge in 1968, atomic collision experiments were performed only in modest self-contained laboratories on neutral or low-charge-state atomic systems. Although x-ray production cross sections were measured for incident heavy ions as early as 1962[2], typical ion-beam energies in the early and mid 60's were well below 3 MeV, and higher-energy accelerators were (for collision studies) the exclusive domain of nuclear and elementary particle physicists.

Then in the late 60's and early 70's, few adventurous nuclear physicists began to investigate K-shell ionization by heavy ions at energies above 3 MeV. In x-ray spectra recorded with nondispersive Si(Li) detectors, researchers observed energy shifts in x-ray lines produced by heavy- as opposed to light-ion bombardment of solid targets[3]. The magnitude of the shift was attributed to varying degrees of multiple ionization. This contention was conclusively demonstrated when higher-resolution crystal spectrometers began to be used[4]. Soon many laboratories around the world were actively involved in this new area of research, which became known as accelerator-based atomic physics.

1.2 Beam-Foil Spectroscopy

About five years before the early days of high-energy collision work, the field of beam-foil spectroscopy was developing independently at other laboratories[5]. As the name implies, the major thrust of this work was spectroscopic studies of excited states in ionic systems produced by the beam-foil interaction. In addition, the beam-foil time-of-flight method was developed to experimentally determine lifetimes and absorption oscillator strengths (f values) for particular excited states and transitions. This field is now often renamed "fast-ion spectroscopy" because experimenters have sought alternative methods, such as beam-laser studies, to selectively excite particular states of interest[6]. The limiting factor has now become the low incident photon energy of laser light which limits studies to excitation of only weakly bound electrons.

1.3 Highly-Charged Ion Source Development

The conventional way to produce highly-charged ions for atomic physics studies is by acceleration to high energy followed by foil stripping, but in 1969, the concepts of so-called "novel" ion sources were established[7]. By

1972 the first cryogenic Electron Beam Ion Source (EBIS) was created[8], and by 1980 Electron Cyclotron Resonance Ion Sources (ECRIS) were operational[9]. After many years of development, several versions of these sources now operate routinely and as Arianer[10] remarked in a recent review: "there appears to be an unceasing enlargement of their possibilities". Also mentioned[10] was the concept for yet another novel ion source, PHOBIS: the PHOton Beam Ion Source[11,12].

1.4 Synchrotron Radiation Research

The importance of synchrotron radiation (SR) to atomic physics research was first demonstrated in 1963 in the pioneering study of autoionizing states of helium by Madden and Codling[13]. Since then, as reviewed recently by Crasemann and Wuilleumier[14], there has been considerable activity in the areas of absorption and photoelectron spectroscopy, and some promising beginnings in Auger and fluorescence spectroscopy as well as in X-ray scattering. Although much has been accomplished, "hard synchrotron light is not yet being used nearly as widely in atomic and molecular physics as in other areas such as surface science, solid state physics and biology"[14]. For example, in the recently released Users Manual[15] for the National Synchrotron Light Source (NSLS) at Brookhaven only two of the forty-five operational beam lines on the VUV and X-ray rings are listed as having atomic physics as a primary research area. The narrow scope of earlier investigations of atomic physics with SR is attributable to the limited photon flux and energy range of earlier synchrotrons and to the relatively small community of researchers. To date, most studies have used rare gases and a few metal-vapor targets, only monochromatic incident radiation, and mostly VUV and soft x-ray radiation, that is only low-Z atomic systems and/or outer-shell excitations.

2. Introduction

In a 1980 workshop[16] on the possibilities for research in atomic physics at the NSLS many potential areas for study were identified and plans for building a versatile atomic physics beam line on the x-ray ring at the NSLS were presented[17]. NSLS Beam Line X-26C is now operational for this purpose. Since most atomic physics studies use thin gas targets, the downstream beam is still useable, therefore X-26C was designed to allow for simultaneous applied physics studies[18]. Here we will review the first three atomic-physics studies performed on X-26C. Although the experimental equipment and techniques were distinct, the primary research goal was common, namely to study the production of highly-charged ions produced by hard synchrotron radiation and thereby to develop the concept for PHOBIS: the PHOTon Beam Ion Source.

Calculations of expected PHOBIS performance (using Ar gas at a pressure of 1×10^{-8} torr as an example[11,12]) indicate that if a suitable ion trap were irradiated with truly white SR, then as many as 10^7 Ar¹⁸⁺ ions/cm could be produced and confined after a few seconds trapping time. The incident photon flux used for these calculations was obtained assuming a stored electron beam current of 500 ma, a 25-pole wiggler, and focussing optics to deliver maximum flux at the target. Presently the operational electron beam current is typically 120 ma, and beam port X-26C is a bending magnet (1 pole) port without focussing optics. Furthermore the calculations assumed truly white radiation containing lower energy photons which efficiently remove outer-shell electrons, but vacuum considerations currently require the use of one or two beryllium windows. This cuts off the spectral distribution below 3.2 keV, as shown in fig. 2. These factors reduce effective photon flux by three or four orders of magnitude, which is still sufficient to develop the

basic principle of PHOBIS. Encouraging results have been obtained for synchrotron radiation fluorescence spectroscopy, recoil-time-of-flight studies, and the use of a Penning ion trap.

3. Experimental Results

Over the past few months three experimental programs in atomic physics research with SR have been initiated to study various aspects of the excitation and ionization of argon produced by filtered white synchrotron radiation. First, VUV photoemission spectroscopy has revealed that numerous highly-excited states in many ionization stages are simultaneously produced by primary excitation, by multiple Auger and shakeoff processes, and perhaps by secondary ion-atom interactions. Second, recoil-time-of-flight studies have shown that Ar^{4+} is the most probable charge state produced, and that Ar^{5+} and Ar^{6+} are more probably produced than Ar^{3+} . Third, Penning ion trap experiments have demonstrated that it is indeed possible to trap ions at room temperature, although successive photoionization to very high charge states will have to await higher photon flux.

3.1. Synchrotron Radiation Fluorescence Spectroscopy

The bottom of fig. 3. shows a typical VUV photoemission spectrum[19] of Ar produced by filtered white SR (two Be windows, see the lowest curve in fig. 2). This spectrum was recorded using a normal incidence grating spectrometer[20] positioned at 90° to the incident photon direction. The Ar pressure in the excitation region was 2×10^{-2} torr, the average incident flux integrated over all energies was 2×10^{15} photons/sec, and the spectrum shown in fig. 3 required over 24 hours of accumulation time.

The other spectra in fig. 3 illustrate the predicted appearance of lines from individual ionization stages of Ar. These spectra were created by taking tabulated values for wavelengths and relative intensities from Kelly[21], and

folding in a spectrometer instrumental resolution FWHM of 2 Å (assuming Gaussian line shapes). The relative intensities are not necessarily relevant to SR production, because they are derived from compilations of previous experimental results for plasma discharges, sparks, exploding wires, etc.

In addition, both in the compilation[21] and in fig. 3, the intensities are normalized to the strongest line for each ionization stage, and there has been no attempt to reflect the charge state distribution produced by SR. Careful visual examination and quantitative analysis[19] of the spectra shown in fig. 3 reveals that: (1) many lines in the SR spectrum can be conclusively identified, (2) the relative intensities for transitions of the same ionization stage differ substantially from the tabulated values[21], (3) many lines in the SR spectrum, such as those centered about 970 Å, do not line up with any tabulated values, and (4) considerable excitation of neutral argon is observed as evidenced by the two strong lines at 1048.2 and 1066.2 Å, which are the transitions from the $3p^5(2p_{1/2,3/2})4s$ states to the $3p^6(1S_0)$ ground state in Ar I. This last observation seems surprising at first in light of the indication (mentioned above and discussed below) that Ar^{4+} (Ar V) should be the most probable charge state produced. Since high target pressures were used to achieve reasonable count rates, the mean free path for thermal velocity ions was less than 1 mm. Neutral argon atoms could therefore be excited by collisions with ions produced by the synchrotron radiation, rather than by the SR itself. Obviously, systematic studies as a function of pressure are needed, but again more photon flux would be required to allow significantly lower target pressures. This study of SR fluorescence spectroscopy has yielded interesting results, while future possibilities include time resolved lifetime studies and state selective excitation using tuneable radiation as more photon flux becomes available.

3.2. Production and Storage of Low-Energy Highly-Charged Ions

Based on the assumptions used in the PHOBIS calculations[11] discussed earlier, it was also shown[22] that a Penning ion trap might be used to produce and store as many as 5×10^3 ions with a single charge state, e.g. Ar^{17+} . It was further predicted that slightly lower charge states, e.g. Ar^{12+} , could be made and kept in the trap with mean energies as low as 0.025 eV or 300 K, that is at room temperature! Quite recently it has been demonstrated[23] that ions with recoil energies corresponding to room temperatures can be produced using filtered white and monochromatic x-rays from a wiggler port at the Stanford Synchrotron Radiation Laboratory (SSRL). Essentially the same recoil-time-of-flight apparatus used at SSRL was also employed in similar experiments[24] on beam line X-26C at the NSLS. For the case of Ar, nearly identical results were obtained for the charge state distribution, although somewhat different experimental techniques were employed[23,24].

Recently, the first Penning ion trap studies[25] with filtered white SR have been performed on X-26C. Fig. 4 shows the apparatus and a portion of the beam line. The experimental results, discussed elsewhere in these proceedings[26], verify the predictions that SR can be used to produce and trap ions at very low energies. Table I shows the comparison of measured charge state fractions for production only[23,24] and for production plus trapping[25]. Note that for Ar^{4+} and higher charge states the results are similar, but the fractions for Ar^{3+} and Ar^{2+} are considerably higher with trapping. The former observation reflects the insufficient photon flux to achieve successive photoionization, while the latter is attributed to charge exchange between ions and neutrals within the trap which reduces the mean charge state. The very large fraction observed for Ar^{2+} is attributed[26] to

a low charge exchange cross section for Ar^{2+} interacting with Ar^0 , which leads to a build up of Ar^{2+} as higher charge state ions quickly capture electrons, while progress to lower charge states is impeded.

4. Conclusions

The promising inaugural results from X-26C provide valuable scientific information, demonstrate the use of energetic SR to produce and study multiply-charged ions, and support further development of the PHOBIS concept. Moreover, the ability to produce and study highly-charged ions using SR represents the unifying bridge to consolidate the subfields of accelerator-based atomic physics and provides the opportunity for complementary studies of ion-photon, ion-atom, and ion-electron interactions at a single location. Although conventional ion sources could be used in conjunction with SR for a variety of ion-photon interaction studies, access to the widest range of atomic physics experiments would best be provided by a complete Atomic Physics Facility[1], which would result from the construction of a heavy ion storage ring at a synchrotron radiation facility.

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of immeasurable assistance in our previous ion-atom collision work; and who as a valuable friend will be sorely missed.

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Table I. Charge-state distributions for producing and storing Ar ions created by filtered white synchrotron radiation. Quoted relative intensities are normalized to 1 for Ar⁴⁺.

charge state	Production only ^{a)}	Production plus trapping ^{b)}
Ar ²⁺	0.06	2.46
Ar ³⁺	0.2	0.8
Ar ⁴⁺	1	1
Ar ⁵⁺	0.8	0.6
Ar ⁶⁺	0.3	0.3
Ar ⁷⁺	0.09	0.08
Ar ⁸⁺	0.01	0.05

a) Results of recoil-time-of-flight measurements [23,24].

b) Typical results of Penning ion trap studies [25,26].

Figure Captions

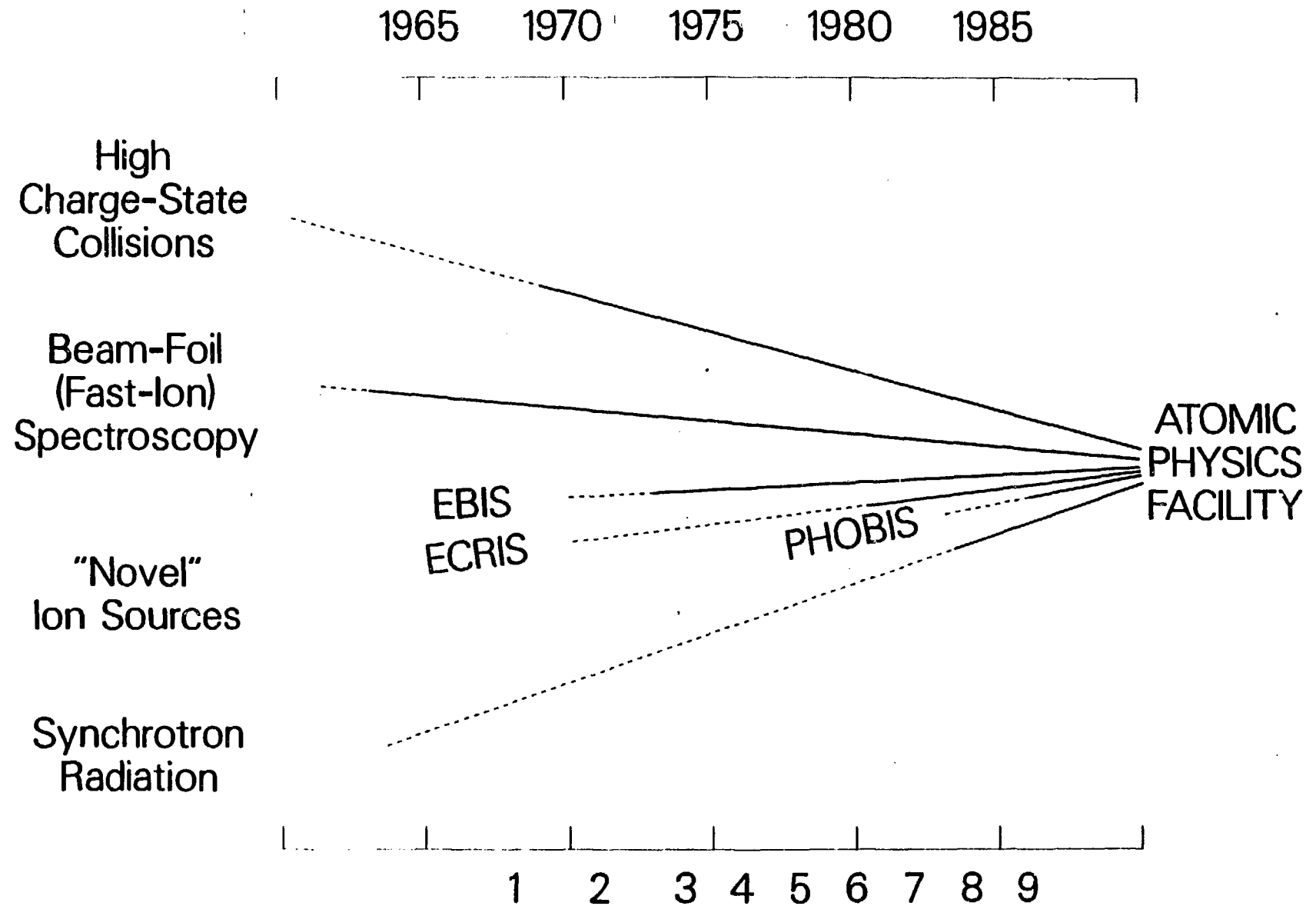
Figure 1. Graphical representation of the convergence of various subfields of atomic physics research. The dashed lines are drawn from the time early concepts were presented or early experiments with low beam energies or charge states were begun. Solid lines begin when high-charge state experiments commenced and then converge in the future upon the proposed Atomic Physics Facility[1], which would combine at a single location a source of highly charged ions, a heavy ion storage ring, and a synchrotron light source. The acronyms EBIS, ECRIS, and PHOBIS stand for Electron-Beam, Electron-Cyclotron-Resonance, and PHOton-Beam Ion Source, respectively.

Figure 2. Plot of the photon intensity for broad-band "white" synchrotron radiation produced by a bending magnet on the x-ray ring of the NSLS at Brookhaven[19]. The uppermost curve is the unfiltered spectrum, the middle and lowest curves show the effect of filtering with one or two 10 mil (254 micrometer) thick Be windows.

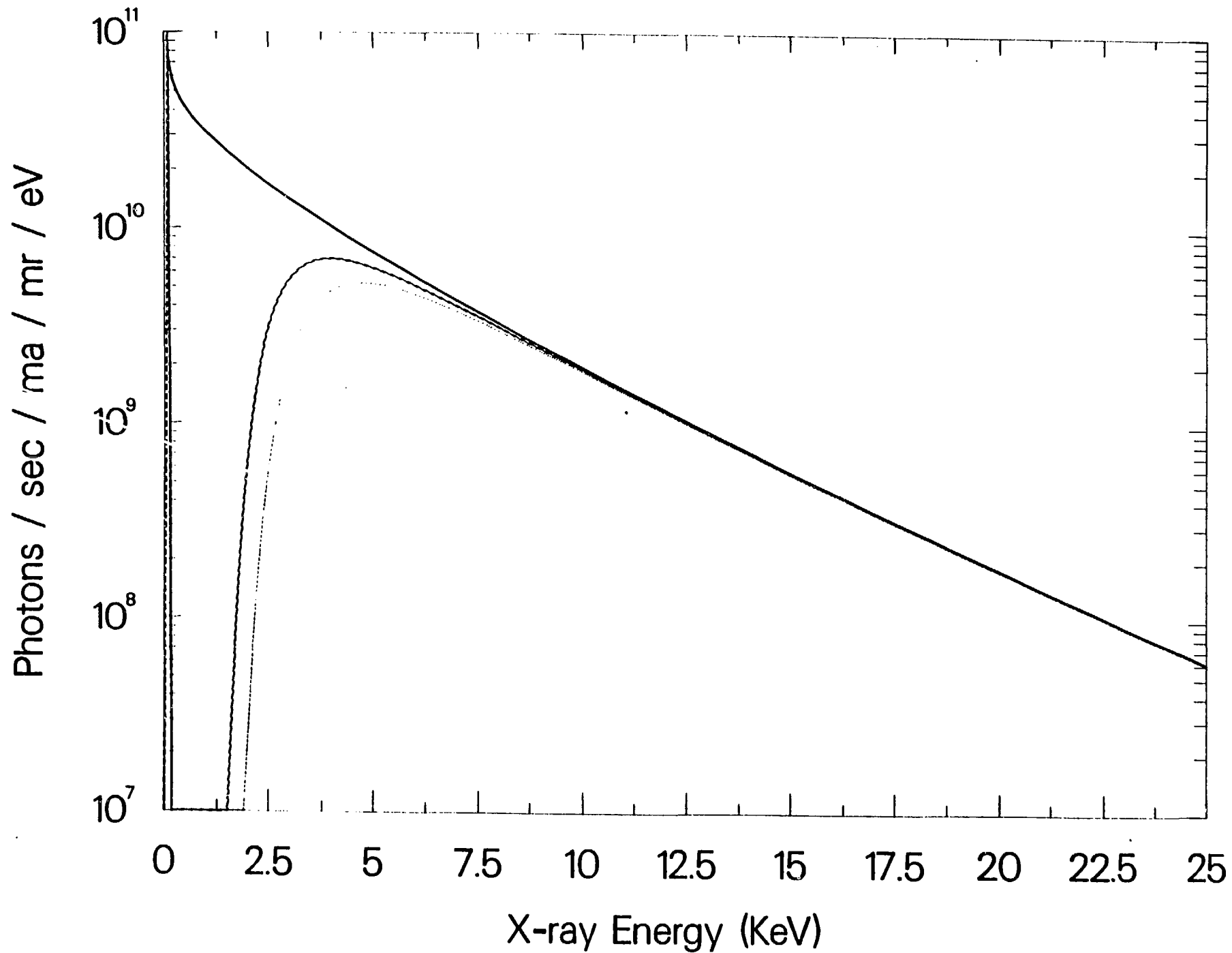
Figure 3. A portion of the VUV fluorescence spectrum of Ar (bottom) produced by filtered white synchrotron radiation from a bending magnet (beam port X-26C) on the x-ray ring of the NSLS at Brookhaven. The upper spectra show the predicted appearance of lines from individual ionization stages (Ar I to Ar VIII) or charge states ($q = 0$ to 7) of Ar created by folding the tabulated values of ref. [21] for wavelengths and relative intensities with a Gaussian peak shape assuming an instrumental resolution FWHM of 2 angstroms.

Figure 4. Photograph of the dedicated atomic physics beam line, X-26C, on the the x-ray ring of the NSLS at Brookhaven. Photons originate at the x-ray ring (to the left of the picture), filter through a 10 mil Be window (not shown), travel to the right, pass through apertures go through the experimental apparatus (Penning ion trap[25]), exit through another Be window and stop in a pile of lead bricks (not shown). The Penning trap itself is centered in the vacuum pipe between the 6" pole tips of the large electromagnet. The experimental module (from upstream valve to downstream Be window) is mounted on wheels to be completely removable for replacement with other modules such as fluorescence[19] or recoil-time-of-flight[23,24].

CHRONOLOGY OF ACCELERATOR-BASED ATOMIC PHYSICS RESEARCH

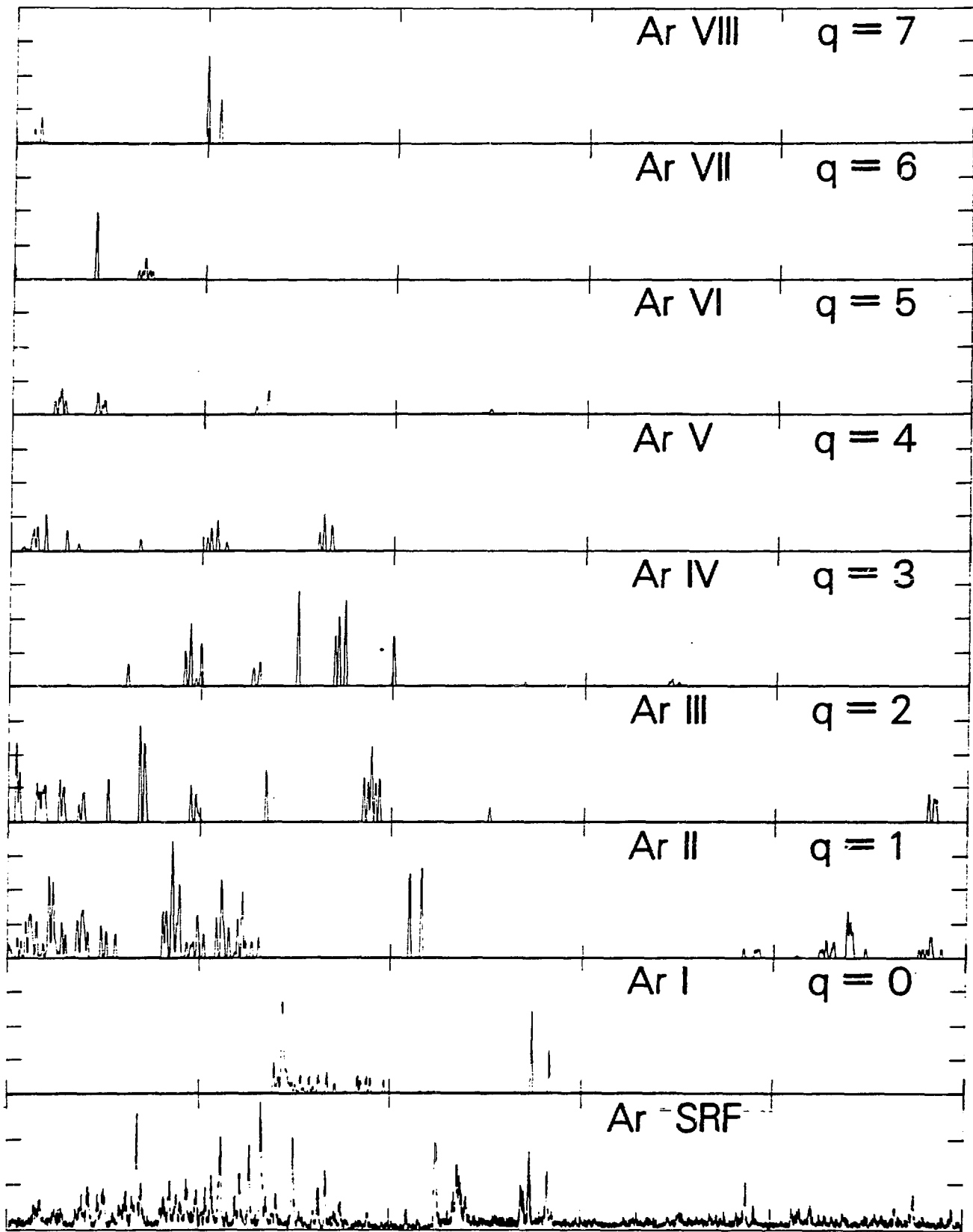


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