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THE ARGONNE PULSED NEUTRON SOURCE PROGRAM

by

J. M. Carpenter, R. K. Crawford, S. W. Peterson,
M. H. Mueller, J. D. Jorgensen, and A. H. Reis, Jr.

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J. M. Carpenter, R. K. Crawford, S. W. Peterson,
M. H. Mueller, J. D. Jorgensen, and A. H. Reis, Jr.
Argonne National Laboratory, Argonne, IL. 60439 U.S.A.

INTRODUCTION

Neutrons for thermal neutron scattering experiments and for basic fast neutron irradiation effects studies have typically been obtained from fission reactors. However, both types of research are limited by the available neutron flux which has reached $\sim 10^{15}$ in the case of current high flux research reactors, but is close to an effective upper bound determined by heat transfer limitations. To extend the range of problems which can be studied effectively, a new-generation neutron source is required.

The pulsed neutron source program at Argonne National Laboratory was established to develop a new-generation source based on the spallation principle for neutron production. Principles of pulsed spallation neutron source are illustrated in Fig. 1. Protons are accelerated in a synchrotron to a very high velocity and directed at one of two alternate heavy-metal targets. When struck by the high-energy protons, atomic nuclei in the target emit neutrons which are delivered in individual beams to many different experiments. Because the synchrotron produces the protons in bursts, the neutrons generated in this "spallation" process likewise are delivered in short high-intensity pulses. These pulses of fast neutrons (spectrum peaked at about 1 MeV) can be allowed to fall directly on a sample to produce radiation damage, or can be moderated by small hydrogenous moderators placed around the target to produce pulsed

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beams of thermal or epithermal neutrons ($\sim 10^{-3}$ – 10 eV) with pulses typically a few microseconds long. Such pulsed neutron beams are inherently prepared for efficient use with the "time-of-flight" (TOF) neutron scattering technique in which energies of neutrons are determined by timing their travel over a known path length.

The step-by-step approach to the development of such a source at Argonne is indicated in Table I. The Argonne Pulsed Neutron Source Program started in 1972. After successful prototype experiments (ZING-P in 1974-75), and the present operation of a more intense prototype (ZING-P'), funds have been requested of Congress by DOE for an Intense Pulsed Neutron Source (IPNS-I). This will be fully operational in 1981 and has, from its inception, been envisaged as a National Facility open to all interested users from universities, industry, and other National Laboratories.

ZING-P'

As shown in Table 1 some experience has already been gained with a pulsed beam during the ZING-P operation. The present source of neutrons, ZING-P', is a prototype spallation pulsed neutron source using 500 MeV protons from the Booster-II synchrotron directed onto a tungsten target. A beryllium reflector and three separately tailored polyethylene moderators provide two vertical and three horizontal pulsed beams of thermal and epithermal neutrons with provision for one cold moderator. When operating at design intensity, the peak thermal neutron flux will be typically 10^{14} n/cm^2-sec and the peak epithermal flux will be typically 2×10^{14} n/cm^2-sec-eV. By cooling one moderator, the thermal neutron spectrum in one beam can be shifted to lower energies to provide an intense beam of long-wavelength neutrons.

NEUTRON INSTRUMENTATION

The development of appropriate neutron scattering instrumentation to make efficient use of pulsed neutron beams is deemed to be equal in importance to source optimization. Current plans call for the construction and operation of seven different instruments on the neutron beams at ZING-P'. These instruments will be used in support of scientific programs, as well as to provide essential information for the development of IPNS-I instrumentation. Planned ZING-P' instruments include:

1) High Intensity Diffractometer
2) High Resolution Powder Diffractometer
3) Crystal Analyzer Spectrometer
4) Chopper Spectrometer
5) Single Crystal Diffractometer
6) Small-angle Neutron Diffractometer
7) Ultra Cold Neutron Bottle Experiment

At most only five of these instruments can be operated simultaneously at ZING-P'.

We will now briefly describe three of the above instruments, namely Nos. 2, 5 and 6.

a. High Resolution Powder Diffractometer. The principle involved in pulsed neutron powder diffractometry is that a polychromatic wave length distribution "scans" the various d spacings giving intensity peaks spread out in time. Thus a diffraction pattern is detected at a fixed angle by measuring intensity as a function of time of flight. The main instrumental variables influencing resolution are neutron pulse width and flight path length. A high resolution diffractometer, located on a horizontal beam at

ZING-P' is currently in operation. Its design parameters involve an incident flight path of 20 meters, a sample to detector distance of 1 meter and time focussed detector bank centered at scattering angles of 160° and 90° which yield 0.3% and 0.5% ($\Delta d/d$) resolution, respectively. This diffractometer is currently the highest resolution neutron diffractometer in the United States and because of the unique wavelength dependence of the time width of the source pulse, the resolution is independent of momentum transfer (at a given angle). The 160° bank (0.3% resolution) can cover a useful momentum transfer range of 2 - 21 Å^{-1}. Sample sizes up to 1 cm x 5 cm can be accommodated. Typical data collection times (for one given sample at one sample temperature, pressure, etc.) will range from 1 day to 1 week, depending on the sample. This instrument which was installed at ZING-P' in December 1977 is shown schematically in Fig. 2. A powder pattern of Si obtained with the 160° banks during the initial tune-up period is shown in Fig. 3. This pattern amply demonstrates the high resolution achieved with this instrument; peaks up to and beyond the (13,3,3) are fully resolved even though the instrument adjustments were not completely optimized at the time. Fig. 4 shows a pattern taken more recently of a metal storage hydride compound $LaNi_{4.5}Al_{0.5}D_{\sim 6}$. This pattern is currently being analyzed by the Reitveld method of profile analysis and use is being made of resolved peaks for a Fourier analysis.

b. <u>Single Crystal Diffractometer</u>. This instrument is under construction and is currently scheduled for installation at ZING-P' on a horizontal neutron beam tube late in 1978. Several unique design features are involved and these must be thoroughly tested before routine operation of such an instrument will be possible. The instrument design is based on the Laue technique which is not currently used in single crystal neutron diffractometry but which is ideally adapted to pulsed white beam operation. In this technique, a "white" incident neutron beam, with each pulse containing all neutron wavelengths between some effective cutoffs λ_1 and λ_2, is allowed to impinge on the sample. A large number of reciprocal lattice points of the crystal sample will satisfy the Bragg condition during this pulse, each for a different wavelength within this range and/or a different scattering angle. Time-of-flight analysis is used to separate scattering events corresponding to different wavelengths, while angular resolution is provided by the spatial resolution of the detector. The use of a large area position-sensitive detector coupled with TOF measurement makes it possible to resolve and collect simultaneously $10^2 - 10^3$ Bragg reflections. Since the broad wavelength band provides the necessary integration over the crystal mosaic spread, thus eliminating the usual scan, data gathering rates may be increased by factors of 10-100 over normal methods. The price that must be paid for this tremendous gain in rate of data accumulation is the need for a very fast and sophisticated electronic data acquisition system with a large memory.

The instrument being built for ZING-P' has a 20 cm x 20 cm x 2 cm thick position sensitive detector (∿2 mm x 2 mm resolution) mounted on a movable arm so that both scattering angles and the sample-detector distance can be independently varied (∿0 ± 150° and 0.2 - 1 meter, respectively). The incident flight path is 8.5 meters. Sample orientation will be automatically variable using a computer controlled two circle orienter. Depending on unit cell symmetry and size, from 20 to 100 different orientations will be required to collect all of the independent reflections for a given sample. The instrument is shown schematically in Fig. 5.

c. Small Angle Neutron Diffractometer. The position-sensitive detector and the data-acquisition system being developed for the Single Crystal Diffractometer will be used for a small fraction of the time in a different configuration on the same neutron beam to form the nucleus of a prototype Small Angle Neutron Diffractometer based on time-of-flight analysis. This prototype will provide the experience necessary for the design of the IPNS small angle instrument. Although the prototype instrument will probably not be well optimized for small-angle diffraction, a limited experimental program involving metallurgical and biological samples is planned. A schematic of this instrument is shown in Fig. 6.

IPNS

The IPNS-I facility is scheduled to begin operation in 1981. Like ZING-P', this facility will utilize protons from the Booster-II accelerator. However, unlike ZING-P', IPNS-I will be a fully instrumented research facility rather than a prototype intended primarily for development work. The IPNS-I target area will contain a U^{238} target with three moderators (one cold) supplying thermal and epithermal neutrons to twelve beams for neutron scattering. Peak thermal and epithermal fluxes of 7.5×10^{14} n/cm^2-sec and 2×10^{15} n/cm^2-sec-eV, respectively, are expected. A separate uranium target will provide fast neutrons ($\sim$1 MeV) for radiation damage studies, and the proton beam can be steered to this target when desired. This radiation effects source should provide a time-averaged fast neutron flux of 3.5×10^{12} n/cm^2-sec. The target area will be surrounded by a large experimental hall housing the flight paths and instruments for the neutron scattering experiments. This hall is part of the existing Zero Gradient Synchrotron (ZGS) facility as shown in Fig. 7. This figure also indicates that a planned second phase, namely IPNS-II, would make use of other areas of the ZGS Facility for both a High Intensity Synchrotron as well as targets for neutron generation.

Fig. 8 shows the projected spectrum for the IPNS-I neutron facility compared with that from both the ambient and the hot source at the best current research reactor, namely the High Flux Reactor at the Institute Laue-Langevin (ILL) in Grenoble, France. It can be seen that in the thermal range IPNS-I will be roughly equivalent to the current high flux reactors, while offering unprecedented opportunities in the utilization of the epithermal spectrum. Planned instruments will utilize both portions of the spectrum. It should be emphasized, however, that there are a number of experiments which can still best be carried out at steady-state reactor, hence the pulsed and steady-state neutron sources should nicely complement each other as major research tools for solid state, chemical, and biological science.

Table I

Argonne National Laboratory Pulsed Neutron Source Program

Facility	Protron Accelerator	Frequency c/sec	Protons/ Pulse	Proton Energy (MeV) and Target	Neutrons/ Proton	No. of Neutron Beams	Peak Thermal Neutron Flux (n/cm^2-sec)	Operation
ZING-P	ZGS Booster I	10	2.5×10^{10}	200 Pb	2	2	5×10^{11}	Jan.1974
ZING-P'	ZGS Booster II	30	1×10^{12}	500 W	8	4	10^{14}	Nov.1977
IPNS-I	ZGS Booster II	45	3×10^{12}	600 U^{238}	25	12	7.5×10^{14}	Apr.1981
IPNS-II	High Intensity Synchrotron	60	5×10^{13}	800 U^{238}	30	12	10^{16}	To be determined

FIGURE CAPTIONS

Fig. 1. Schematic of Intense Pulsed Neutron System for Neutron Scattering and Radiation Damage Studies.

Fig. 2. High Resolution Powder Diffractometer installed at ZING-P'. Instrument has a 0.3% resolution.

Fig. 3. Silicon Powder Pattern from ZING-P' High Resolution Powder Diffractometer.

(a) Plot of raw data and calculated profiles based on an asymmetric Gaussian.

(b) Difference plot between raw data and calculated profiles.

Fig. 4. Powder Pattern of $LaNi_{4.5}Al_{0.5}D_{\sim 6}$ obtained on ZING-P' High Resolution Powder Diffractometer from the 160° detector bank.

(a) Region from d = 1.64 to 0.93 Å.

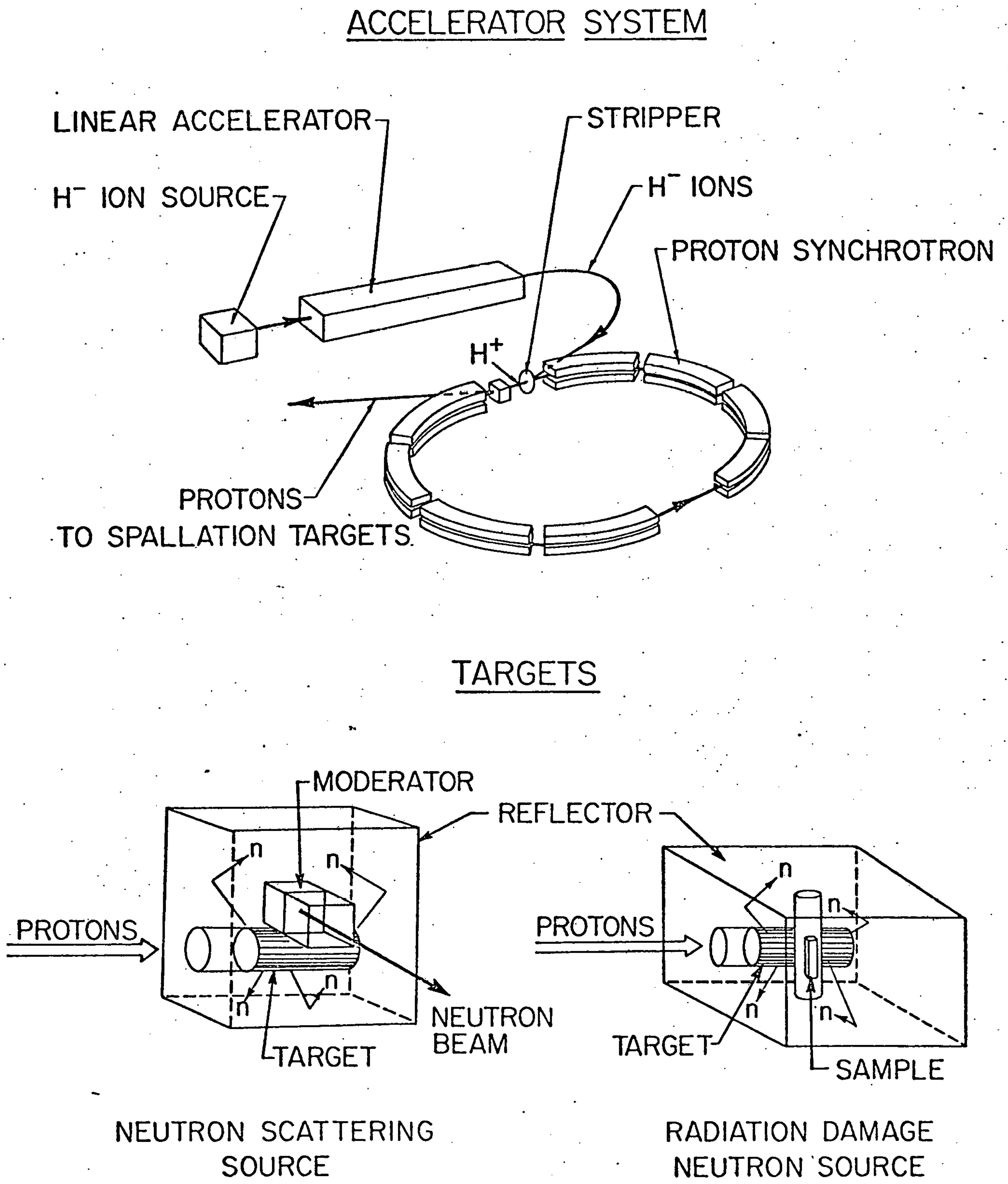
(b) Region from d = 0.93 to 0.62 Å and beyond.

Fig. 5. Pulsed neutron single crystal diffractometer.

Fig. 6. Schematic diagram of IPNS Small Angle Diffractometer. The collimating system is represented only by entrance and exit aperatures, but channels are assumed to be continuously defined.

Fig. 7. IPNS site layout in present High Energy Physics complex.

Fig. 8. Neutron source spectra for IPNS and ILL.



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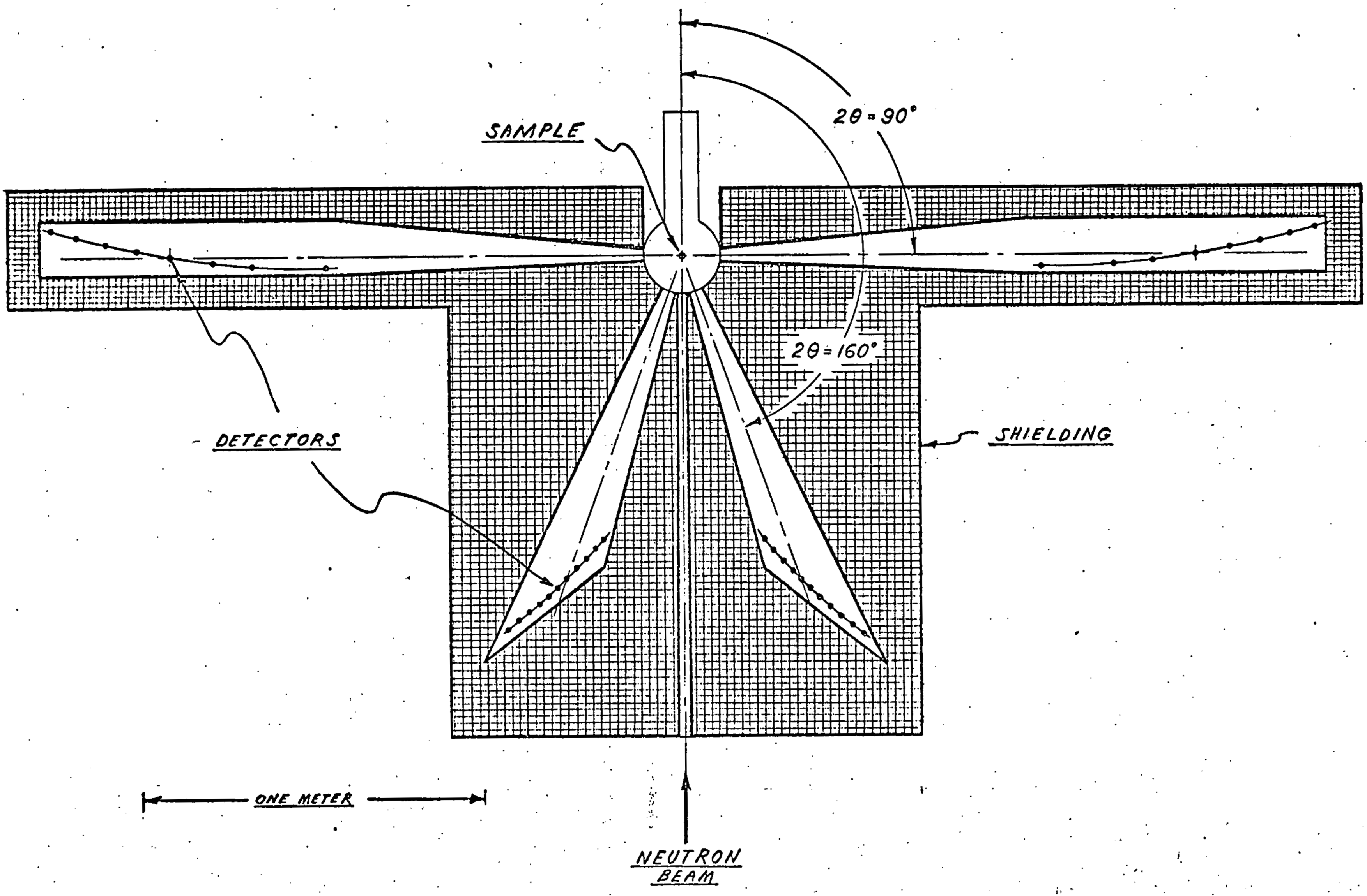

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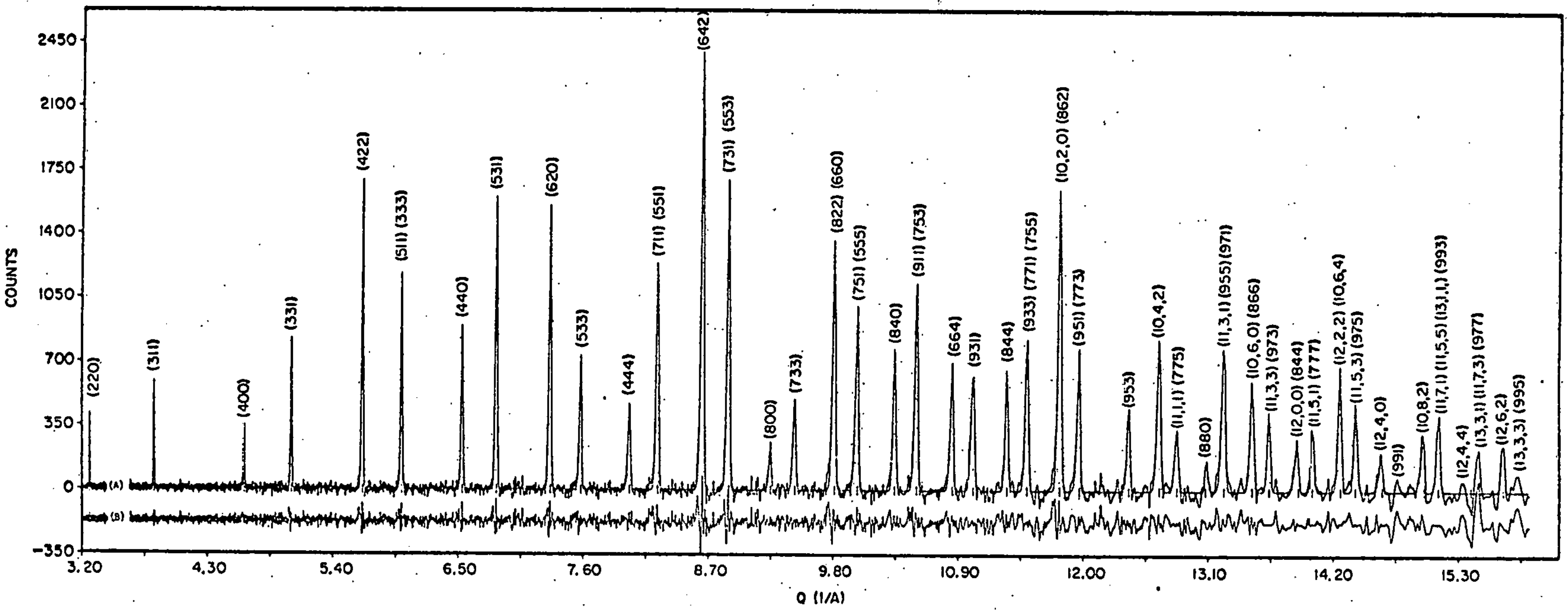

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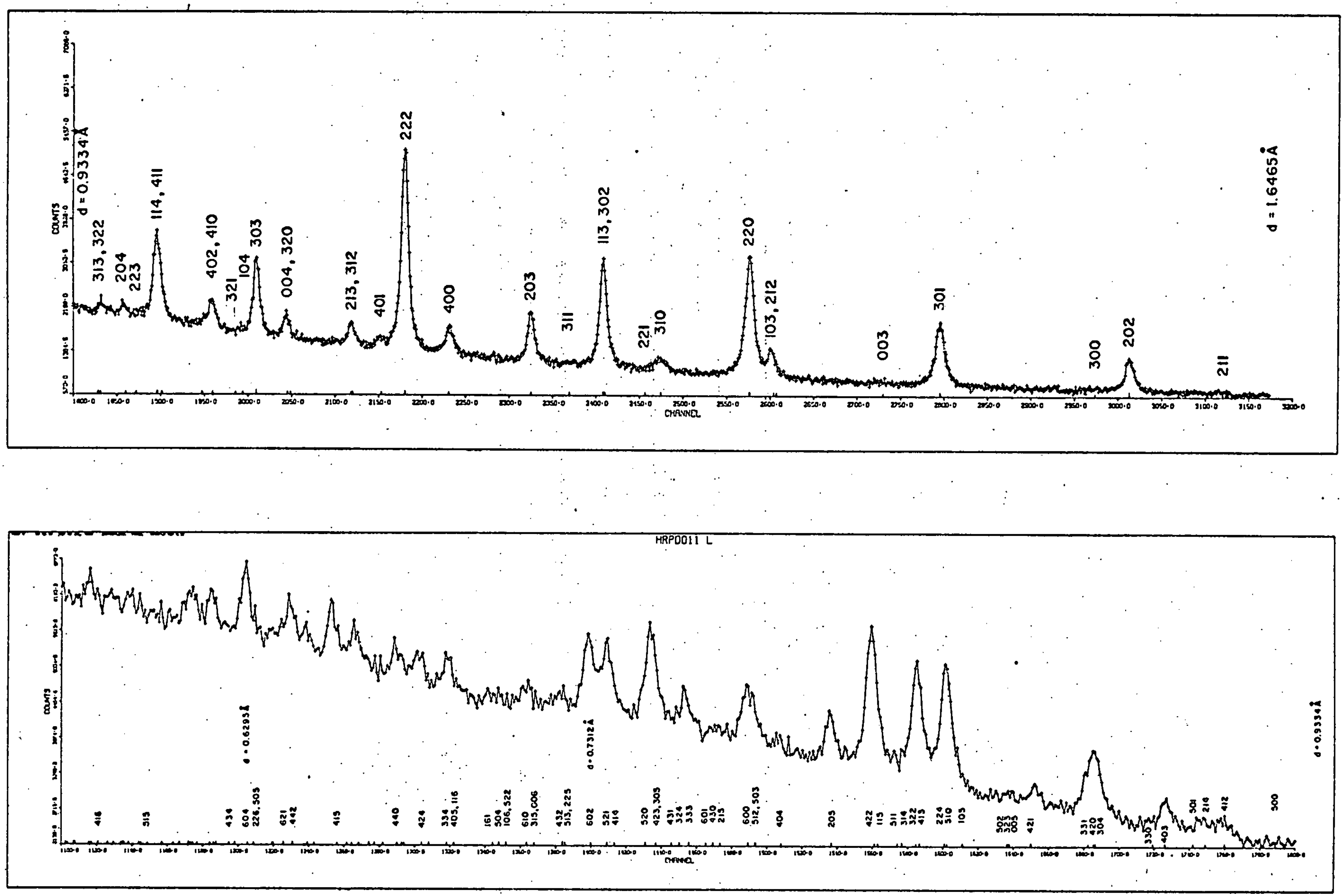

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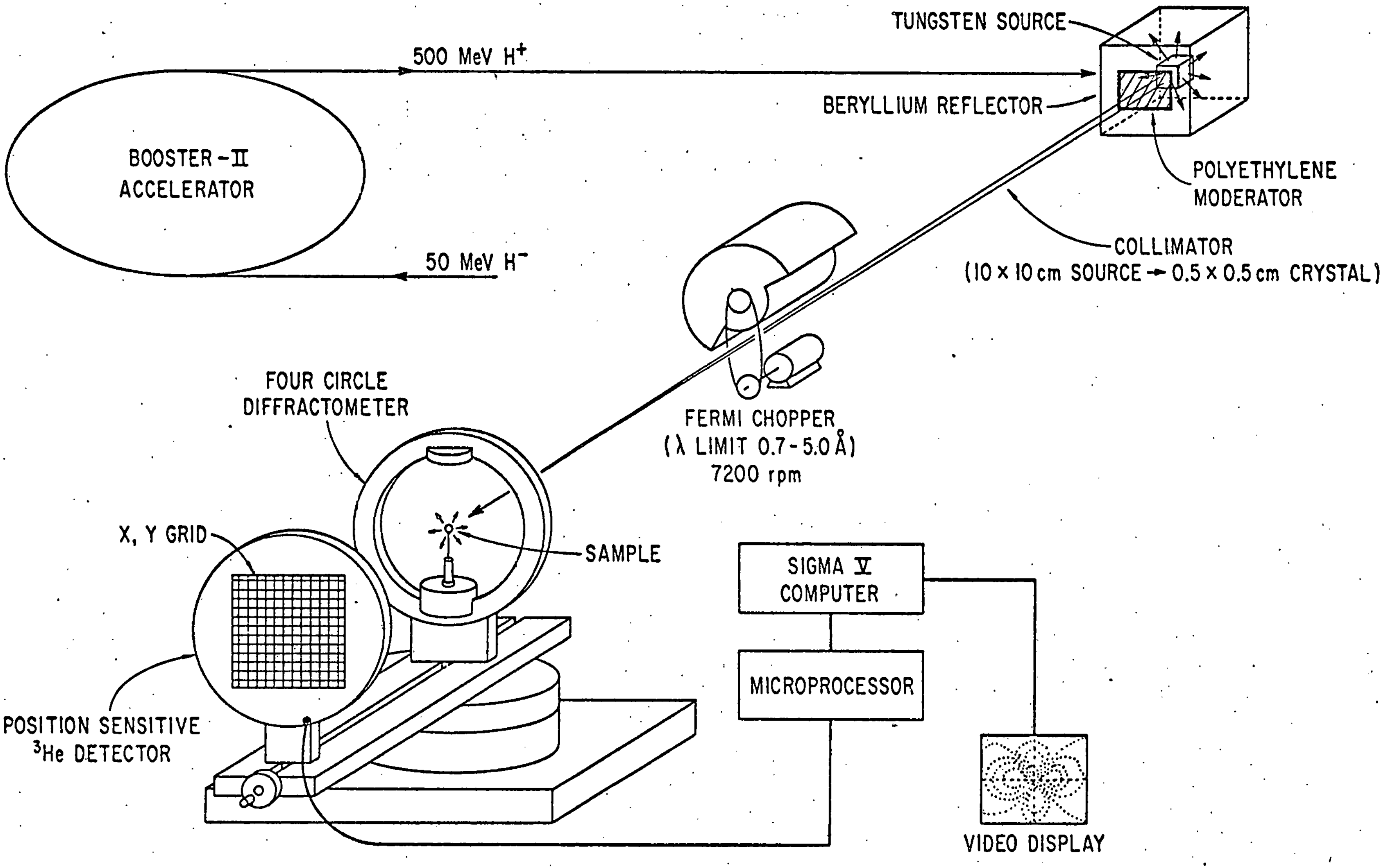

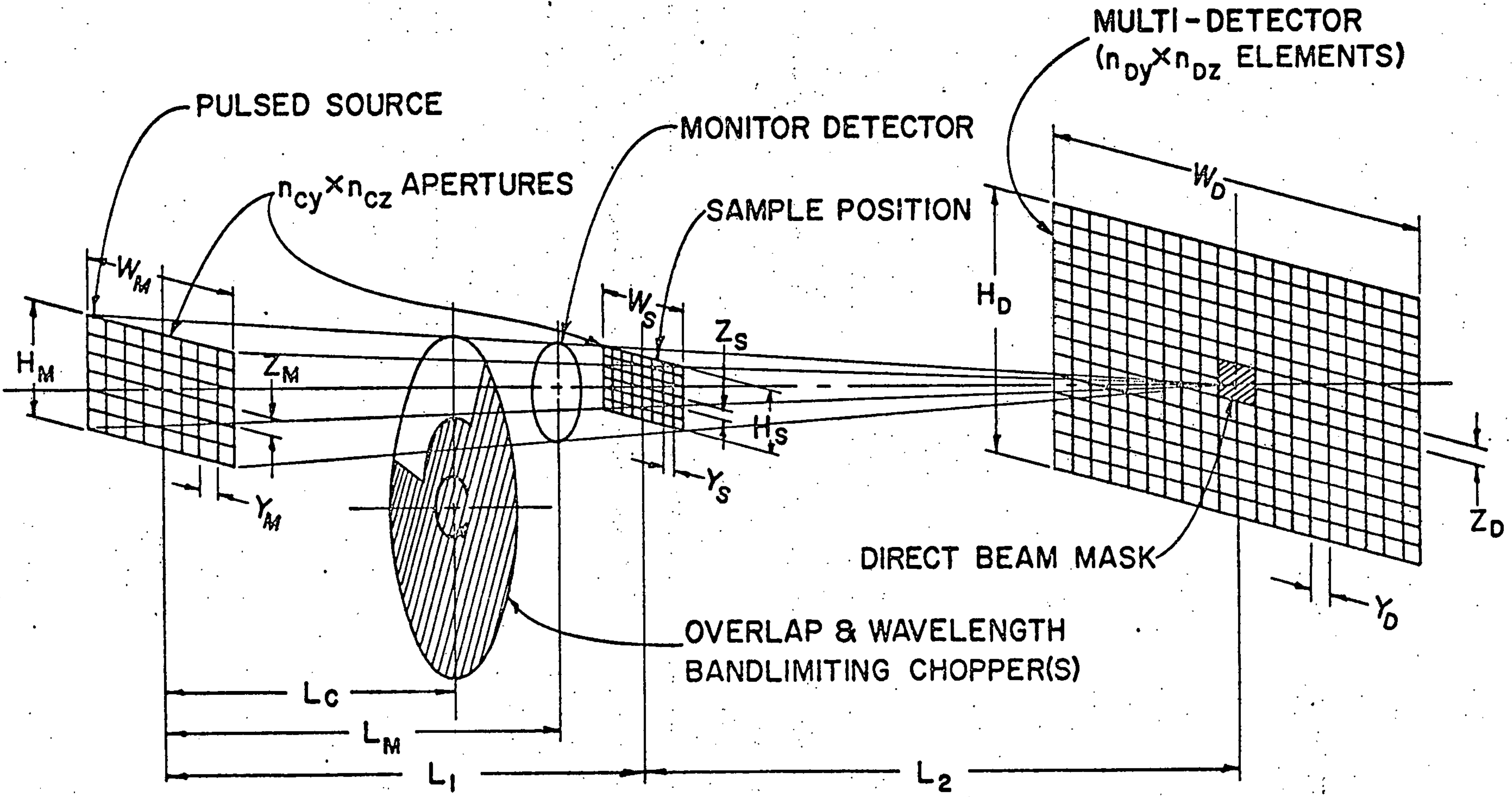

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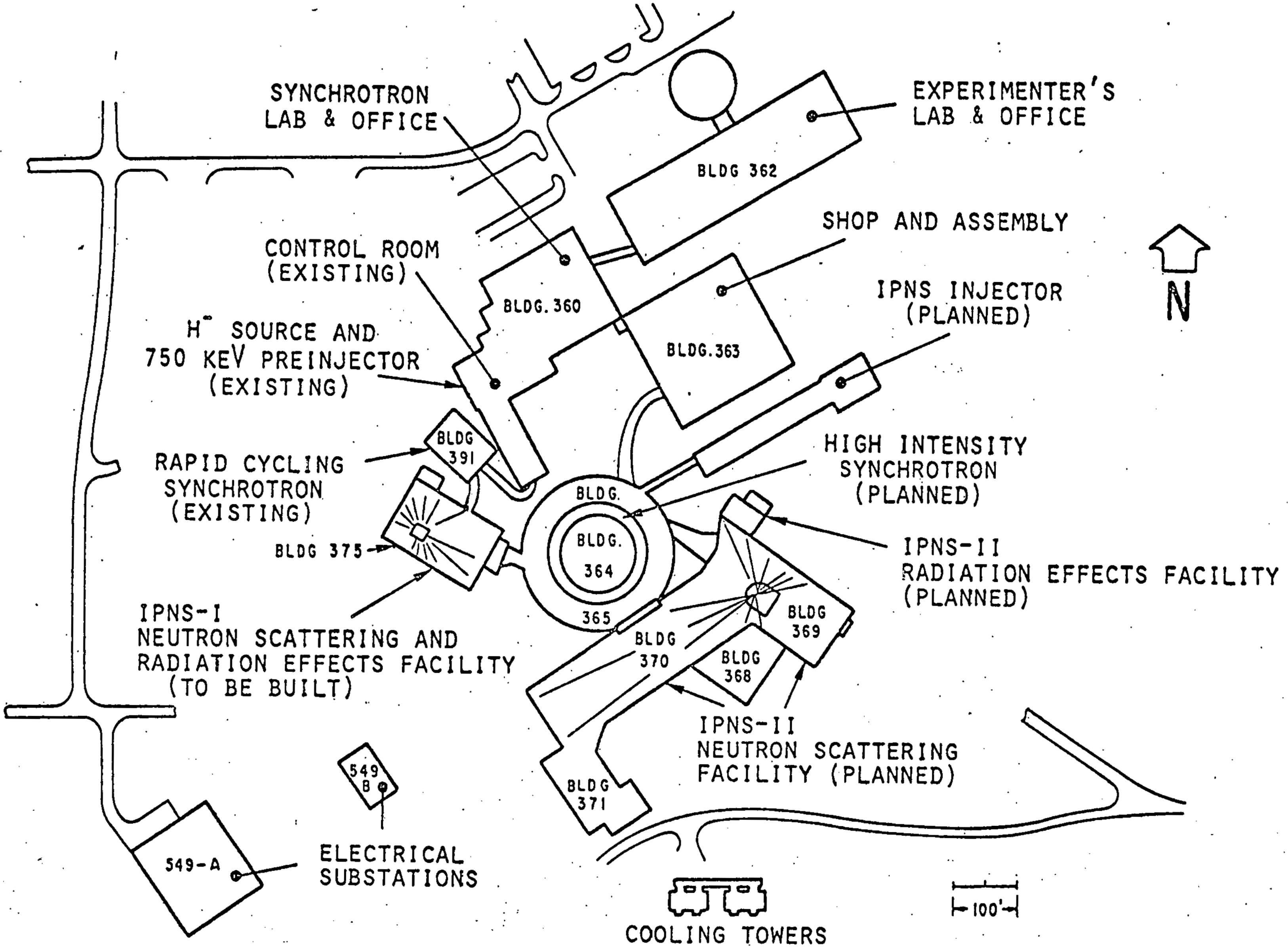

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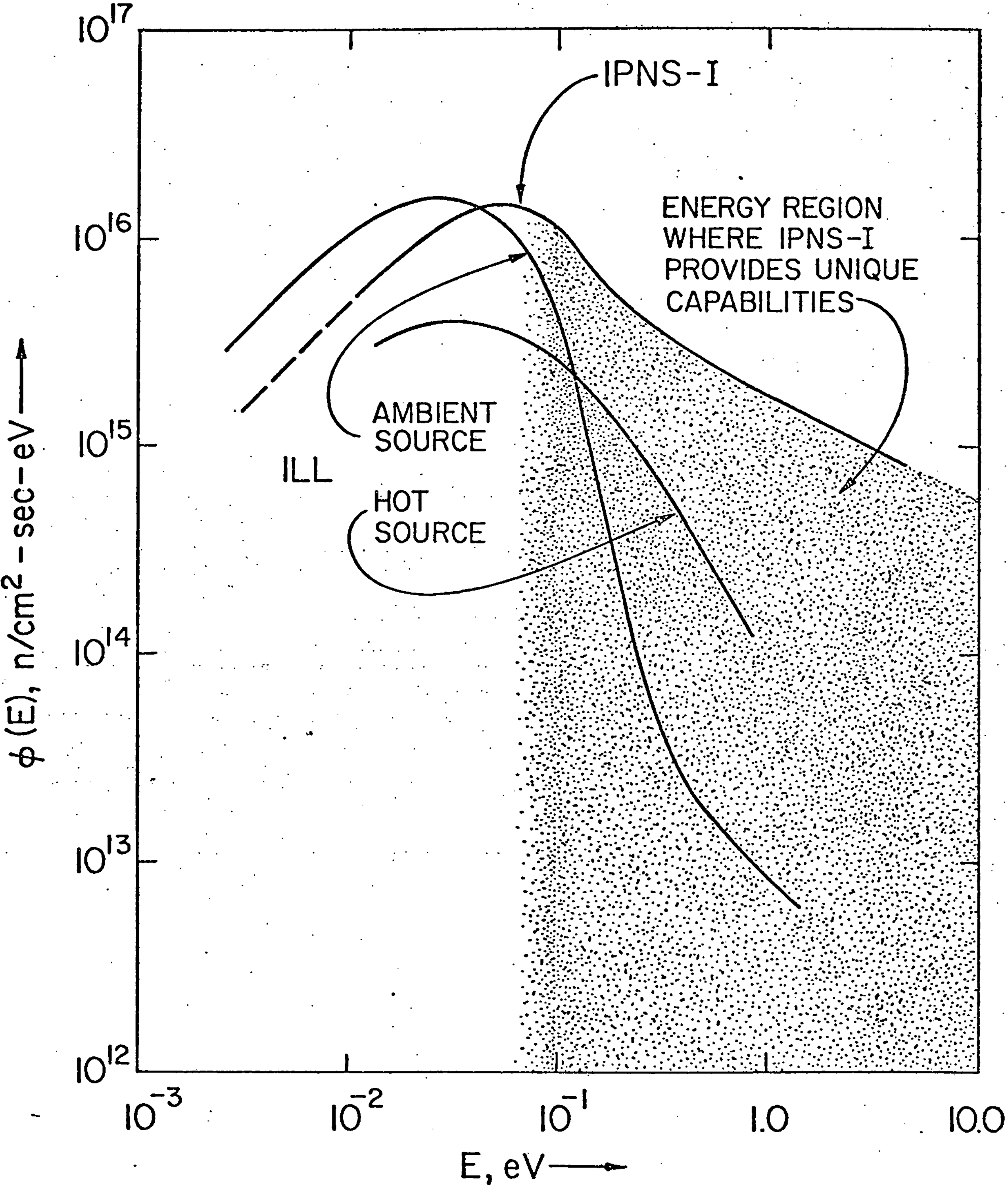

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