

ELECTRON VOLT SPECTROSCOPY USING RESONANCE ANALYSIS AT THE SNS

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ABSTRACT

A brief description is presented of the instrumentation which will be available in the early period of SNS operations for the development and scientific use of electron volt inelastic neutron spectroscopy.

I INTRODUCTION

The eVS project on the SNS has as its present aim the development and scientific use of a spectrometer in which the scattered neutron energy is determined using a nuclear resonance absorption filter. Two basic methods of using absorption resonances will be investigated in tandem on the one beam line. The resonance detector spectrometer (RDS) relies on the detection of promptly emitted  $\gamma$ -rays after resonant neutron capture. The resonance filter spectrometer (RFS), on the other hand, detects neutrons and utilises the difference in signal between the filter being out of and in the beam. This paper seeks to outline the present status of the eVS beam line instrumentation.

A principal feature of any eVS design must be the ability to measure high energy transfers with low associated momentum transfer  $\hbar Q$  (see Figure 1). Of central interest here are magnetic and electronic excitations which

require  $Q \leq 5 \text{ \AA}^{-1}$ . Because of the large electron-neutron mass difference, the "easiest" experiments will be those on tightly bound, high effective mass electrons. For high energy excitations, which we arbitrarily define as  $\hbar\omega > 0.5 \text{ eV}$ , this can be realised only by using high energy incident neutrons ( $E_0 > 5 \text{ eV}$ ) and detecting within a few degrees of the forward direction. Resolution requirements also place limitations on the science that can be done; the two "sharpest" nuclear resonance energies in this region are at 1.056 eV for  $^{240}\text{Pu}$  and at 6.671 eV for  $^{238}\text{U}$  and these can in principle provide  $\hbar\omega$  and  $Q$  resolutions of  $\sim 10\%$ . The intense epithermal spectrum of pulsed spallation sources also allows, for the first time neutron studies of condensed matter at high momentum transfer (see Figure 2). Values of  $Q \sim 150 \text{ \AA}^{-1}$  are attainable (e.g. using the 6.67 eV  $^{238}\text{U}$  resonance and detecting at  $\phi \sim 170^\circ$ ); such  $Q$  values approach the impulse approximation.

A full review of the current status of eVS instrumentation development has been presented in a previous report [1], together with a discussion of some areas of scientific application. For more detailed descriptions of the techniques and experimental results see references [2] (RDS) and [3] (RFS) and the references therein.

The two detection methods will use a common collimation package, beam dump, data acquisition electronics (DAE), front end minicomputer (FEM), etc. and both view the same ambient (A) moderator. Incident and scattered beam neutron monitors will be common to the two instruments. The sample/detector tanks are sited within a blockhouse of wax/borax shielding tanks, thereby allowing relatively convenient close access. In this way a full assessment of the two methods can be made during early SNS operation, with the aim of determining the best eV spectrometer design.

## II BEAMLINE LAYOUT

The eVS beamline has five major components beyond the moderator: the shutter, incident beam collimation, the Resonance Filter Spectrometer (RFS), the Resonance Detector Spectrometer (RDS) and the beam dump. The general layout of the beamline is shown in Figure 3. More detailed diagrammatic information on the beamline components external to the biological shield can be found in Figure 4. Nominal RFS and RDS sample positions are at 10 m and 11.5 m respectively with secondary flight paths of  $\sim 1 \text{ m}$  in both cases.

### III COLLIMATION

The incident beam collimation package defines a circular beam of diameter  $\sim 30$  mm at a sample position  $\sim 10$  m from the moderator. The sample views a 100 mm diameter circle inscribed on the 100 x 100 mm ambient (A) H<sub>2</sub>O moderator at 89.3° to the moderator surface.

To measure electron volt energy transfers in inverse geometry at low Q we are constrained to use relatively high energy neutrons and detect at scattering angles  $\phi \sim 2^\circ$ . Implied in this statement is the importance of a design that limits both the sample and the detectors' view of collimation surfaces directly illuminated by the moderator, and a choice of materials that minimise problems associated with inelastic collimator transmission and  $\gamma$ -ray fluxes. The ability to collimate a relatively high energy neutron beam without introducing significant off-energy contamination is of paramount importance to the successful development of an eVS.

The eVS collimation may be divided into 3 sections: that within the shutter, the insert (i.e. within the biological shield) and that external to the biological shield. Figure 5 gives a schematic view of the assembly.

The shutter collimation consists of nine octagonal irises separated by iron spacing sections and held within an iron shot/borax/resin moulding which serves to decouple the collimation from the surrounding bulk shielding. The irises are formed from eight 6 mm thick sintered boron carbide tiles set at 45° to one another. The inner surfaces of the iron sections are set back from the B<sub>4</sub>C - defined beam edge such that they cannot be viewed directly from the sample and should not, therefore, contribute to the instrument background as an extended secondary source.

Within both the insert and external collimator sections the beam is defined by a series of twenty 50 mm thick circular section B<sub>4</sub>C/(10%) resin irises. This low hydrogen content mix should help to reduce the albedo of the inner, illuminated surface of the iris and moderation of the incident beam energies. The problem is ameliorated further by the intrinsic roughness ( $\sim \frac{1}{2}$  mm) of the iris inner surface. (Recent development work [4], motivated by the longer term requirements of the eVS project, has led

to  $B_4C$ /resin formulations with resin contents as low as 3%). Upstream of eight of the irises there is a 50 mm thick lead ring which serves to collimate the  $\gamma$  ray flux generated within both the target assembly and the collimation itself.  $\gamma$  collimation is particularly important when using a RDS-type system but will also be of benefit to a RFS-type system using Li-glass scintillator detectors. The lead rings are set back from the beam edge and cannot be viewed directly from either sample or detector positions, thereby avoiding the secondary neutron emission problem inherent in the use of lead. It is also worth noting that relatively pure lead ( $\sim 99.8\%$ ) has been used in the fabrication to avoid possible background problems arising due to the large antimony content of most low grade lead stock: (antimony has a strong  $\gamma$ -emitting resonance at 6.24 eV). Downstream of each  $B_4C$ /resin iris there is a thin aluminium disc plasma spray coated with  $\sim 1$  mm  $B_4C$ . These discs penetrate the epithermal beam sufficient to prevent a direct view of any iris from the sample or detector positions. These ensure that no hydrogenous collimator surface material may be seen directly by either the sample or by any detector.

The gaps between iris assemblies are filled with a lead + iron shot, borax and resin moulding with an inner surface lining of  $B_4C$ /resin. The inner surfaces of these are set well back from the beam edge.

The insert and most of the external collimation is evacuated to  $\sim 1$  mbar, individual sections being separated by thin aluminium windows. The vacuum window immediately upstream of the spectrometers is sufficiently far from the RFS detector positions that it cannot be viewed at  $\phi > 5^\circ$ , and lower angle detectors will have only a limited view. This arrangement also allows relatively easy access to the final three collimator irises for modification, if necessary. The final vacuum window is at the end of the beam dump drift tube. A gap in the collimation just outside the biological shield facilitates the insertion of resonance absorption blocking filters and will also allow direct geometry measurements.

The above collimation gives an umbra diameter of  $\sim 28$  mm at the RFS sample position converging at 3.0 mrad, and, with a penumbra annulus of width  $\sim 4$  mm which diverges at 6.8 mrad. At full SNS intensity the neutron flux through this sample position will be  $\phi \sim 3 \cdot 10^7$  n  $eV^{-1}$   $s^{-1}$  at  $E = 1$  eV (note,  $\phi \approx E^{-0.95}$  for  $E > 1$  eV). Within the umbra the flux is  $\sim 5 \cdot 10^6$  n  $eV^{-1}$   $cm^{-2}$   $s^{-1}$ . Provision has been made for the insertion of up to 250 mm

of beam trimming collimation just upstream of the RDS; assuming a nominal 30 mm diameter beam/sample within the RDS, this implies a flux of  $4 \times 10^6$   $\text{eV}^{-1} \text{cm}^{-2} \text{s}^{-1}$ .

Two lithium glass scintillator monitors [5] may be installed using vacuum feedthroughs into the collimation (at either 6.5 m or 8.6 m from the target) and beam dump tube (at 13.5 m from the target). These low efficiency monitors have a fast response time, and their  $E^{-\frac{1}{2}}$  efficiency (at  $E > 1$  eV) makes them highly suitable for eV spectroscopy applications.

#### IV DETECTION SYSTEMS AND ELECTRONICS

As described above, there will be two detection systems under development within the eVS project, one downstream of the other. A prototype RFS sample/detector chamber has been installed on a beam line at the WNR facility, Los Alamos, for tests, but has now been returned to RAL to form the basis of a RFS on the SNS. The RDS equivalent will be based on the existing system installed on the Harwell Linac as part of a Harwell/Oxford University/RAL collaboration.

The two spectrometers will both use the same standard SNS instrument DAE package and VAX 11/730 FEM. The FEM will have access to the HUB computer via a Cambridge Ring and to all standard SNS instrument software packages.

Clearly, the two methods cannot be pursued simultaneously; during RFS runs the downstream RDS assembly will be left void (i.e. forming part of the beam dump drift tube), whilst for RDS runs it will be necessary to insert some collimation into the RFS sample/detector chamber. Having outlined the substantial areas of instrumentation overlap we now describe briefly the major features peculiar to each method.

- (a) RFS. The prototype design has been discussed in some detail elsewhere [6] and the salient features only will be summarised here. The incident flight path is  $L_1 \sim 10$  m, the scattered flight path will normally be  $L_2 \sim 1$  m though there exists an option to reduce this to  $L_2 \sim 0.75$  m. The 10 atm He-3 detector tubes are housed within a moulded  $\text{B}_4\text{C}$ /resin shielding block which allows the use of 1, 2 or 3 tubes (ganged or singly) at  $\phi \sim \pm 10^\circ$ ,

$\pm 15^\circ$  and  $\pm 20^\circ$  and a four-detector square around the beam at  $\phi \sim 5^\circ$ . It will be possible to extend the range to smaller scattering angles by installing an array of scintillator detector elements. Both the sample and the secondary beam fixed filters [3] are suspended from a sample tank flange. Rotary vacuum feedthroughs allow the user to perform a full double difference experiment without breaking vacuum. There is a liquid nitrogen feed through/cold finger assembly mounted on the sample flange which may be used to cool a filter or, exceptionally, the sample. The vacuum tank is lined internally with 6 - 8 mm of  $B_4C$ /resin (the resin concentration being limited to 3% in the sample tank and 5-10% elsewhere).

The inelastic scattering from  $ZrH_2$  was studied during a series of test measurements at the WNR pulsed neutron source using both  $^{149}Sm$  ( $E_R = 0.872$  eV) and  $^{181}Ta$  ( $E_R = 4.28$  eV) analysers. Figure 6 shows the derived double difference spectra at  $\phi = 5.5^\circ$  as well as computer simulations based on an Einstein oscillator model.

- (b) RDS. The RDS makes use of the detector and sample boxes previously used on the Harwell Linac. These are positioned to give an incident flight path of approximately 11.5 m and scattered flight path of 1 m, though variations are possible. An important component of the external shielding around the detector box will be a 100 mm layer of pure (antimony-free) lead to screen the detector from any  $\gamma$ -ray background. Initially the gamma ray detector will be a high resolution, high purity Ge detector (HPGe). The detector system has its own electronics, including an A.D.C., and can give output signals for events in pre-selected  $\gamma$ -ray energy bands as well as a digital signal which can be used to give 2-dimensional data analysis (i.e. T.O.F. and  $\gamma$ -ray energy) at some future date. Plans are well advanced to install a low resolution BGO detection system which will allow an investigation of coincidence techniques.

Initial development of the RDS on the Harwell Linac is described in [7]. This includes a comparison of HPGe and BGO detectors. A double difference spectrum obtained on the Harwell Linac for scattering from  $ZrH_2$  at  $15^\circ$  using Sm analyser foils is shown in Figure 7.

## V eVS DEVELOPMENT

It is impossible to define, a priori, the precise line of development that the eVS project will take but it is possible to indicate the general directions. Optimisation of the incident beam collimation together with both detector and general instrument shielding is important. The resonance filter technique must be pursued to lower scattering angles, which will necessitate the development of a low angle scintillator bank and a re-design of the scattered beam fixed filter assembly. The resonance detector must likewise be pushed to low angles using both high and low resolution  $\gamma$  detectors together with an investigation of coincidence or anti-coincidence techniques.

In parallel with the development of the instrumentation, and as a necessary part of it, the areas of science made accessible by the spectrometers will be investigated.

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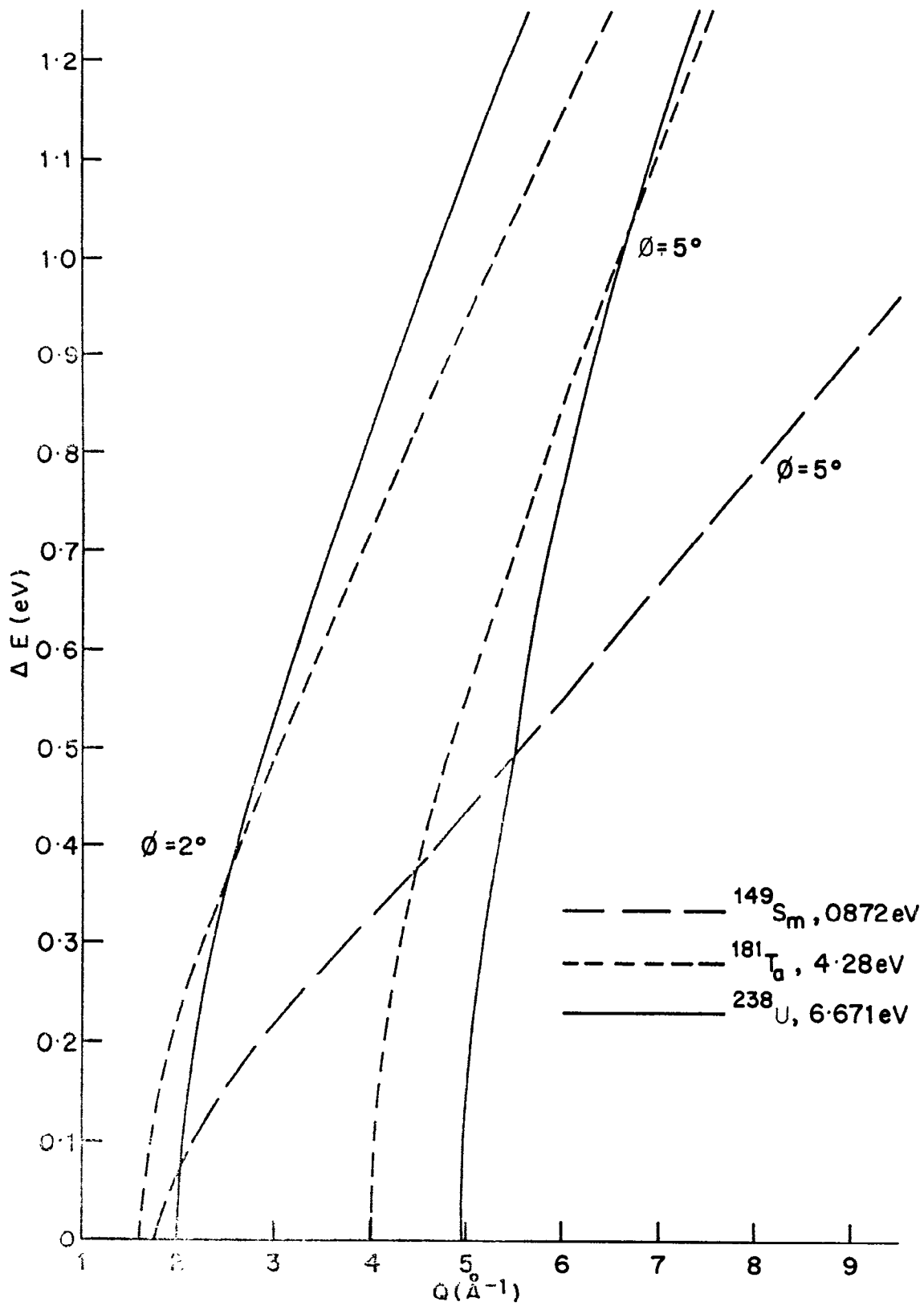


FIGURE 1 : DYNAMIC RANGE ACCESSIBLE WITH LOW ANGLE DETECTION

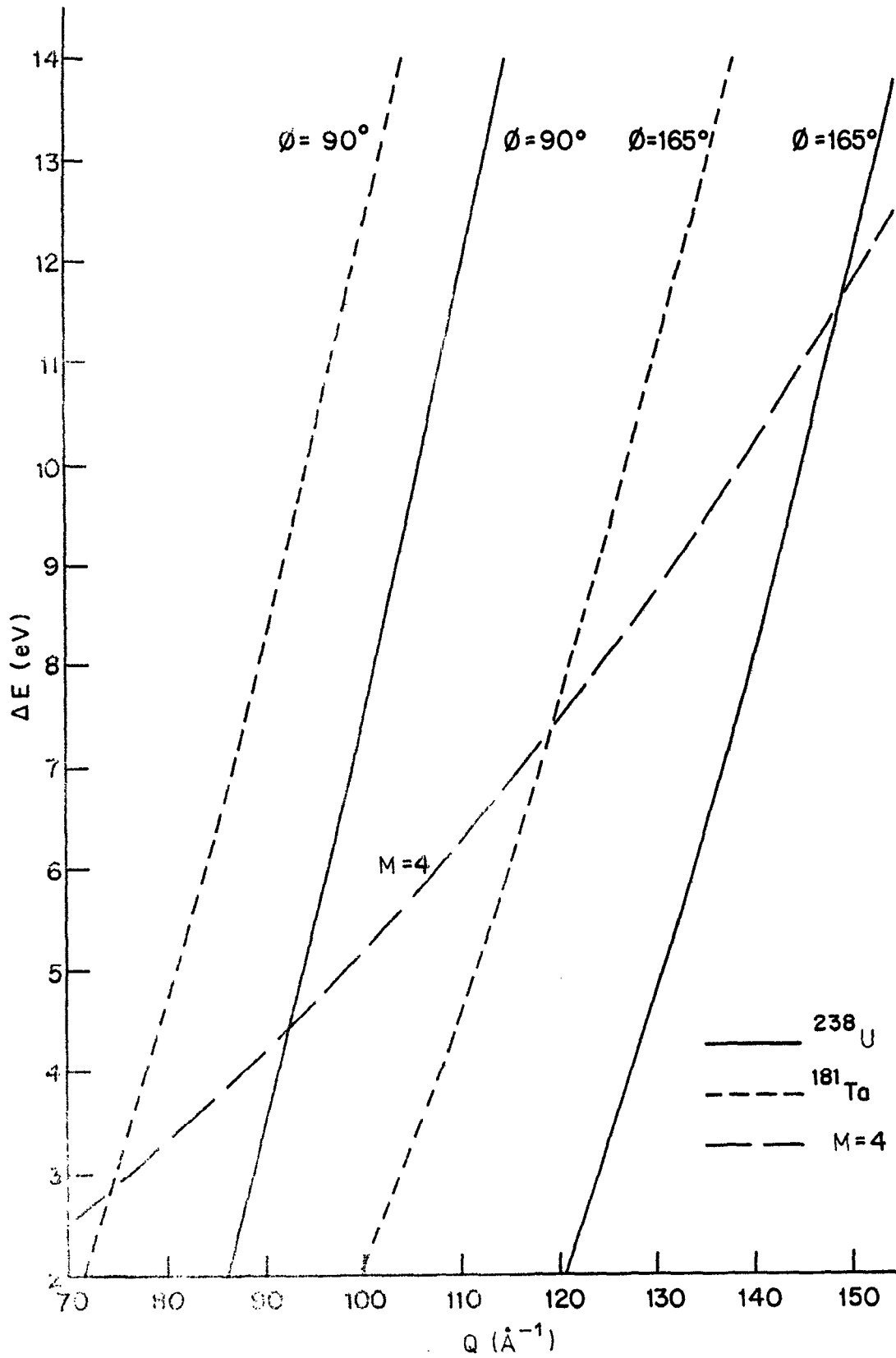
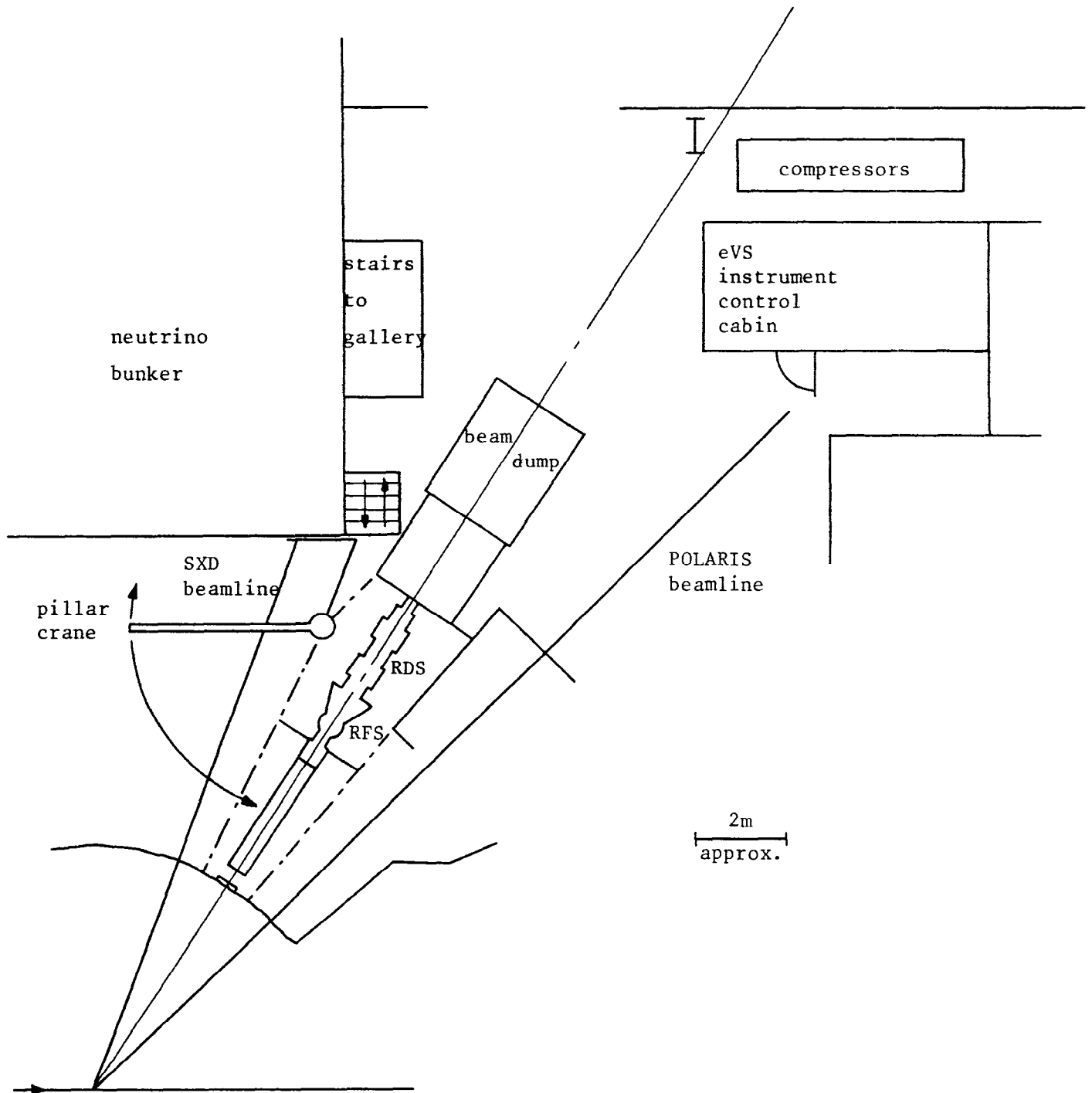


FIGURE 2 : DYNAMIC RANGE ACCESSIBLE WITH HIGH ANGLE DETECTION



**FIGURE 3** THE eVS BEAMLINE AND ITS ENVIRONS

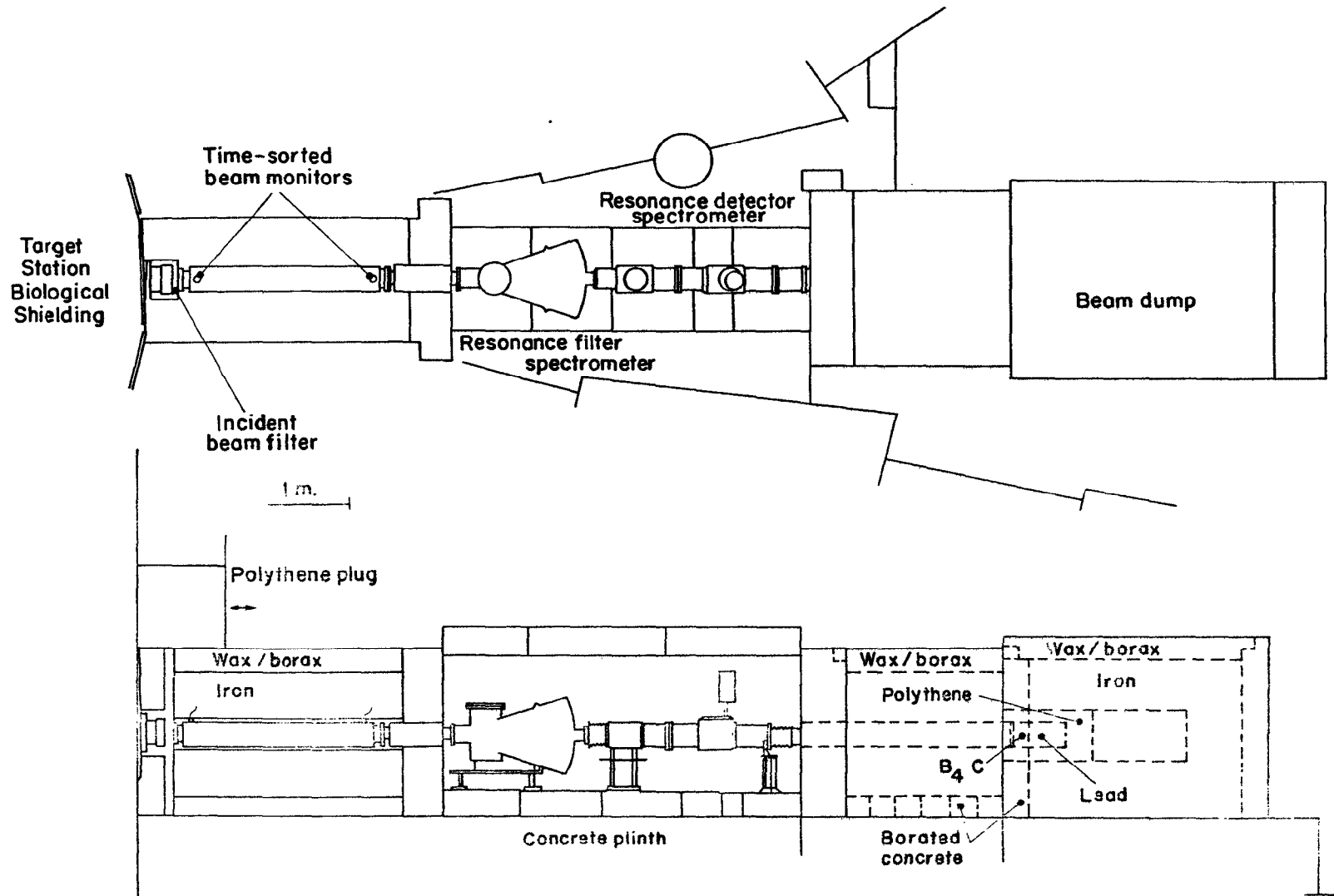


FIGURE 4 : Schematic Plan and Section of the eVS Beamline.

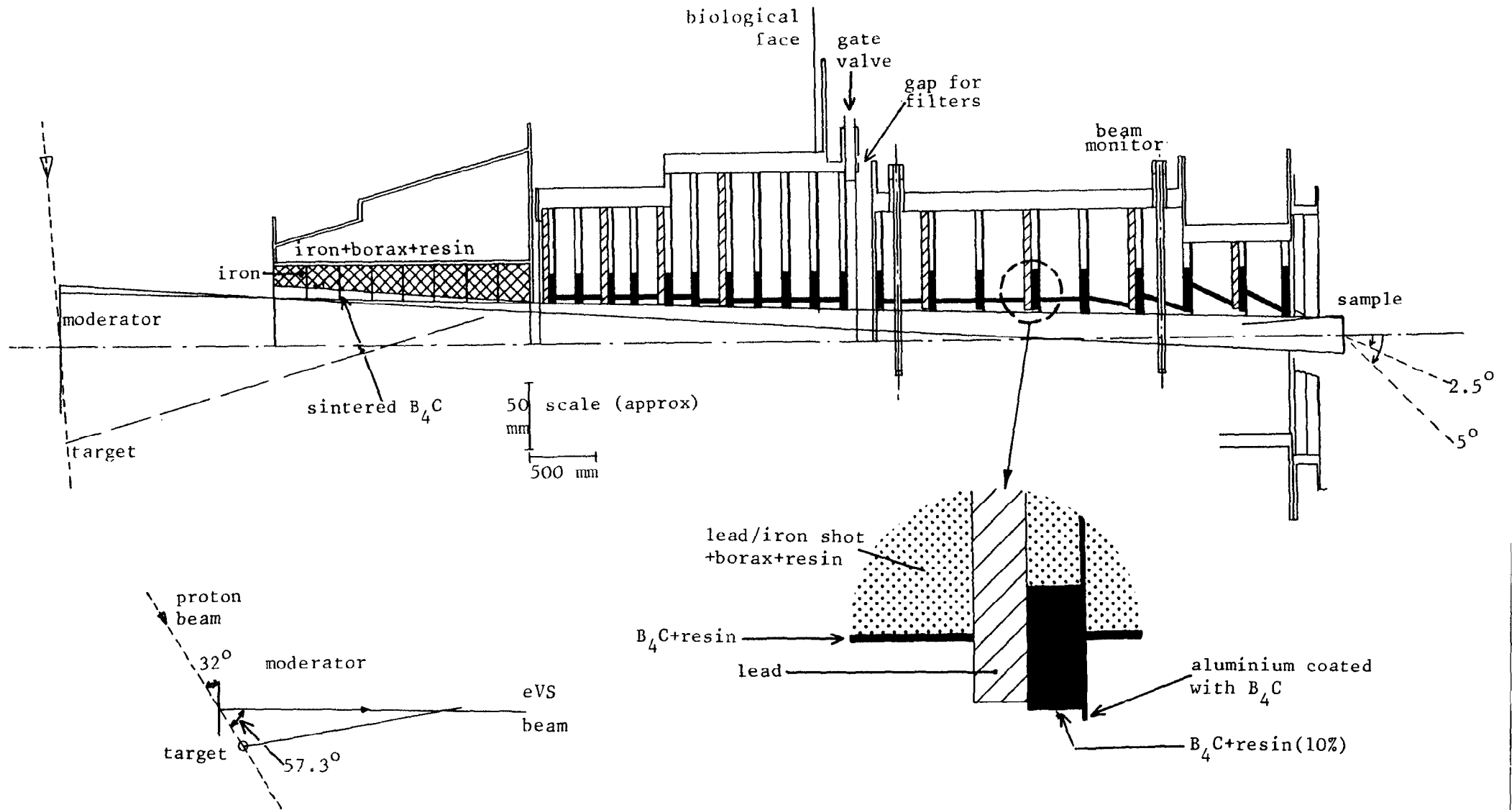


FIGURE 5 SCHEMATIC SECTION AND DETAILS OF THE eVS INCIDENT BEAM COLLIMATION ASSEMBLY

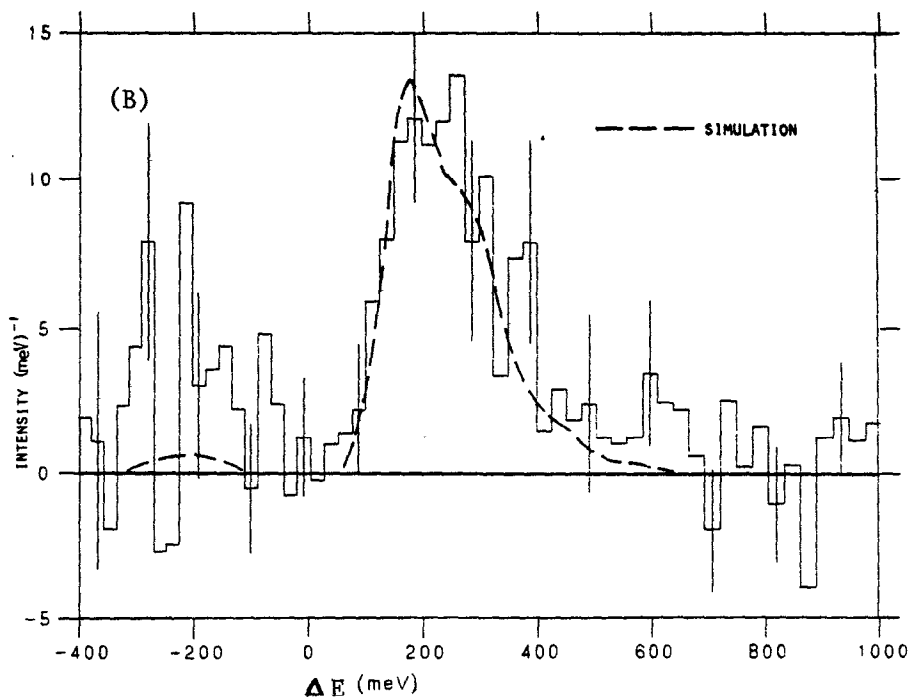
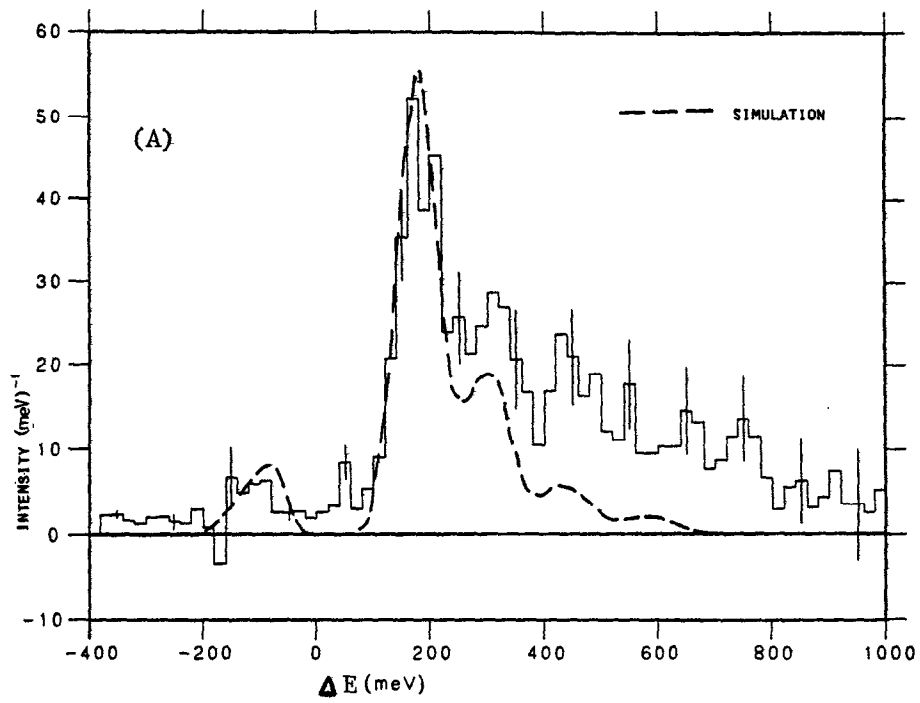


FIGURE 6 : INELASTIC SCATTERING FROM  $\text{ZrH}_2$  USING THE RFS:

(A) SAMARIUM ANALYSERS

(B) TANTALUM ANALYSERS

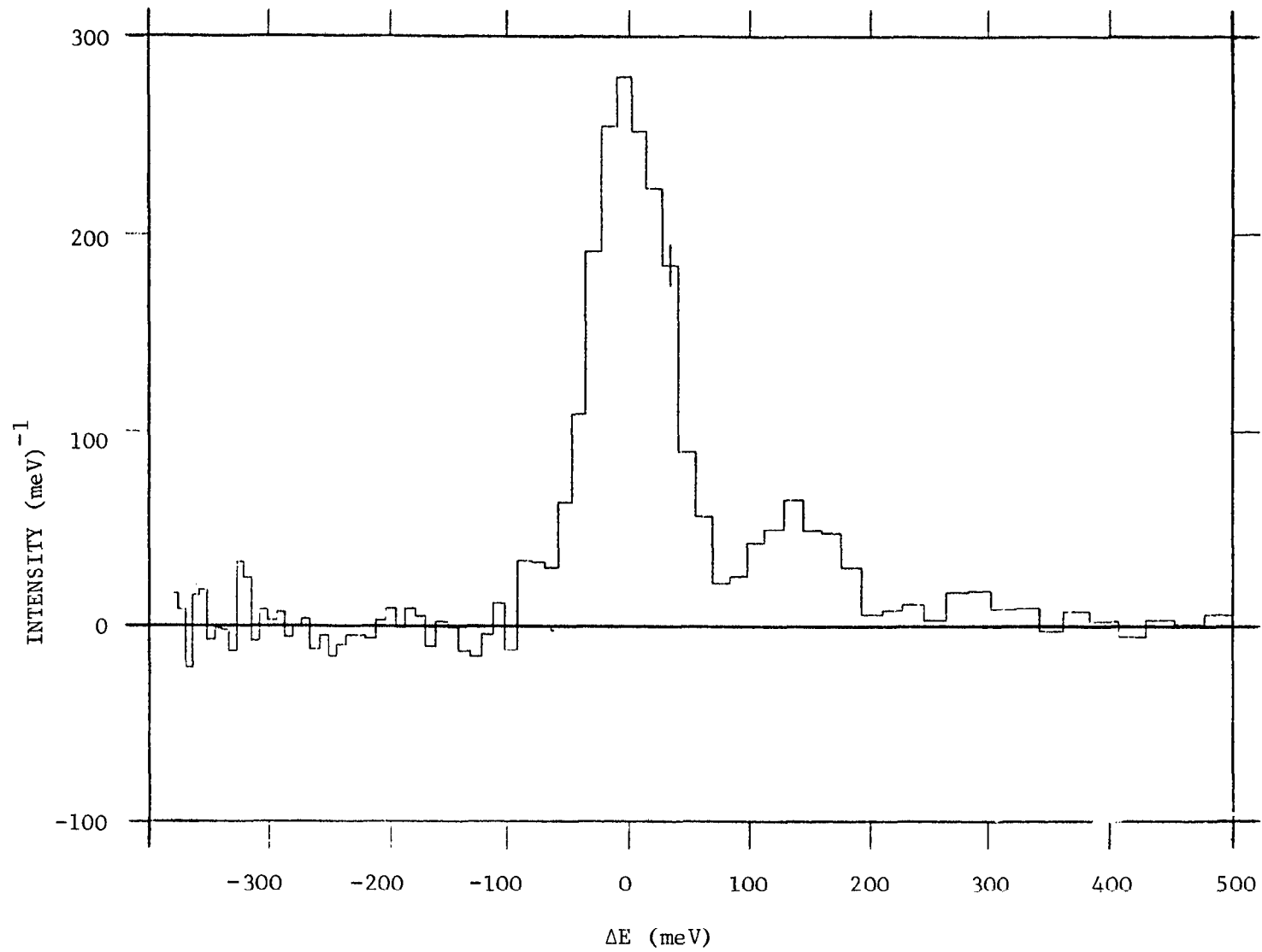


FIGURE 7 : INELASTIC SCATTERING FROM  $ZrH_2$  USING THE RDS:  
SAMARIUM ANALYSERS