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H E T : THE HIGH ENERGY INELASTIC SPECTROMETER AT ISIS

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INTRODUCTION

The HET spectrometer is a direct geometry chopper spectrometer designed specifically to study systems at high energy transfer ε with low associated momentum transfer Q and good energy transfer resolution $\Delta\varepsilon$. This is achieved by monochromating at high incident energies (100-2000 meV) and scattering at low angles (3^o-30^o)

Fields of interest include:

- High frequency localised magnetic excitations such as crystal field and spin-orbit transitions in 4f and 5f electron systems, where the electronic form factor severely reduces intensities when Q is greater than 4 $Å^{-1}$.
- Vibrational spectroscopy where the low momentum transfer minimises multiphonon effects and the excellent energy resolution, particularly at high energy transfer allows new features to be distinguished.
- Coherent excitations at high energy transfer in itinerant ferromagnets where again form factor considerations severely limit the available intensity.

In addition a high angle detector array allows measurements to be made at high energy transfer and high associated momentum transfer. Such measurements probe the momentum distribution of particles and is of particular interest for quantum systems such as hydrogenous materials and the helium liquids.

INSTRUMENT

The spectrometer is illustrated in Figure 1 and details are given in Table I. The monochromating chopper, located 10 m from the source, spins on magnetic bearings at frequencies up to 600 Hz and produces a sharp ($\sim$1μs) burst of neutrons on the sample. Three different slit packages each made of boron-fibre/aluminium composite are optimised to give peak transmission for 1000 meV, 500 meV and 250 meV neutrons. By rephasing the chopper opening time with respect to the accelerator pulse and by spinning at reduced multiples of the ISIS frequency, monochromatic beams of neutrons ranging from 100 meV to 2000 meV may easily be produced, see Figure 2. Such versatility has been invaluable in optimising the resolution and momentum transfer characteristics of the spectrometer for a given experiment.

The primary flight path and the entire spectrometer are under rough vacuum, and the sample chamber is turbo-pumped to a cryogenic vacuum. Almost all samples are run at low temperature. All internal surfaces of the spectrometer are lined with tiles of a low-hydrogen B_4C mix to minimise the scattering of high energy neutrons, and the same material is used as a shield behind each detector.

Detectors

Scattered neutrons are detected 4 m from the sample in a low angle array (3^o-7^o) of ^{3}He gas tubes, at 4 m in a movable high angle array (80^o-140^o) and at 2m in an intermediate angle array (10^o-30^o). Originally the low angle bank was complete, but only a test octant of 2mm glass scintillators were used at the 2 m position. It was known that the intrinsic background (0.5 n/cm^2/min) in the considerably less expensive scintillator detectors was a factor of 100 worse than achievable with gas detectors, but before operational experience it was not known whether this intrinsic background would be dominated by all other sources. One result of the quality of spectrometer shielding and the separation by time-of-flight of fast neutron background from signal neutrons was that the major background source for incident energies below 350 meV was the intrinsic noise of the detectors. The difference in intrinsic noise levels may be clearly seen in Figure 3. Signals clearly observed with the gas detectors were not seen in the scintillators. An early decision was taken to use gas detectors throughout the spectrometer. All detectors are now 10 atmosphere ^{3}He tubes 2.54 cm diameter. The majority have 30 cm active lengths, but some 20 cm and 15 cm active length detectors are used in constrained geometries at low angles.

For safety and reliability reasons, the large thin window at 2 m is being replaced in Spring 1987 by an eightfold array of locally thin windows, see Figure 4. This configuration will have 32 detectors mounted immediately behind each window at a distance of 2.5 m from the sample. A total of 256 detectors will be used. The inner four detectors of each group will have a 20 cm active length. The outer 28 will have a 30 cm active length. In the current (October 1986) configuration there are four arrays of 25 large detectors in the horizontal and vertical locations.

Choppers

The chopper control system, whereby the chopper is phased to a master 1MHz oscillator which controls extraction from the accelerator, has proved to be very effective in the majority of cases. An additional veto system has been introduced which rejects data on a frame by frame basis unless an optical monitor of the chopper indicates that the chopper is at the correct time delay with respect to the extracted proton burst. This has eliminated a 10% jitter which occurred under some accelerator operating conditions.

The background suppressing chopper, located upstream of the monochromating chopper, has been recently commissioned. This chopper blocks the beam during the power pulse (see Figure * in reference [1]) with 30 cm of nimonic alloy and prevents fast neutrons from entering the spectrometer. A substantial reduction in background was obtained, allowing experiments to be performed with incident energies up to 2000 meV.

EXPERIMENTAL PROGRAMME

Proposals from the UK neutron scattering community for time on HET have been considered in four categories: Vibrational Spectroscopy; Isotropic Magnetic Scattering; Momentum Distributions and Single Crystal Excitations. Preliminary experiments have now been performed in all four categories with considerable success.

Vibrational Spectroscopy

The versatility resulting from the ease of energy selection proved to be invaluable in optimising many experiments. This is well illustrated in a study of structure in the harmonics of the librational modes in NH_4Cl by Goyal et al [2]. The harmonics occurring around 95 meV and 140 meV are shown in Figure 5(a) where the incident energy is 160 meV. Reducing the incident energy to 120 meV (Figure 5(b)) significantly improves the resolution of structure in the first harmonic, but of course does not allow the second harmonic to be observed. The resolution characteristics of the spectrometer are summarised in Figure 6 as a function of the fractional energy transfer ε/E. In this case the choice of momentum transfer Q is not critical.

Isotropic Magnetic Scattering

In magnetic systems the choice of incident energy is more subtle, since the incident energy controls both the momentum transfer (and hence through the form factor, the intensity) and the resolution. The experiments on crystal field transitions in UO_2 by Osborn et al [3] used 360 meV neutrons to show the magnetic nature of the signal by studying the form factor variation, then sacrificed intensity for resolution and Q range by using 290 meV incident energy neutrons, see Figure 7, to resolve the $\Gamma_5 \text{-->} \Gamma_4$ triplet (7 meV), and finally resolved the $\Gamma_5 \text{-->} \Gamma_3$ doublet (3 meV) on the limit of the form factor intensity with 230 meV neutrons.

The strong epithermal component in the ISIS spectrum was exploited in several experiments. The J-->J+1 spin-orbit transition at 129 meV in Sm^{3+} ions in metallic $SmPd_3$ was observed for the first time [4] and its form factor dependence measured (see Figure 8). An incident energy of 610 meV was used exploiting a minimum in the absorption cross-section of natural samarium. Previous neutron studies of samarium compounds have only been possible on isotopically enriched systems.

Momentum Distributions

Using the high angle bank and neutrons with incident energies in the range 800 meV to 2000 meV, preliminary studies were made of high momentum transfer scattering from polycrystalline lithium [5], again without the need for isotopic enrichment, and liquid helium, both in the superfluid and normal phase.

Excellent data [6] were obtained on normal helium at high resolution and free from background from the cryostat. These data are shown in the Figure 9. The mean kinetic energy determined from a fit of the impulse approximation lineshape is significantly lower than previous measurements and is closer to the value from Green Function Monte Carlo calculations. Deviations from the impulse lineshape due to final state effects are being investigated. The error analysis is still in progress, but conservative estimates give <KE> = 13.2 +/- 0.5 K.

Single crystal Excitations

Preliminary test measurements were carried out on a single crystal of Pd_3Fe by Mitchell et al [7] to study methods of crystal alignment for future coherent excitations experiments. The crystal was easily aligned on a room temperature goniometer using white beam techniques (chopper removed) and then checked with monochromatic neutrons. Inelastic scans gave encouraging results and several points on the magnon dispersion curve previously measured by a triple-axis spectrometer [8] were observed. For example, Figure 10 shows a magnon at 66 meV along the 111 direction . Unlike their isotropic counterparts, such experiments use only a few in-plane detectors and hence require significantly greater neutron intensity.

CONCLUSIONS

In addition to commissioning the spectrometer, an excellent start to the scientific programme has been achieved. In the field of vibrational spectroscopy, measurements have been made on the metal hydride systems $ZrNiH_x$ and VH_2, on molecular species such as KH_2PO_4 and solid and liquid formic acid, in addition to the NH_4Cl data described above. The suitability of the spectrometer to study high energy magnetic excitations has been well established by the work on $SmPd_3$ and UO_2. Work on momentum distributions and coherent excitations has only just begun but the low intensity tests are very encouraging.

ACKNOWLEDGEMENTS

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REFERENCES

[1] T J L Jones et al These Proceedings
[2] P S Goyal, ISIS Internal Experimental Report, HET/IER/P7/86
[3] R Osborn, ISIS Internal Experimental Report, HET/IER/P6/86
[4] S Culverhouse, ISIS Internal Experimental Report, HET/IER/P8/86
[5] M Hewitt, ISIS Internal Experimental Report, HET/IER/P20/86
[6] P E Sokol, ISIS Internal Experimental Report, HET/IER/P17/86
[7] P W Mitchell, ISIS Internal Experimental Report, HET/IER/P21/86
[8] A J Smith, W G Stirling and T M Holden, J Phys F7, 2411 (1977)
[9] E Balcar and S W Lovsey, J Phys C19, 4605 (1986)

TABLE I

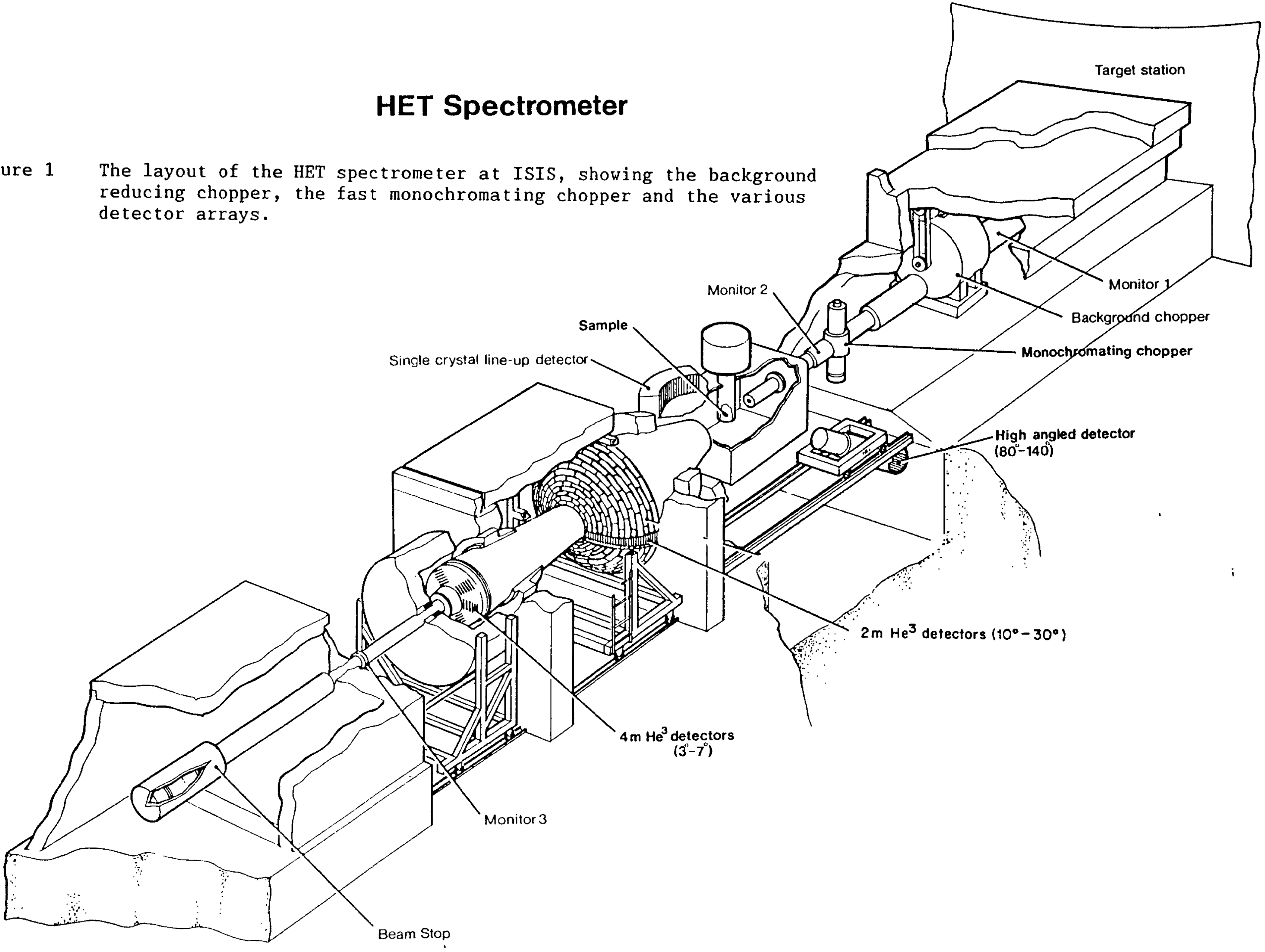
Specification

Incident energy	100-2000 meV
Energy transfer	20-500 meV
Momentum transfer	Depends on incident energy, energy transfer, and angle of scatter, eg 2.5 $Å^{-1}$ at 200 meV transfer; 40 $Å^{-1}$ (maximum) using high angle detector
Resolution	$\Delta E_0/E_0 = 0.7\%$

Instrument

Beam	N9
Moderator	316K H_2O poisoned at 1.5 cm
Background Chopper	8 m from moderator, closed at t = 0.
Chopper	10.0m from moderator, 600 Hz phased to ISIS pulse ± 1/2 μs
Sample position	11.8m from moderator
Beam size	50 x 50 mm^2 max
Detectors	3^o-7^o 4m counter bank: 50 10 atm ^{3}He 10^o-30^o 2m counter bank: 100 10 atm ^{3}He 80^o-140^o 4m movable bank: 10 10 atm ^{3}He
Intensity at sample	4.4×10^3 n cm^{-2} s^{-1} at $E_o = 1$ eV (full ISIS intensity)
Sample environment	Accepts standard sample environment equipment
Data acquisition	VAX11/730

Figure 1 The layout of the HET spectrometer at ISIS, showing the background reducing chopper, the fast monochromating chopper and the various detector arrays.



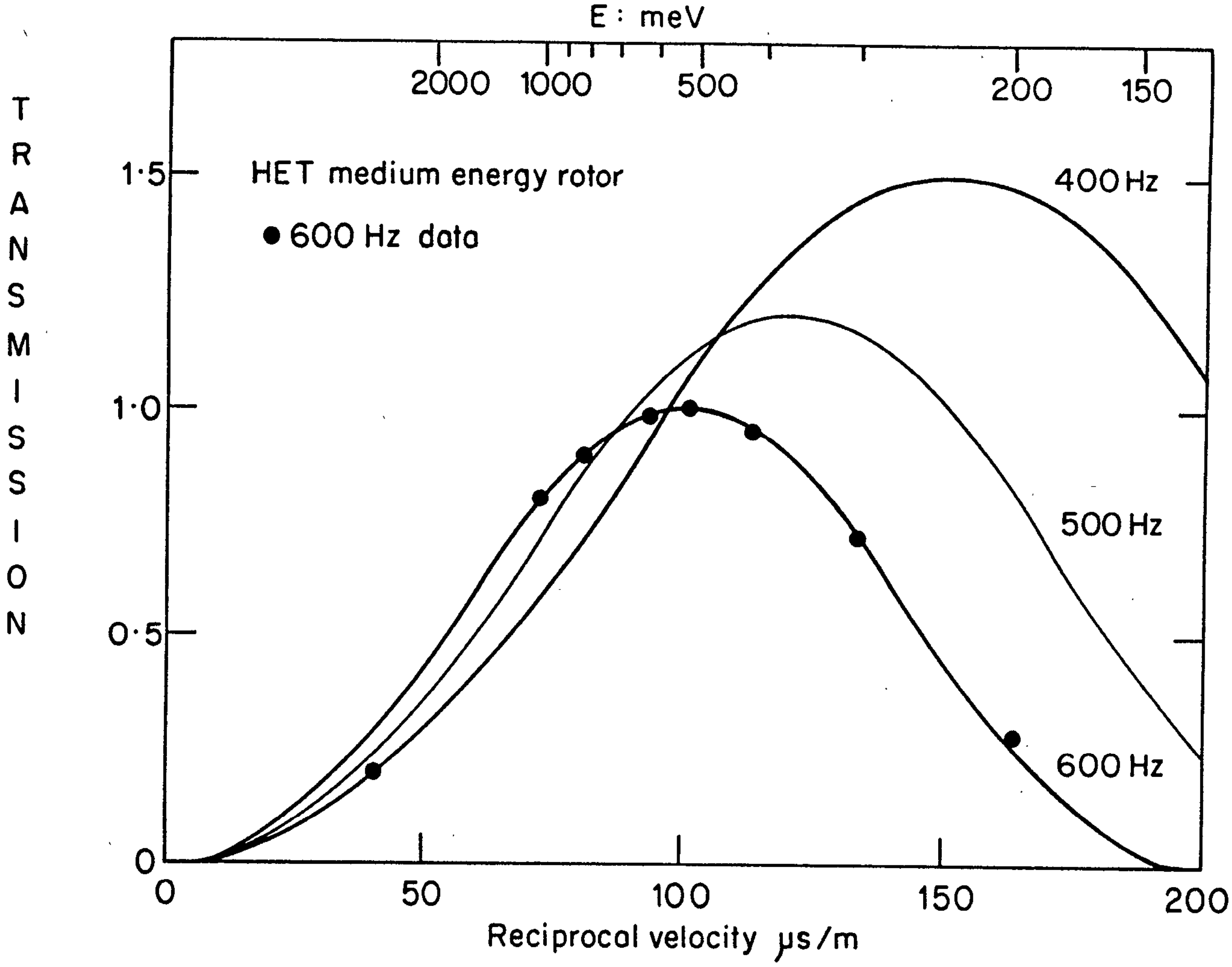


Figure 2 The calculated transmission characteristics of the medium energy HET chopper. It has an effective slit width of 0.89 mm and a radius of curvature of 1.32 m, which give it a peak transmission of 515 meV when spinning at 600 Hz. The data points are an experimental determination of the 600 Hz performance. By reducing the frequency to a lower multiple of the ISIS frequency, the peak transmission may be shifted to lower energies. (Eg: 10*ISIS = 500 Hz is optimised for 360 meV and 8*ISIS = 400 Hz gives 228 meV neutrons.)

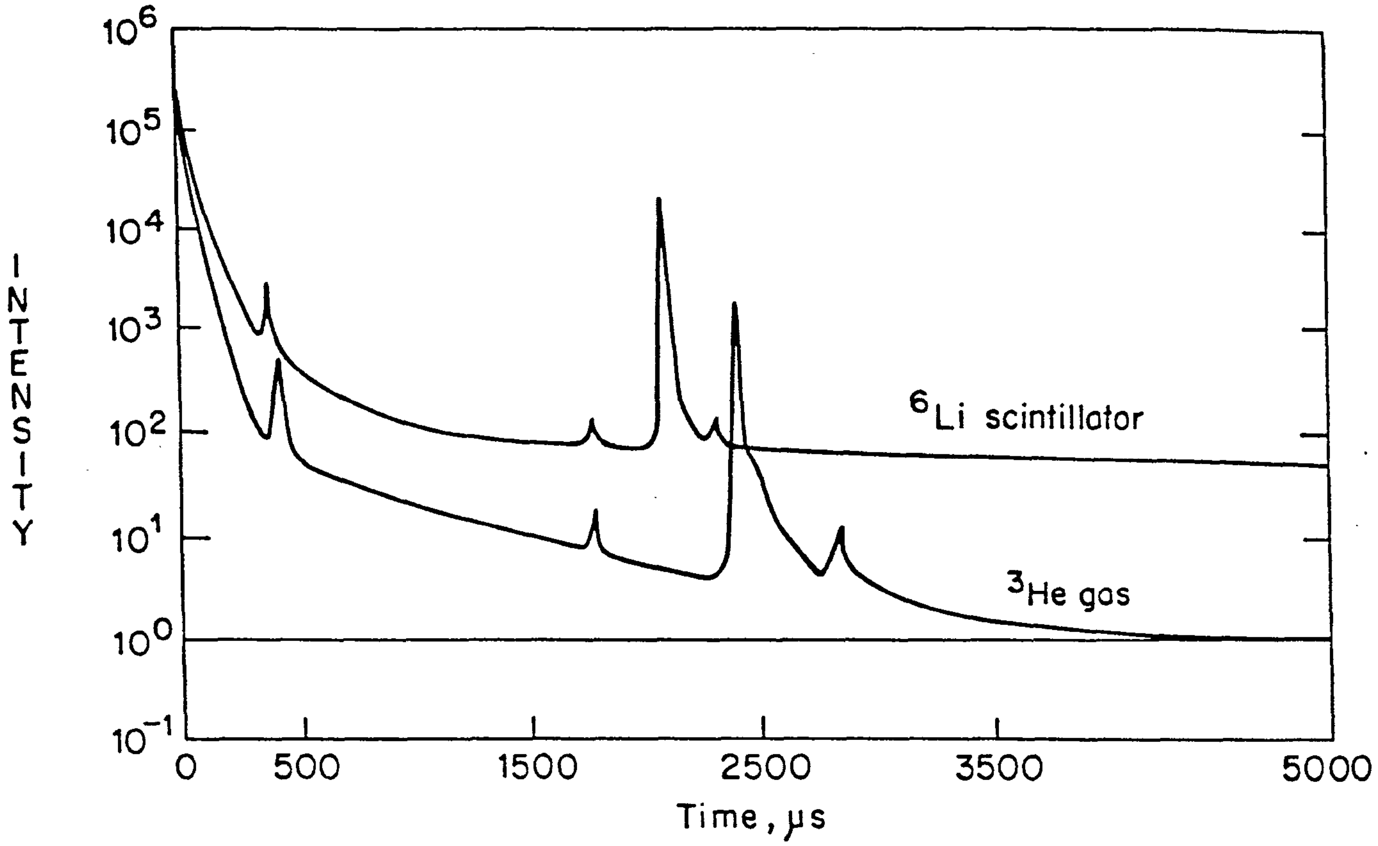


Figure 3 A comparison of the time-of-flight response of the ^{3}He gas detectors and the ^{6}Li glass scintillators. The difference in intrinsic background which dominates at long times is obvious. These data have been corrected for differences in solid angle.

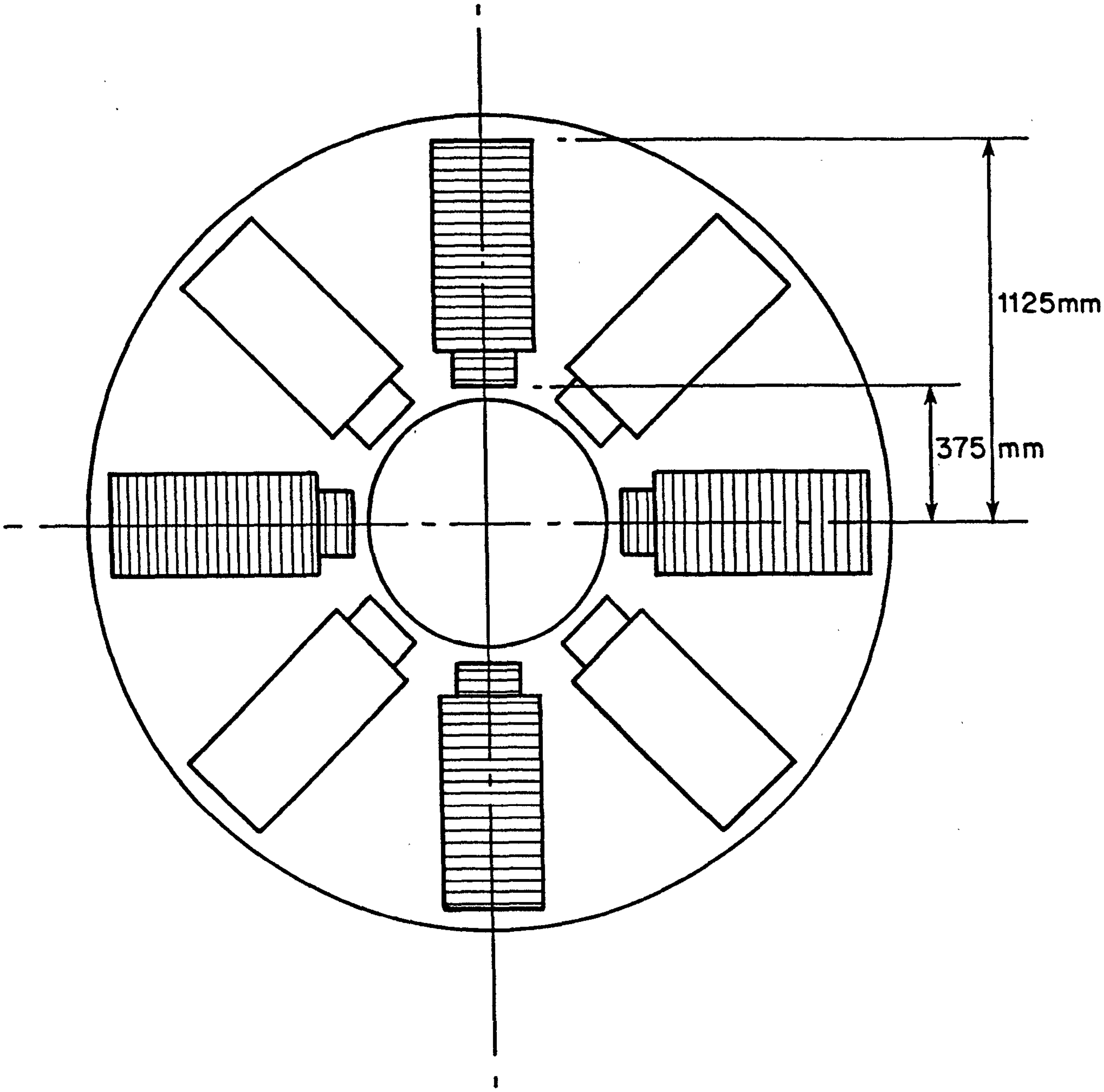


Figure 4 The eight-fold array of thin window and detectors which will provide continuous angular coverage at 2.5 m between 10° and 30°.

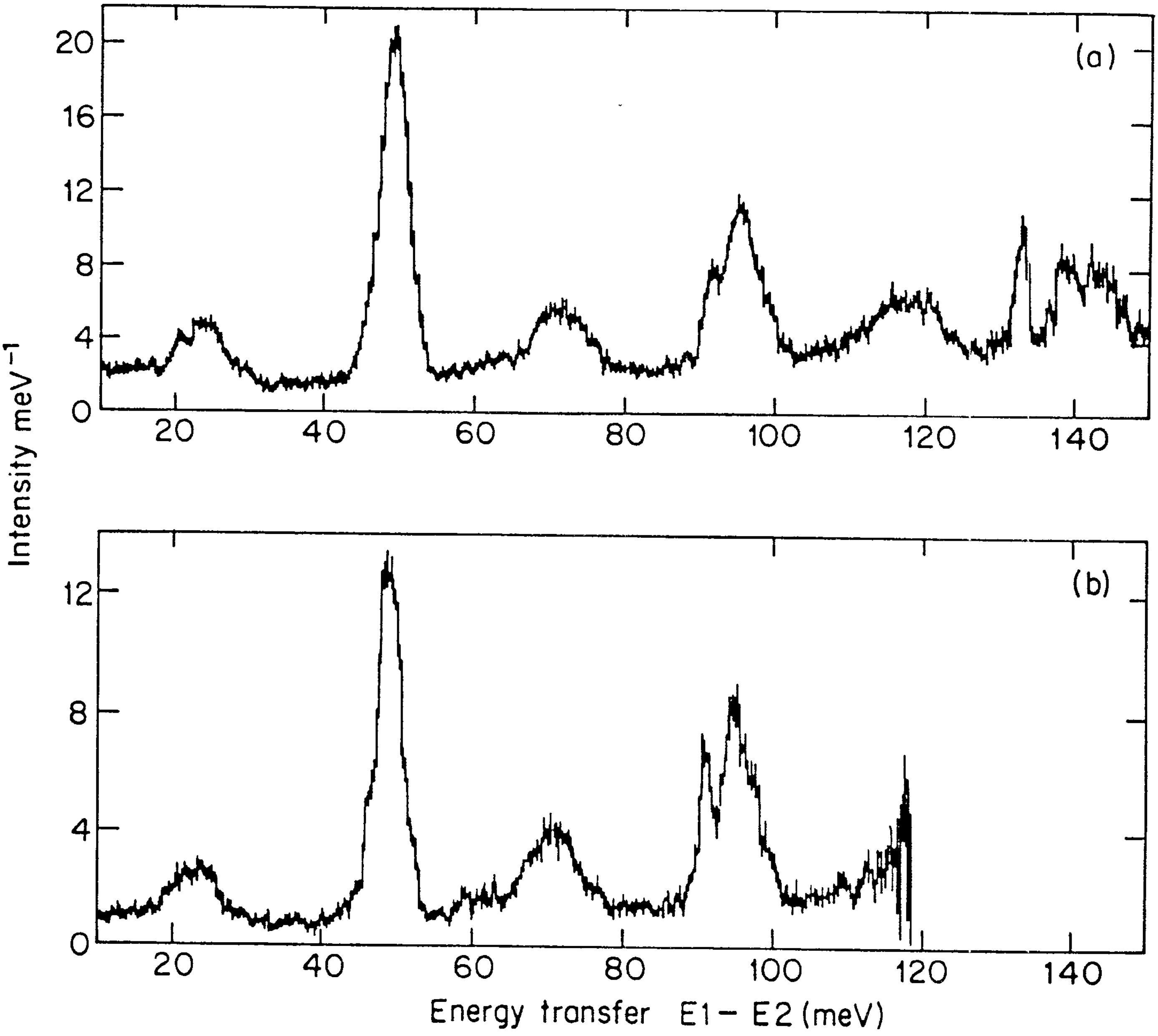


Figure 5 These data show inelastic scattering from the librational modes of NH_4^+ in NH_4Cl at two different values of incident energy. The data from the 4 m low angle array required 140 µA-hr of protons with an incident neutron energy of 160 meV, figure 5(a) and 50 µA-hr at an incident neutron energy of 120 meV, figure 5(b). The fractional energy transfer at 98 meV is 0.61 and 0.82 respectively. The improvement in resolution is in agreement with the calculation of Figure 6.

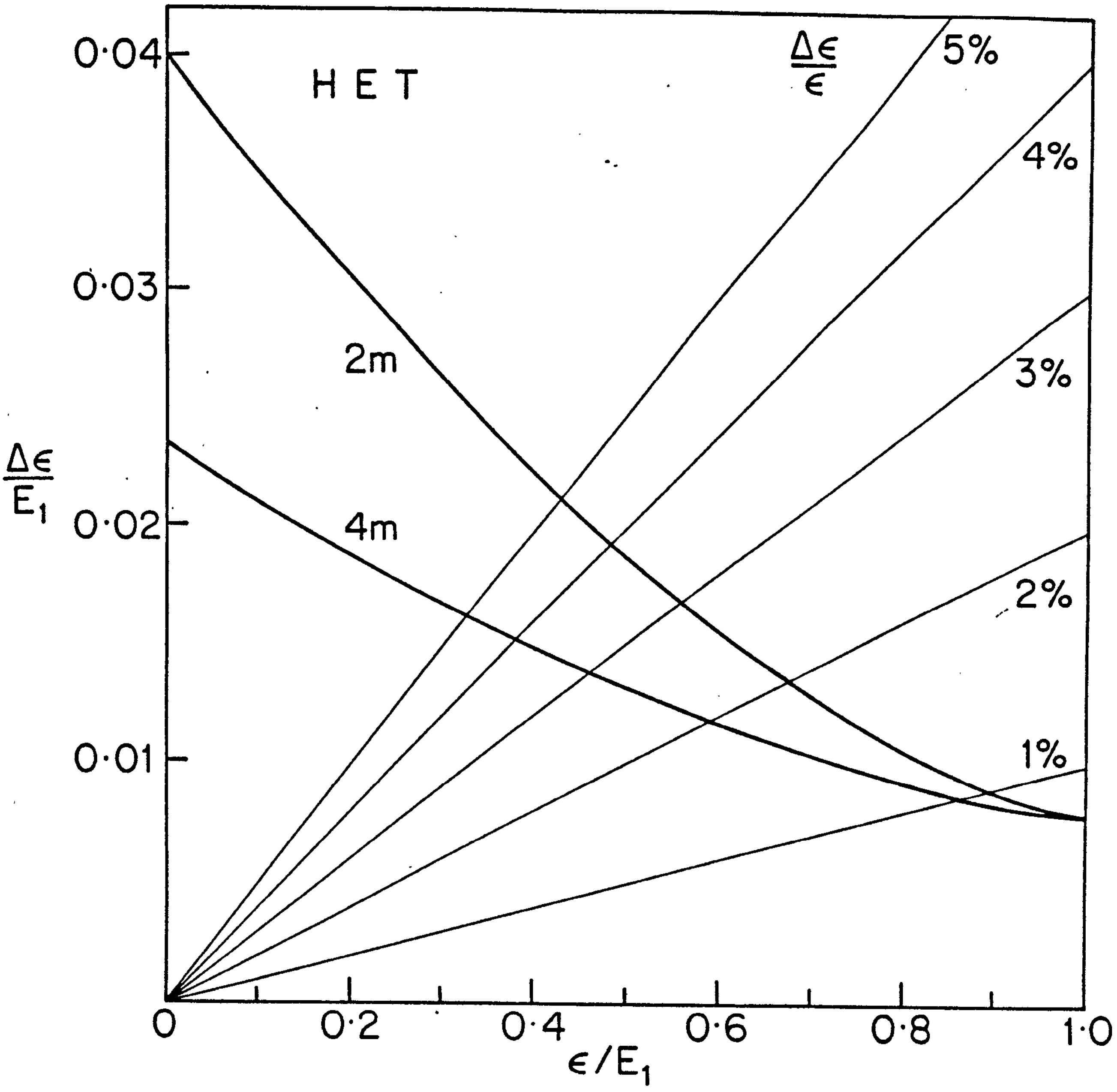


Figure 6 The calculated resolution of the 2 m and 4 m detector arrays on HET as a function of the fractional energy transfer ε/E.

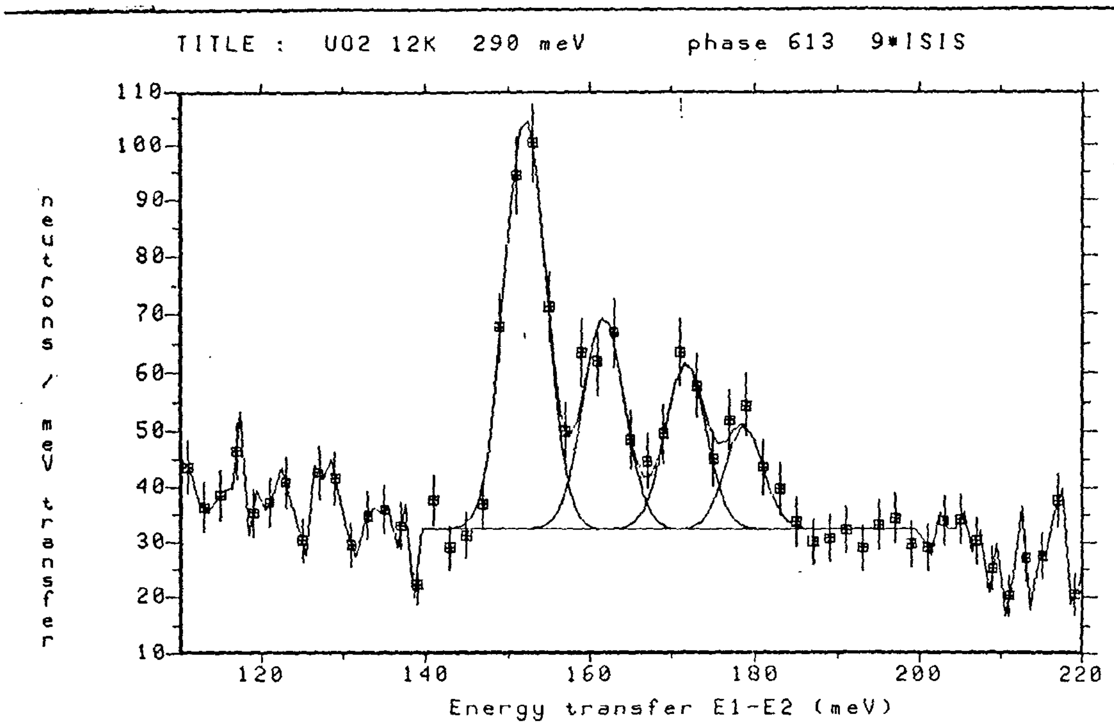


Figure 7a The spectrum of crystal field excitations in UO_2 at 12K measured in the 4 m low angle array ($\phi = 5^{o}$) at an incident energy of 290 meV. These data were recorded in 512 μA-hr using 80 g of sample. Previous measurements on HET at 360 meV had established the form factor dependence of this signal, but with insufficient resolution to identify details of the transitions involved. A subsequent measurement at 220 meV was able further to resolve the signal, but only at the limits of signal to noise.

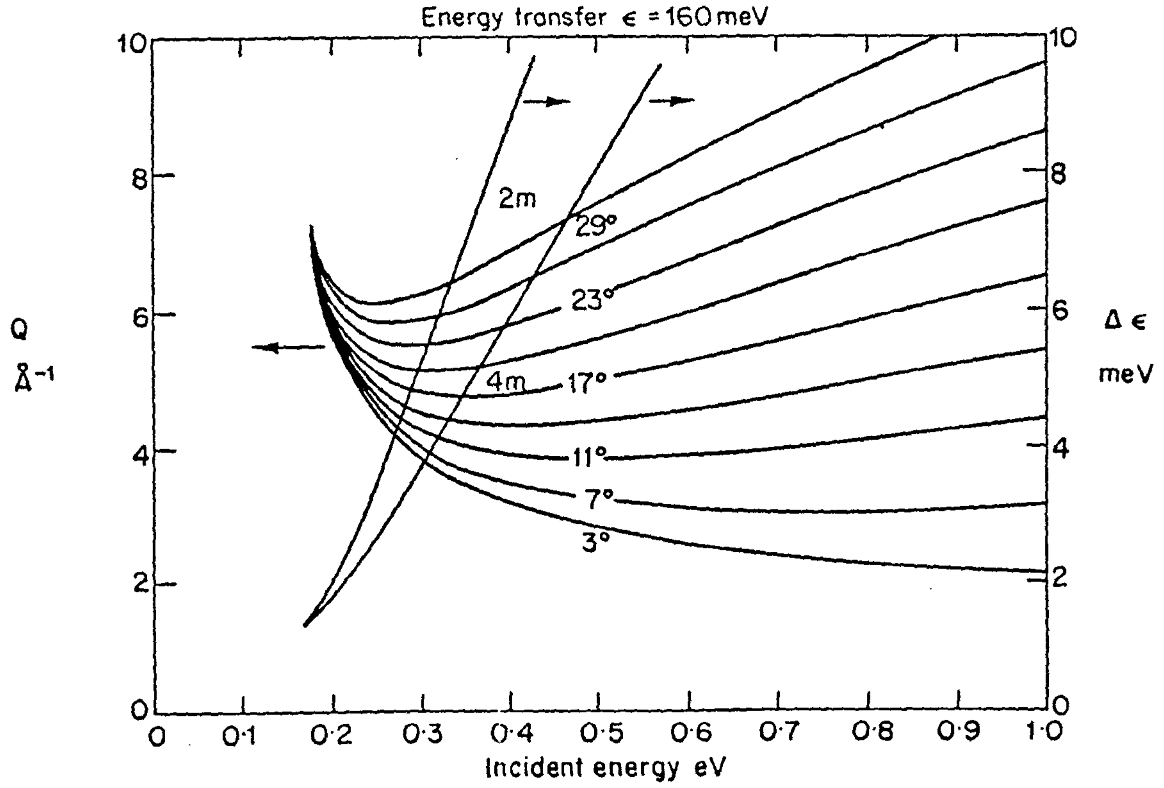


Figure 7b The resolution and momentum transfer characteristics of HET calculated for the 160 meV UO_2 excitations as a function of incident energy. These data allow the experimentalist to optimise the choice of incident energy by balancing resolution against Q (and hence, because of the form factor associated with magnetic scattering, intensity).

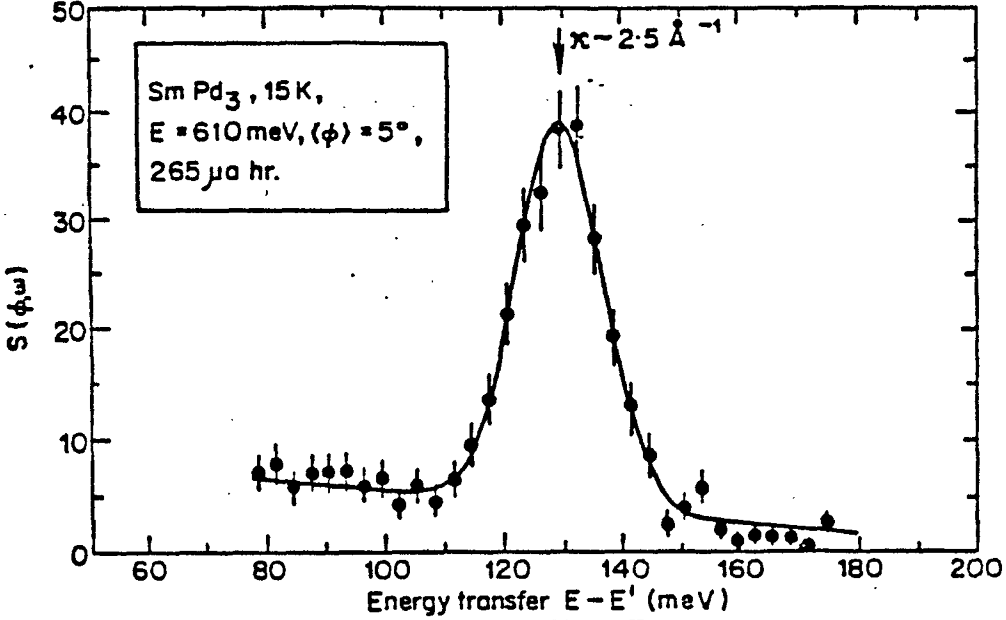


Figure 8a The spin-orbit transition in Sm^{3+} measured in 265 μA-hr at an incident energy of 611 meV. These data correspond to scattering at angles between 3.4° and 7°.

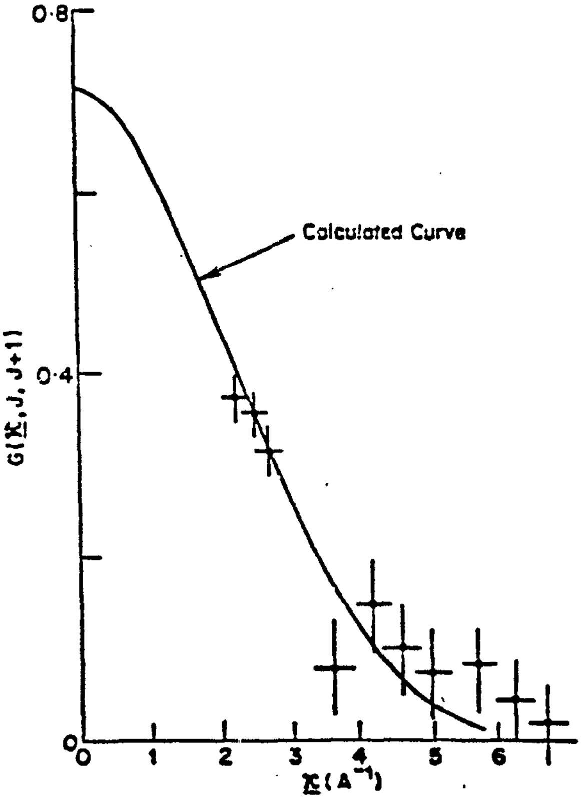


Figure 8b The observed form factor of the above spin-orbit transition in Sm^{3+} compared with the calculations of Balcar and Lovesey [9].

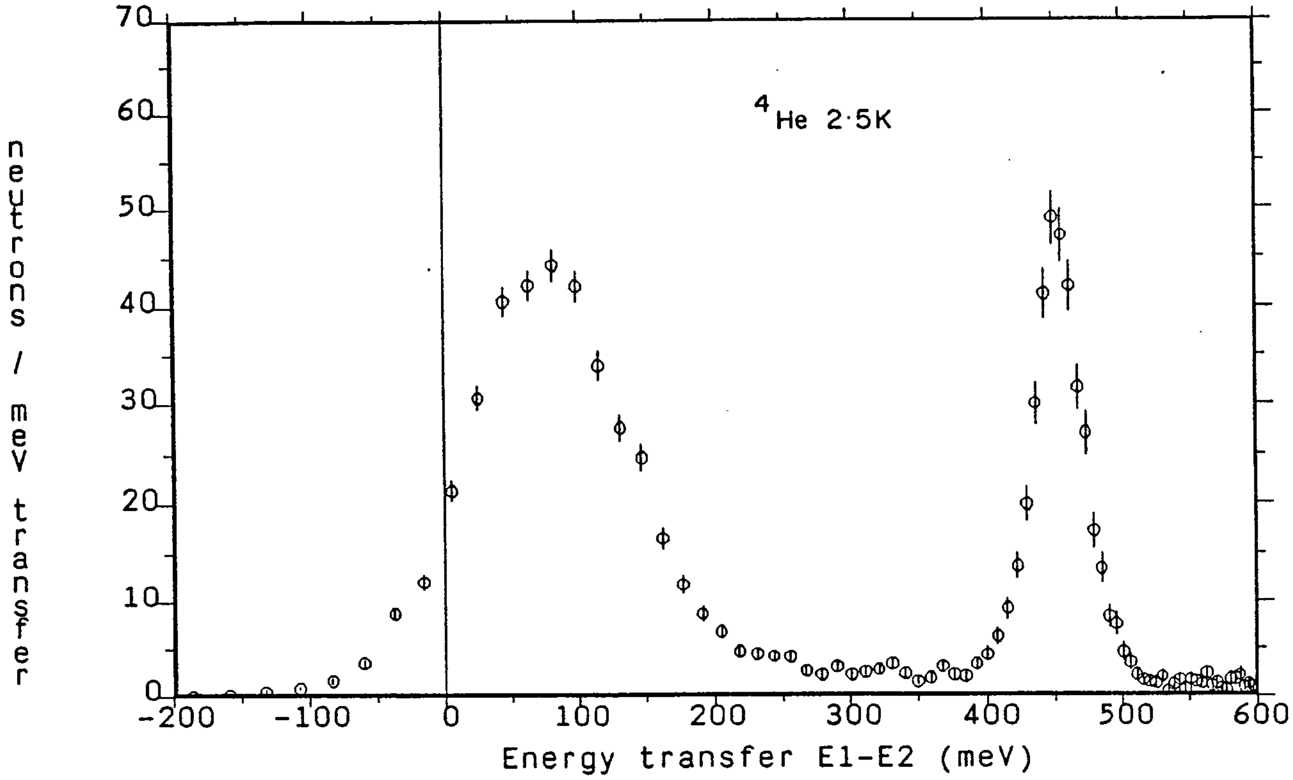


Figure 9 Recoil scattering at 450 meV from liquid helium at 2.5 K measured at an angle of 124°. The incident energy was 828 meV giving a momentum transfer at the recoil energy of 30 Å^{-1}. These data were collected in 1100 μA-hr and the background from the cryostat in 250 μA-hr.

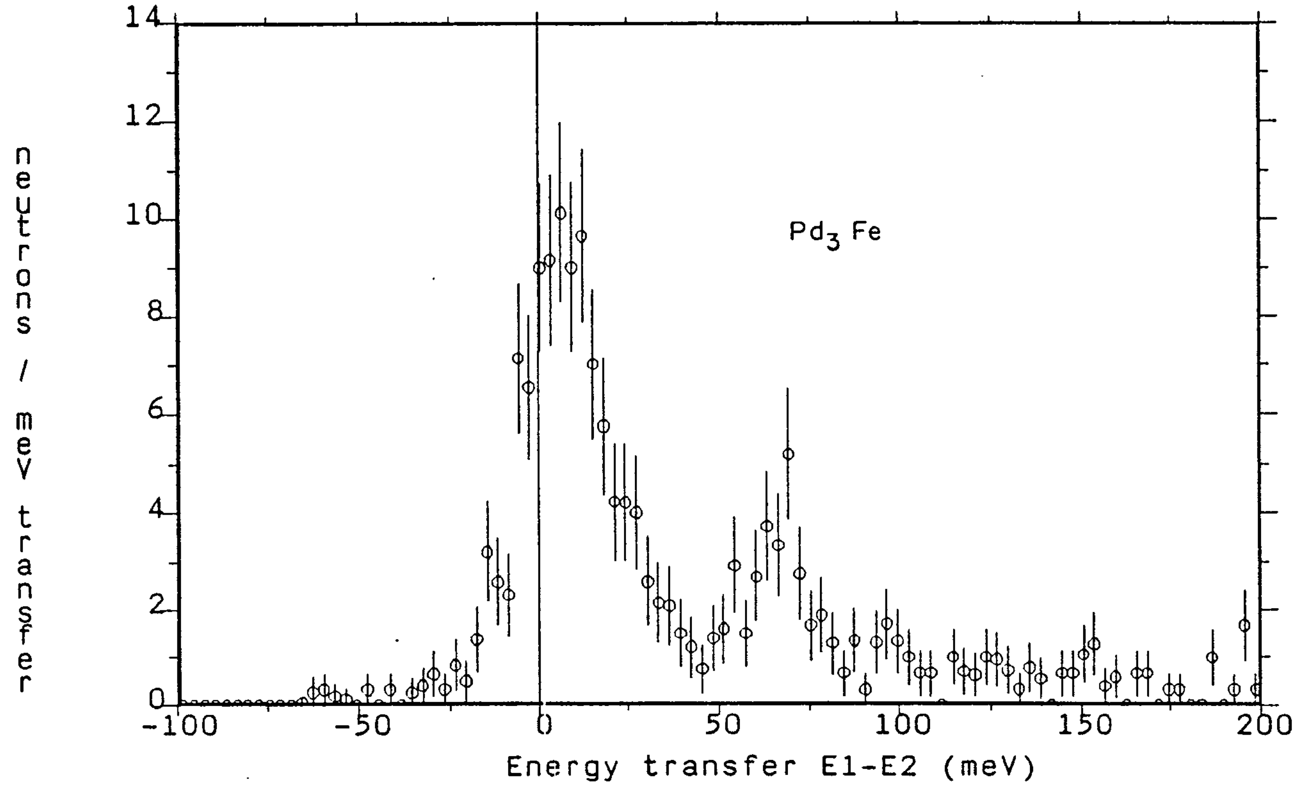


Figure 10 The excitation at 66 meV is a magnon in the 111 direction of Pd_3Fe measured in 300 μA-hr with an incident energy of 230 meV. These data are in agreement with the dispersion curves of reference [8].