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Advances in Crystal Analyser Spectroscopy at ISIS

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Abstract

A review of the development of crystal analyser spectrometers on reactors and pulsed sources is given for both incoherent and coherent inelastic spectroscopy. The status of and developments on the three operational instruments IRIS, TFXA and PRISMA at ISIS are given. Particular attention is paid to the improvement of resolution and background in present and future generation instruments.

Introduction

Crystal analyser spectrometers have a fundamental advantage over direct geometry spectrometers because of the opposite handedness of the Q,ω locii as shown in figure 1. This means that, when studying cold samples in their excitational ground state, the neutron energy loss technique can still be employed, giving access to large energy and momentum transfer ranges, yet maintaining the instrumental resolution. The resolution can thus be decoupled from the energy transfer and the final analysed neutron energy can be very low

if high resolution is required. Since neutrons of the analyser energy need not be present with any intensity in the incident beam the development of synthetic long wavelength analysers, such as Langmuir Blodgett films, with d-spacings beyond that of natural crystalline materials, could be considered in the future.

A disadvantage of the technique however is that the full white beam travels into the spectrometer and falls onto the sample and its environment. This generates a neutron population within the analyser tank which is, perhaps, a hundred times higher than on a similar performance direct geometry spectrometer. The effect becomes more severe the higher the ratio of energy transfer range to resolution width. The problem can be overcome despite early pessimism expressed in an IAEA panel meeting in 1969:

H A Mook	"Perhaps you could use a time-of-flight spectrometer with the flight path first, then use a crystal to analyse the neutrons..."
B N Brockhouse	"I think it would be very difficult to work in the main beam with a single crystal analyser ..."
G Dolling	"It should be pointed out that you really are making things hard for yourself when you put your specimen in the main beam".

However it does mean that collimation of the incident and scattered beams must be effective and that internal shielding must be well defined and universal. Also the crystal used as the analyser itself must have a very high contrast for the desired energy neutrons compared with other energies in the incident beam. In practice this means that the ratio of the coherent elastic cross section to the sum of the coherent inelastic and incoherent cross sections must be minimised.

A significant bonus however on white beam crystal analyser instruments is that a diffraction pattern of the sample can be recorded simultaneously with the inelastic spectrum. This aids the diagnosis of structural phases when studying complex materials with multiple phase transitions and hysteresis effects such as displayed by liquid crystals. It also allows structural parameters to be linked to spectroscopic observations in the study of samples uniquely prepared in beam such as intercalates, or those which are undergoing transformations in time.

Early Instrumentation

The so-called beryllium filter spectrometer is the archetypal crystal analyser instrument. Set against the triple axis spectrometer and the direct geometry chopper time of flight instruments on reactor sources it developed a tarnished reputation to the extent that it almost attained the status of a term of abuse! This was largely due to the poor resolution, caused by the wide band pass of the filter and the poor definition of the incident energy, and the increasingly higher background as the energy transfer increased, mainly due to coherent inelastic scattering in the monochromator. Attempts to improve its performance, such as by using a chopped white-beam to sort the incident neutron energy by time of flight or by the use of filter difference techniques had only limited success.

Quasielastic and Tunnelling

The first high precision instrument in the crystal analyser class was the backscattering machine developed firstly at Munich and then at Jülich before emerging at ILL as IN10. This instrument generates its white beam (over a 30μeV window) by Doppler shifting the Bragg reflection from a silicon monochromator using a mechanical piston, and analyses the scattered beam with a very large area silicon analyser mirror in exact backscattering geometry, giving an energy resolution of 1μeV. This concept has been extended to higher momentum transfers with the design of IN13 which uses a similar geometry to IN10 but employs CaF_2 as monochromator and analyser. The white beam scan over 450μeV, is achieved by temperature cycling the monochromator.

The backscattering principle was applied to pulsed sources in the design of the IRIS spectrometer where the incident white beam is naturally present at the end of a long incident flight path. The analyser consists of a large array of graphite crystals (1350cm^2) in a geometry close to backscattering, giving a resolution of 15μeV over an energy transfer range of 3meV. The decision not to go to exact backscattering in the instrument design eliminated the need for a beam modulation chopper before the sample, a device used on IN10 to discriminate against neutrons scattered directly into the detector from the sample. This decision increased the expected count rate on IRIS by a factor of 4 (2.5 x 1.5) since the saving in the instrument budget was used to increase the graphite area by 50% and the chopper transmission would have been 40%. The instrument is shown schematically in figure 2.

At KENS, the development of such a high resolution instrument followed a different path involving firstly a graphite analyser instrument with an 80° take-off angle (LAM 40) followed later by a near-backscattering instrument with a 160° take-off angle (LAM 80). This has been further developed with the addition of mica analysers and the instrument is now known as LAM 80 ET.

Molecular Spectroscopy

On pulsed sources, the first appearance of a crystal analyser machine was at IPNS where a copper analyser at relatively low take-off angles was used with limited success, largely because of the moderate resolution due to the $\cot\theta$ term and the difficulty of achieving effective focussing. At KENS a fundamental improvement in the design was achieved by the realisation that a simple focusing geometry could be built which allowed the resolution line width to be preserved and the intensity to be enhanced. This geometry is shown in Fig 3. The parallel arrangement of the sample, analyser and detector bank ensures that the time of flight of neutrons from the sample to the detector is independent of the neutron energy selected at different points on the analyser. This concept has been extended in the design of the time focusing crystal analyser spectrometer TFXA at ISIS as shown in Figure 4 to include separate recording of individual detector elements and a subsequent merging in software.

Coherent Excitations

A different developmental direction again of the crystal analyser technique has led to the design of instruments on pulsed sources able to study coherent excitations in single crystals. The first such instrument was the constant-Q spectrometer installed at the Harwell linac. This utilised a large monolithic flat single crystal of germanium as an analyser, placed after the sample. Different points on the analyser select different final energies thus generating a 'constant-Q' scan in reciprocal space. At LANSCE this concept has been further developed with the separation of the analyser into a number of individually controllable cylindrical analyser crystals still with the overall geometry of a flat plate. The mosaic spread of each crystal has been separately selected in an attempt to equalise the resolution width across the scan.

At KENS a variation on this theme led to the development of a multiple analyser crystal spectrometer MAX. This idea is more flexible than the constant-Q design and enables a scan along a particular reciprocal lattice direction to be measured in one setting of the instrument. This has been further refined in the building of the joint Anglo-Italian instrument PRISMA at ISIS with its 16 individually controllable analyser detector arms. It is shown in Figure 5. Recently the rotating collimator has been replaced by a t-zero nimonic chopper to reduce further the flux of fast neutrons onto the sample.

Using a single rotating analyser and a linear position sensitive detector, the ROTAX spectrometer is being built at ISIS by a team from the University of Würzburg. ROTAX is an ambitious attempt to create the flexibility of a triple axis instrument on a pulsed source spectrometer and installation will begin in 1992.

Instrument Performances & Future Developments

IRIS

The present (January 1991) characteristics of IRIS are listed in Table 1. A set of resolutions from 1μeV to 50μeV can be accessed, each with an increasing range of Q. The energy transfer ranges with each resolution is limited by the adjacent analyser reflections. A beryllium filter, cooled to 25K, has been developed for use with the graphite (002) reflection which will increase the energy transfer range to approximately ~10meV, determined by the limit of transmission down the curved neutron guide. Furthercommissioning work is needed before this option comes into service fully.

The underlying broad feature beneath the elastic line from graphite has been positively identified as being due to thermal diffuse scattering from the analyser itself. There are two ways to minimise this. The first involves the use of collimating blades between the analyser and the detector. Unfortunately this solution will also reduce the Bragg intensity and will not affect the relative level of TDS directly under the Bragg line itself. More promising is to reduce the phonon population by cooling the analyser. Tests on IRIS show that there is a linear fall in TDS intensity with temperature down to 50K as shown in Fig 6 for an offset angle from the Bragg reflection of 4°. As a consequence a helium cooling loop is to be installed on the graphite analyser with a design temperature of 25K. When this is operational in mid-1991 the

Table 1: The Characteristics of IRIS (Jan 1991)

Analyser Reflection	Analyser λ Å	Analyser E_f meV	Energy Resolution μeV	Energy and Momentum Transfer Ranges
Mica 002	19.8	0.208	1	250 μeV to ~ 100 μeV 0.09 to 0.7 $Å^{-1}$
Mica 004	9.9	0.83	4.2	300 μeV to ~ 300 μeV 0.18 to 1.4 $Å^{-1}$
Mica 006	6.6	1.87	11	1 meV to -1 meV 0.26 to 1.88 $Å^{-1}$
PG 002	6.7	1.835	15	3 meV to -1 meV 0.25 to 1.85 $Å^{-1}$
PG 004	3.35	7.28	50	5 meV to -4 meV 0.5 to 3.7 $Å^{-1}$

background from TDS sources should improve by an order of magnitude.

Additional valves and filters have been installed in the vacuum system of the analyser tank in order to ensure that vacuum oil vapour does not condense on the graphite and intercalate between the layers.

Interestingly tests of CaF_2 on HRPD also point towards TDS effects being the cause of a similar undiagnosed broad background on IN13.

A recent major development on IRIS has been the realisation of the mica analyser array with an area of ~1500cm^2. This provides a resolution of 11μeV for the (006) reflection with a more clearly defined "moderator edge" than occurs in graphite. This suggests that mica is much more perfect, in terms of

$\Delta d/d$, than graphite and the instrument is clearly mismatched at this distance (38m) from the moderator. The optimum distance for matching is yet to be determined but at 60m the resolution would further improve by 50%.

Despite this mismatch the effective count rate from the mica (006) array is only slightly less than 50% of the count rate from the graphite (002) array. Since mica is 100 times cheaper than graphite a large area array is quite feasible. Plans are in hand to double the present area to 3000cm^2 in the coming months.

The use of the (004) reflection at an analysing energy of 830μeV (λ = 9.9Å) gives an improvement of elastic resolution to 4.2μeV. The intensity of the elastic line is only about 2.5 times lower than for the (006) reflection and is quite feasible to use. Run times of the order 6-10 hours are necessary.

The (002) reflection gives a resolution of 1.0μeV over a wide energy transfer range, as shown in Figure 7. The practical use of this option has been limited up to now because of the 50Hz pulse repetition rate on ISIS. A second pulse leaves the moderator before the 20Å neutrons destined for the mica (002) reflection reach the band pass chopper at 6.4m. Accordingly a second band pass chopper is presently being installed at 10m which should be operational for the first cycle of 1991. The use of 20Å neutrons should then be routine. It does appear that we have underestimated the intensity of long wavelength neutrons in our incident spectrum and the 10 metre chopper should improve the general background in the spectrometer even when working at shorter wavelengths. Because of the low Q range observed (0.07Å^{-1} to 0.7Å^{-1}) this will facilitate the measurement of translational diffusion. Its use for tunnelling spectroscopy is still to be explored - the low Q will be a disadvantage in this case. Plans to study crystal field excitations where the low Q is a positive advantage are in hand.

Mica, as an analyser, offers great promise which has yet to be fully realised. Little is known about its neutron reflecting properties and more development work is required. The value of $\Delta d/d$ for the various chemical isomorphs is unknown and the optimum thickness for maximum reflectivity and minimum background has not been explored. A full survey of the various varieties has yet to be carried out. Nevertheless it is clearly a material worthy of more study. Interestingly it is now being considered as a long wavelength monochromator for IN6 in order to improve the resolution.

On the data analysis side progress has been made in the implementation of maximum entropy techniques for the deconvolation of inelastic spectra. Using the MEMSYS code developed at Cambridge we have been able to deconvolute features in the tunnelling spectrum from lutidine using the graphite (002) analyser data which have subsequently been experimentally observed using the mica (004) analyser. This data, in turn, has been deconvoluted to reveal further structure which, once again has been confirmed in measurements on IN10 using an offset monochromator. In order to obtain reliable results the resolution function of the instrument must be known precisely, as well as its variation with energy transfer. The data and deconvolutions are shown in figure 8. Obviously, until much more experience has been accumulated, the use of the maximum entropy technique will be initially used as a guide to further, higher resolution measurements. However, once confidence has been generated, and the results accepted by the community, it will find universal acceptance in the field of inelastic spectroscopy.

TFXA

The time focussing crystal analyser spectrometer TFXA illustrated in figure 4, has been operational for over five years. During that period it has undergone continous development within the limits imposed by the monolithic design of the analyser shielding tank. It has been established that thick samples may be used, with scattering probabilities as high as 40% of the incident beam, without seriously compromising the spectral quality. The original graphite analysers have been replaced by a more relaxed mosaic spread set of greater thickness. This has increased the detected count rate by a factor of 4. The original detector bank has been replaced with high pressure squashed 1.2cm helium-3 tubes which has improved the instrumental resolution. At the same time substantial reductions in background have been achieved. The quality of the spectra generated on TFXA has moved molecular spectroscopy beyond a threshold and into a region where individual modes can be well-resolved and the effects of lattice modes and multiphonon events can be quantified. This is illustrated in figure 9 where the spectrum from the 72 atom molecule nicotinamide adenine dinucleotide at 30K is shown. As a consequence of this improved spectral quality much recent progress has been made in implementing an iterative data fitting algorithm CLIMAX which uses a generalised molecular force field model to refine the dynamical model of the molecule and its crystalline surroundings. Molecular spectroscopy with neutrons has entered a stage in the quantitative analysis of data which resembles that experienced by powder diffraction in the late 1970's with the advent of Rietveld refinement.

A major development on TFXA in the forthcoming few months will be the installation of a prototype mica analyser replacing one of the graphite analyser arrays. Higher order contamination will be removed with a cooled polycrystalline graphite filter in place of the present beryllium filter. Using the mica (004) reflection it is expected that the resolution of the instrument at energy transfers up to 100meV will improve by approximately 50%.

Separate papers in this conference deal with the PRISMA and ROTAX projects and the reader is referred there for more details.

Future Plans for New Instruments

Operational experience in the past few years has put us in a strong position to capitalise on the advances achieved with the first generation instruments and their developments. It is clear that high resolutions can be obtained quite readily on pulsed source instruments and that scientific advances follow such improvements in resolution. Equally well the original designs were somewhat conservative being designed without the benefit of today's experience.

Therefore in laying down plans for future instruments the direction is clear. We must further improve resolution and range. The improvement of intensity, for its own sake, is of limited value.

Accordingly plans have been drawn up to separate the high and medium resolution options of IRIS into two instruments on twin guide tubes from the same beam hole. IRIS-2 will be a spectrometer which seeks to optimise the use of mica with resolutions improved by a factor of 2 and employing very large area analysers in order to utilise to the full the plentiful supply of cold neutrons from the hydrogen moderator. IRIS-1 will contain the cold graphite analysers, increased in area by a factor of two. The space released by the present mica analyser will be used to install a large position sensitive diffraction detector for long d-spacing diffractometry up to 30Å with high intensities and a resolution $^{\Delta d}/d$ of 2.5×10^{-3}.

Molecular spectroscopy will be further advanced by the construction of an instrument, Cassandra, having 0.3% resolution in energy transfer, a factor of 3 better than currently available, over a range from 5 meV to 100meV. Cassandra will use a large area mica analyser in near backscattering geometry on the end of a 60 metre long curved supermirror guide.

The experience gained on PRISMA in the last two years is already indicating the scientific value of surveying coherent excitations over a two-dimensional net. Tests of a prototype dog-leg double analyser, currently in the design stage, will indicate the optimum layout for PRISMA-2 which it is hoped to define by 1992.

Acknowledgements

We are grateful to Professor R E Hester of York University, Dr F Fillaux, CNRS, Thiais and Dr R N Silver, LANL for the use of their results prior to publication.

Figure Captions

Fig 1. The Q,ω locii for direct and inverted geometry spectrometers with a defined energy, either E_i or E_f, of 1.8meV as a function of neutron energy transfer $E_i - E_2$. The dashed line is the locus for an inverted geometry instrument and the full line is that of a direct geometry machine.

Fig 2. A cut away view of IRIS the backscattering instrument at ISIS showing its dual arrays of graphite and mica analysers.

Fig 3. The time focusing geometry for the crystal analyser spectrometer first implemented at KENS.

Fig 4. A schematic view of TFXA the time-focused crystal analyser spectrometer at ISIS.

Fig 5. The 16 analyser-detector instrument PRISMA at ISIS.

Fig 6. The effect of cooling on the thermal diffuse scattering from graphite around the (002) reflection. The crystal has been offset 4° from the Bragg position. The inset shows the integrated intensity in the phonon "wings" as a function of temperature.

Fig 7. The observation of 1μeV resolution on IRIS using the mica (002) reflection at an analysing wavelength of 19.8Å. Note the wide energy transfer range.

Fig 8. Data and deconvolutions of tunnelling spectra in 2,6 lutidine on IRIS and IN10: (a) Data and deconvoluted image from PG(002) analyser (b) Data and deconvoluted image from Mica (004) analyser (c) Deconvoluted image as in (b) and data from IN10.

Fig 9. The molecular spectrum of nicotinamide adenine dinucleotide at 30K measured on TFXA.

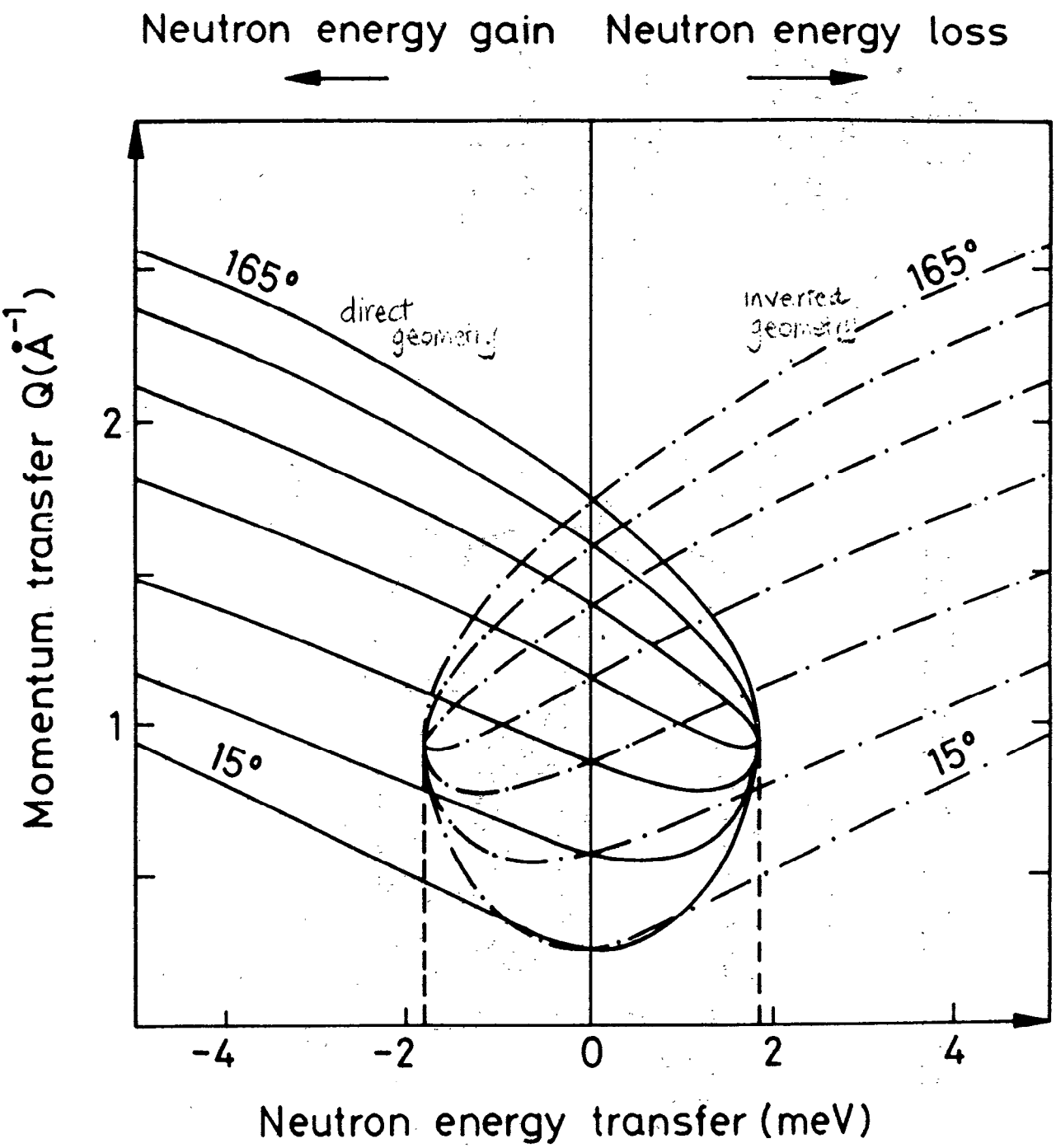

Neutron energy gain
Neutron energy loss
Momentum transfer Q(Å⁻¹)
165°
direct geometry
inverted geometry
165°
15°
15°
2
1
-4
-2
0
2
4
Neutron energy transfer (meV)

FIGURE 1

IRIS CRYSTAL ANALYSER SPECTROMETER

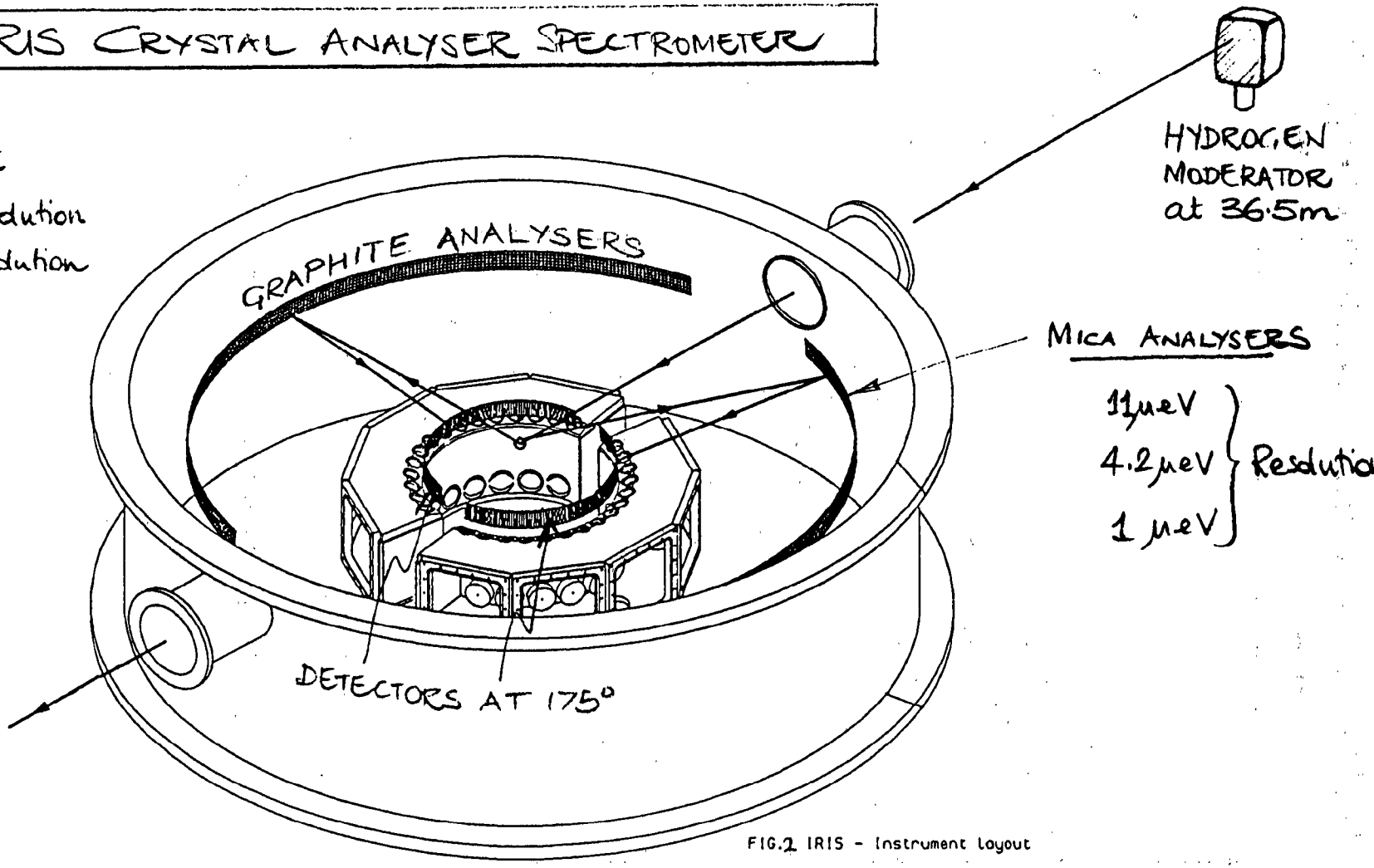


FIG.2 IRIS - Instrument Layout

FIGURE 2

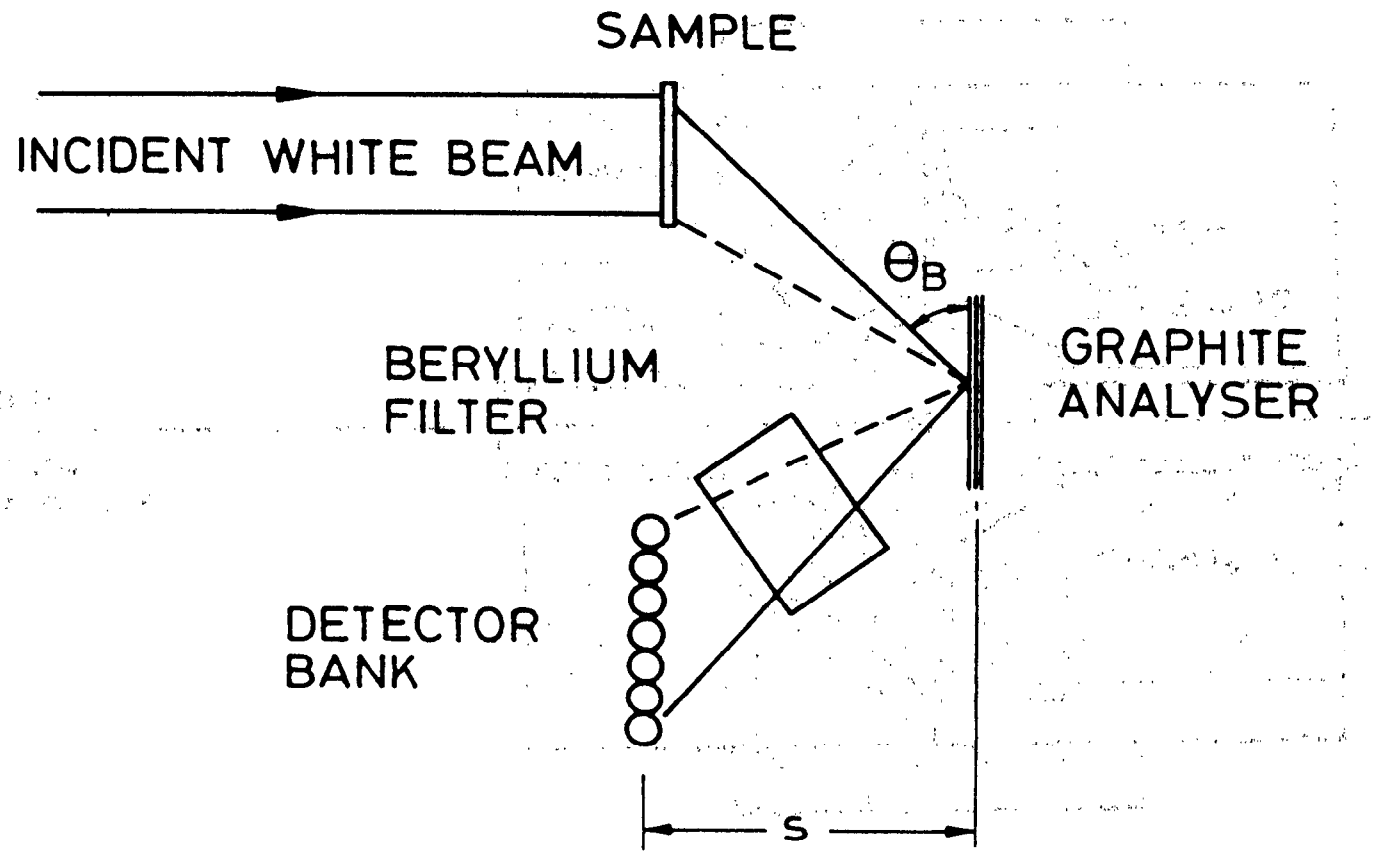

SAMPLE
INCIDENT WHITE BEAM
θ_B
BERYLLIUM FILTER
GRAPHITE ANALYSER
DETECTOR BANK
s

FIGURE 3

TIME FOCUSSED CRYSTAL ANALYSER SPECTROMETER

$\Delta E/E \sim 1.5\%$ $E_1 = 5$ meV to 2000 meV

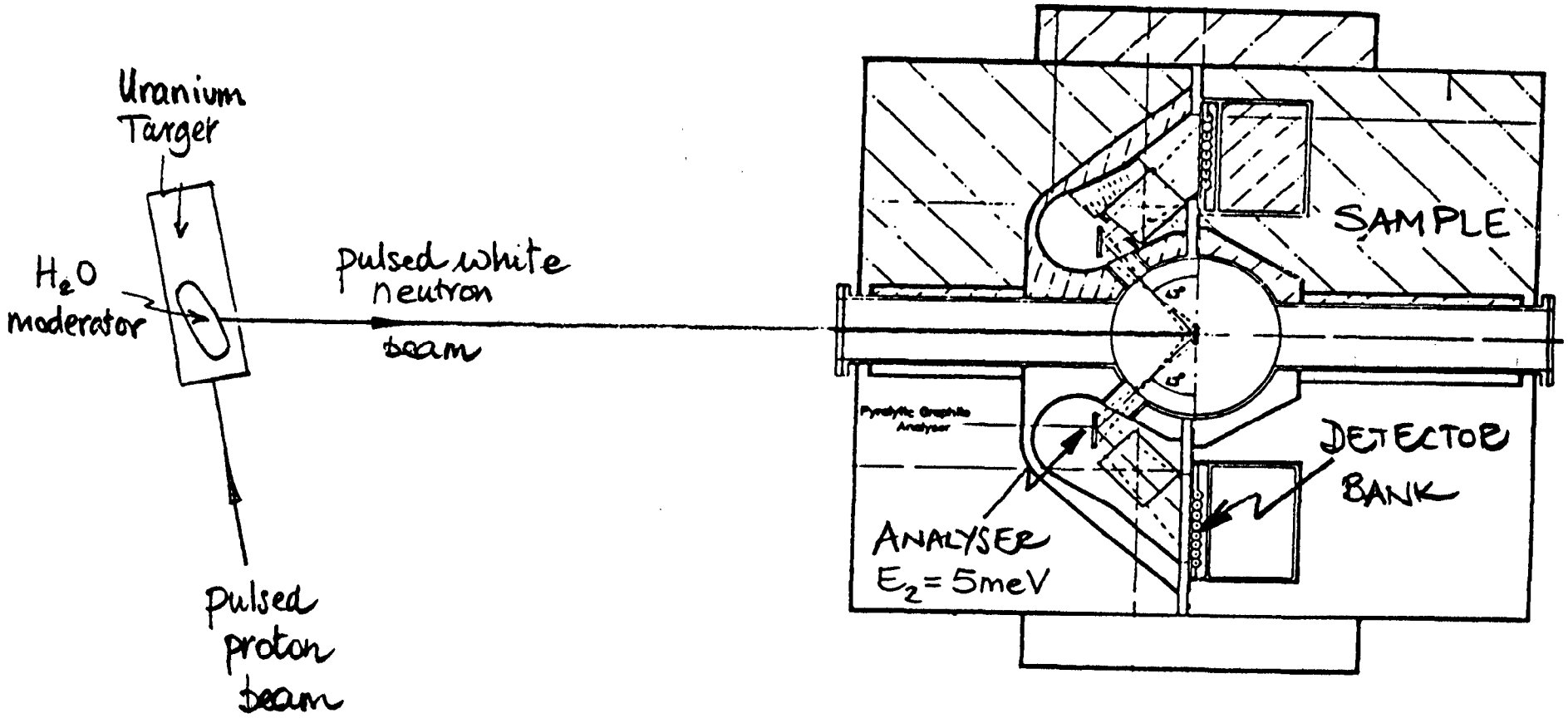
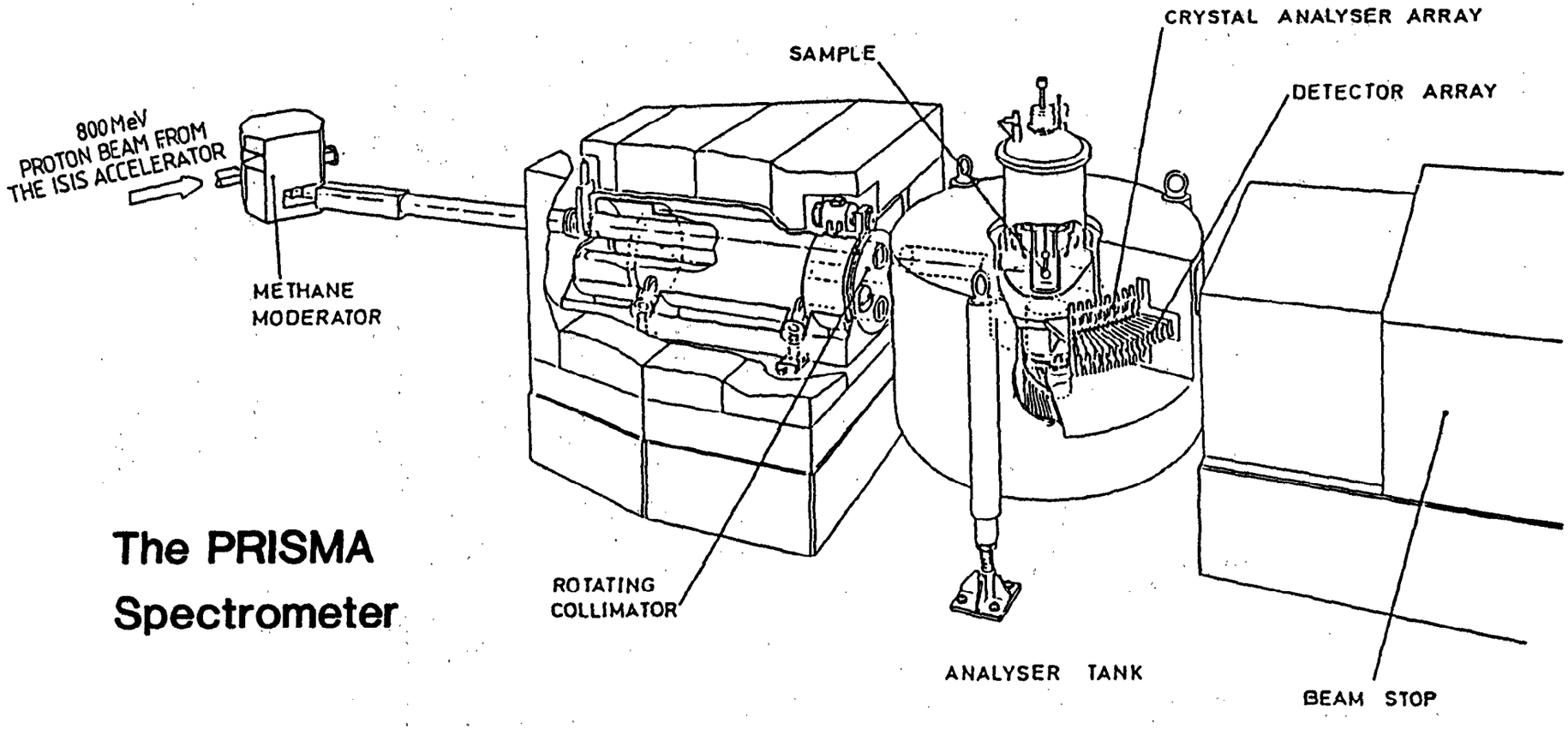


FIGURE 4



The PRISMA Spectrometer

FIGURE 5

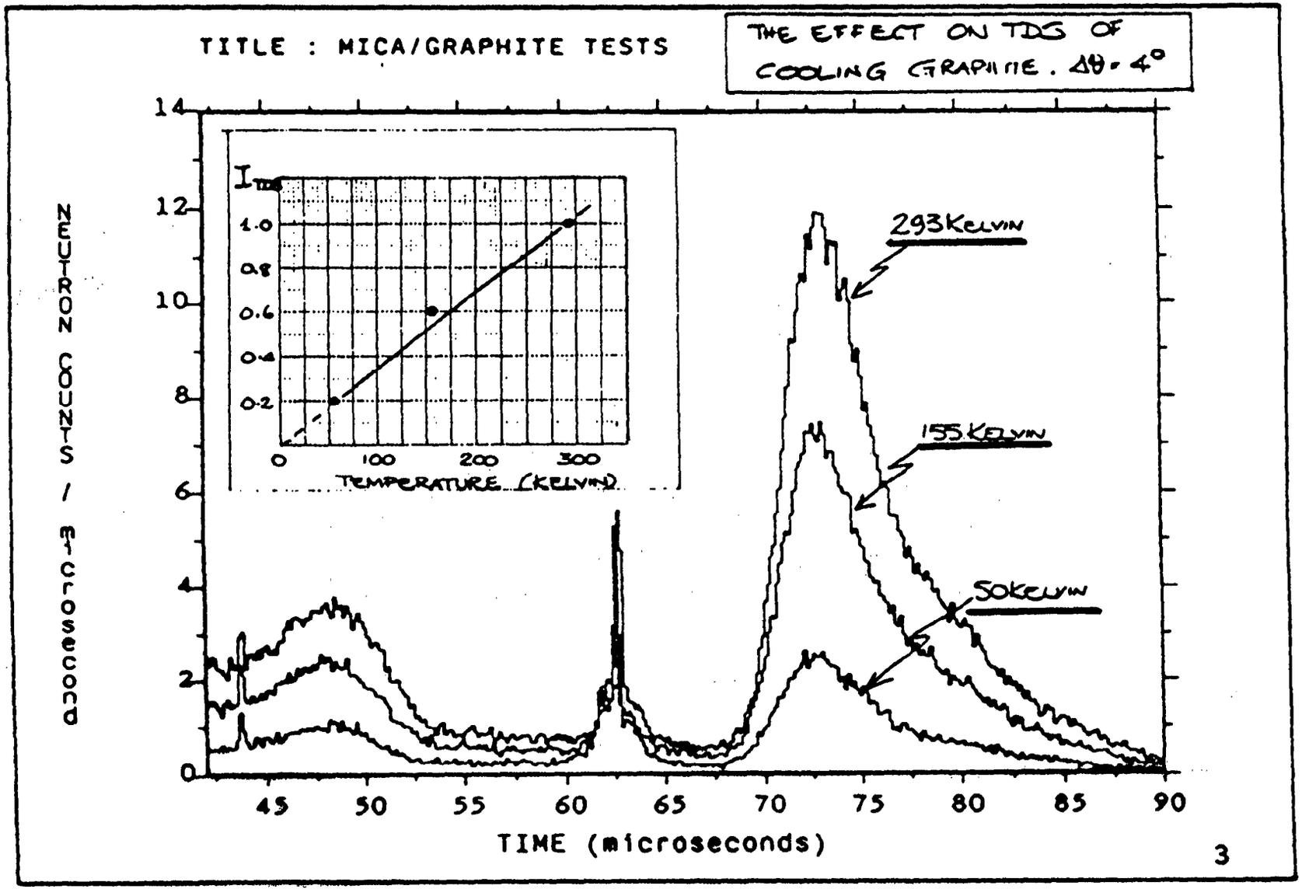

TITLE : MICA/GRAPHITE TESTS
THE EFFECT ON TDS OF COOLING GRAPHITE. Δθ = 4°
NEUTRON COUNTS / 8 microseconds
TIME (microseconds)
293 Kelvin
155 Kelvin
50 Kelvin
TEMPERATURE (KELVIN)
0
100
200
300
0.2
0.4
0.6
0.8
1.0
0
2
4
6
8
10
12
14
45
50
55
60
65
70
75
80
85
90

FIGURE 6

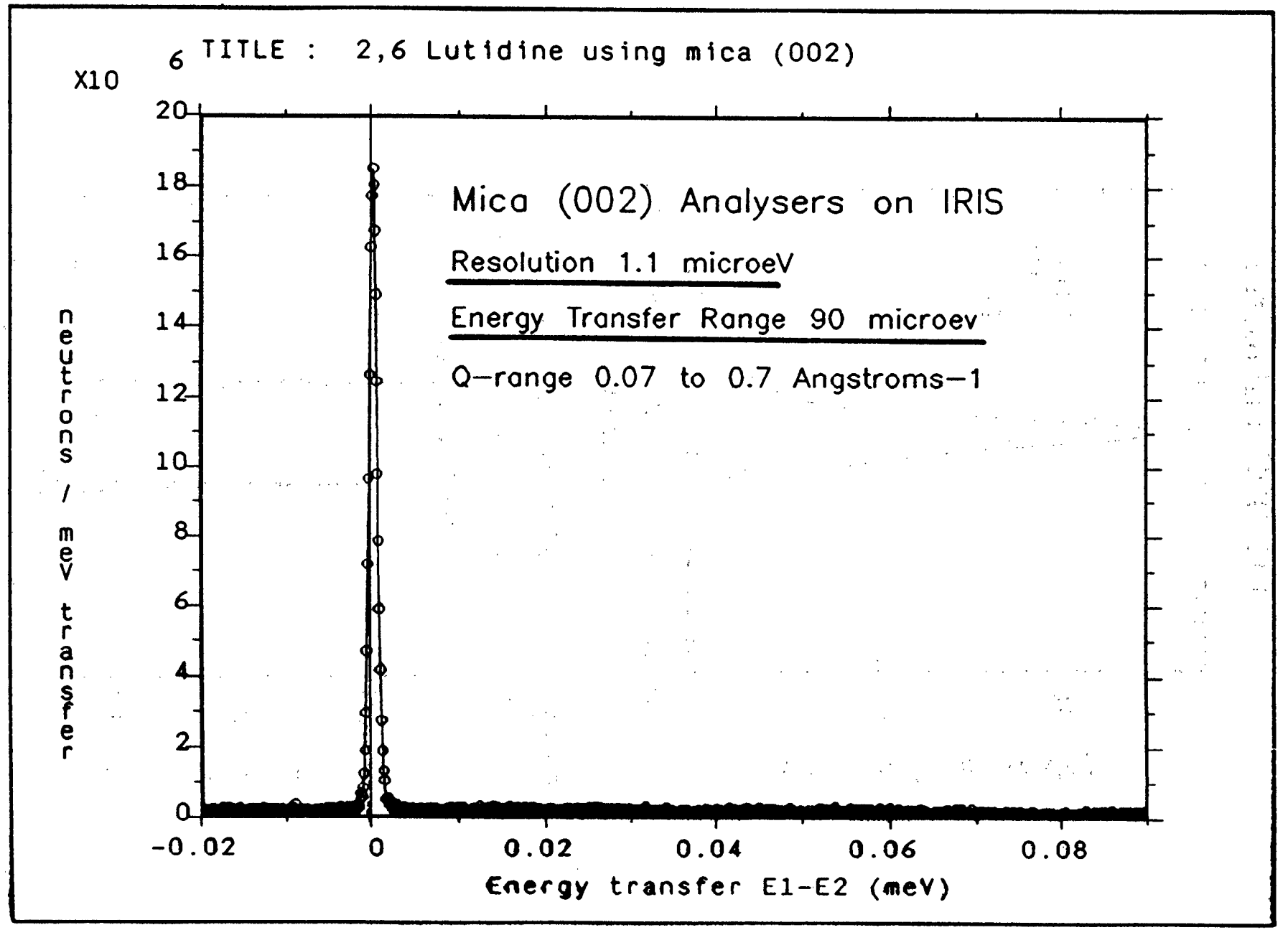

TITLE : 2,6 Lutidine using mica (002)
X10 6
Mica (002) Analysers on IRIS
Resolution 1.1 microeV
Energy Transfer Range 90 microev
Q-range 0.07 to 0.7 Angstroms-1
neutrons / mev transfer
Energy transfer E1-E2 (meV)
20
18
16
14
12
10
8
6
4
2
0
-0.02
0
0.02
0.04
0.06
0.08

FIGURE 7

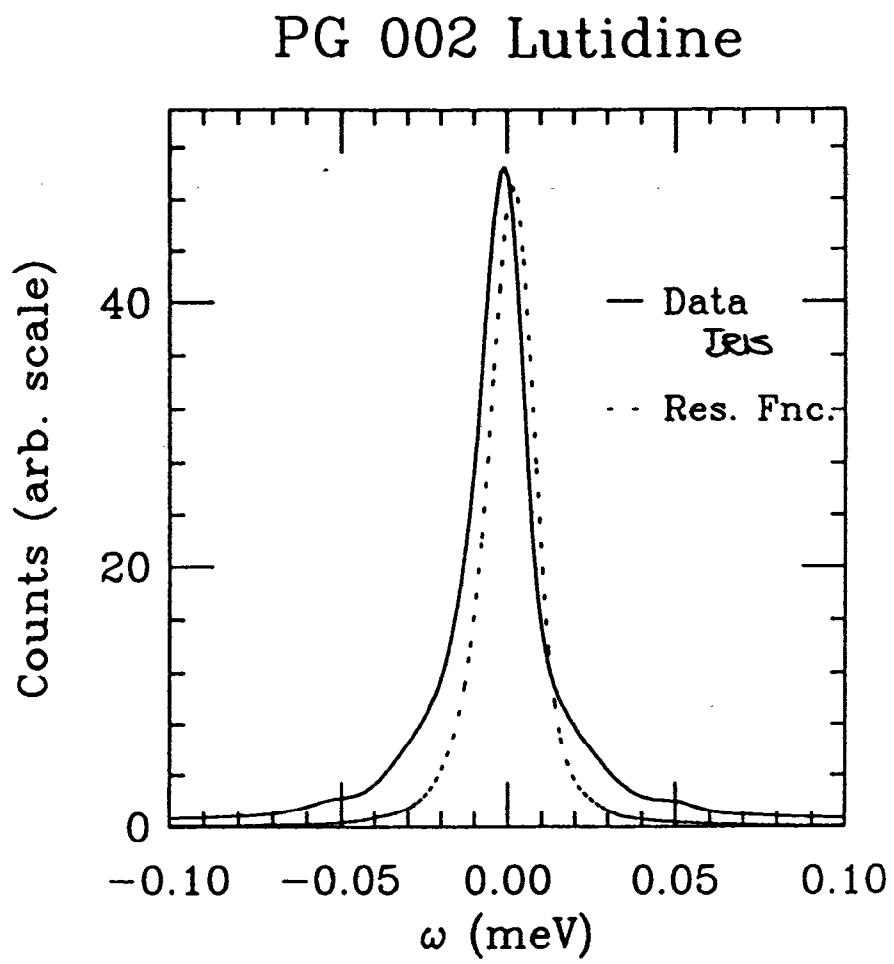
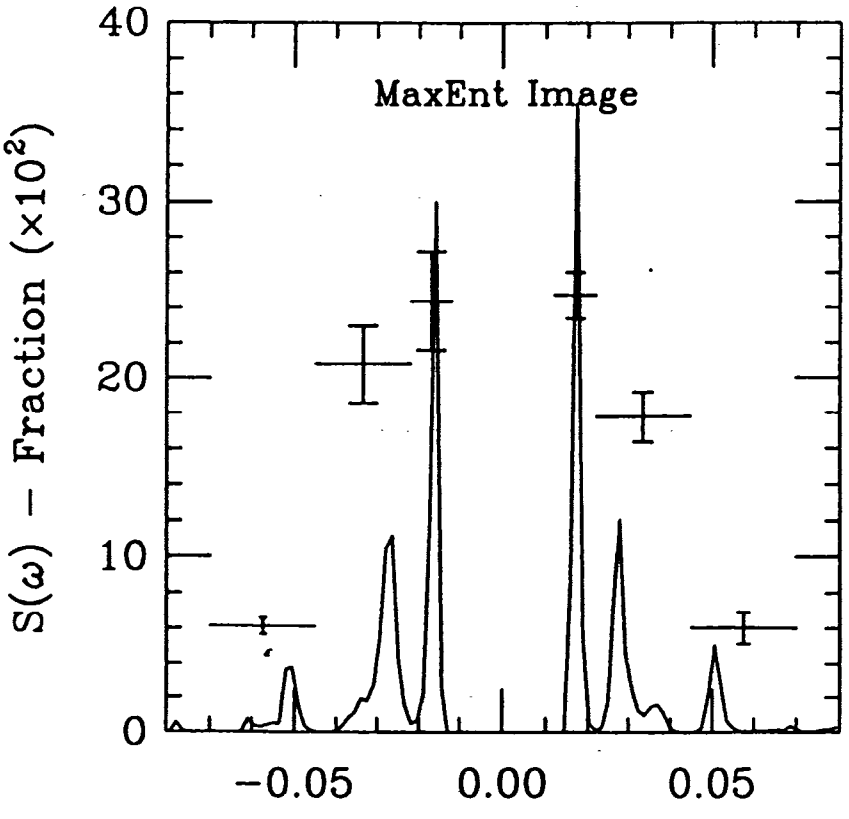


FIGURE 8(a) Lutidine PG002



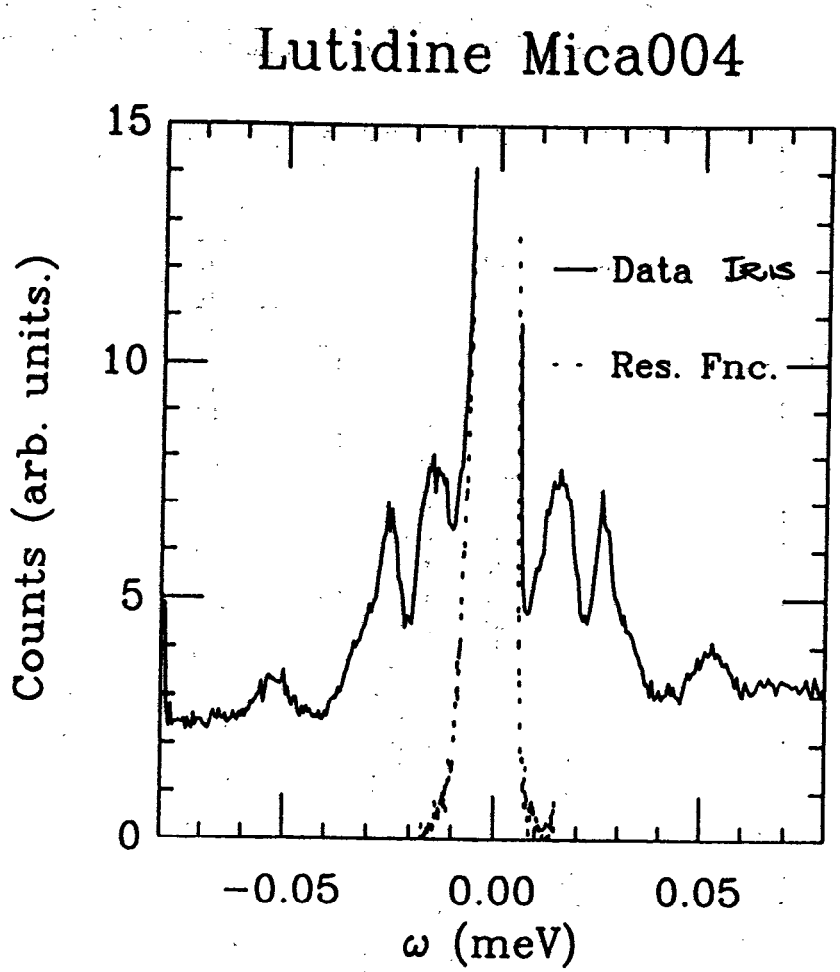
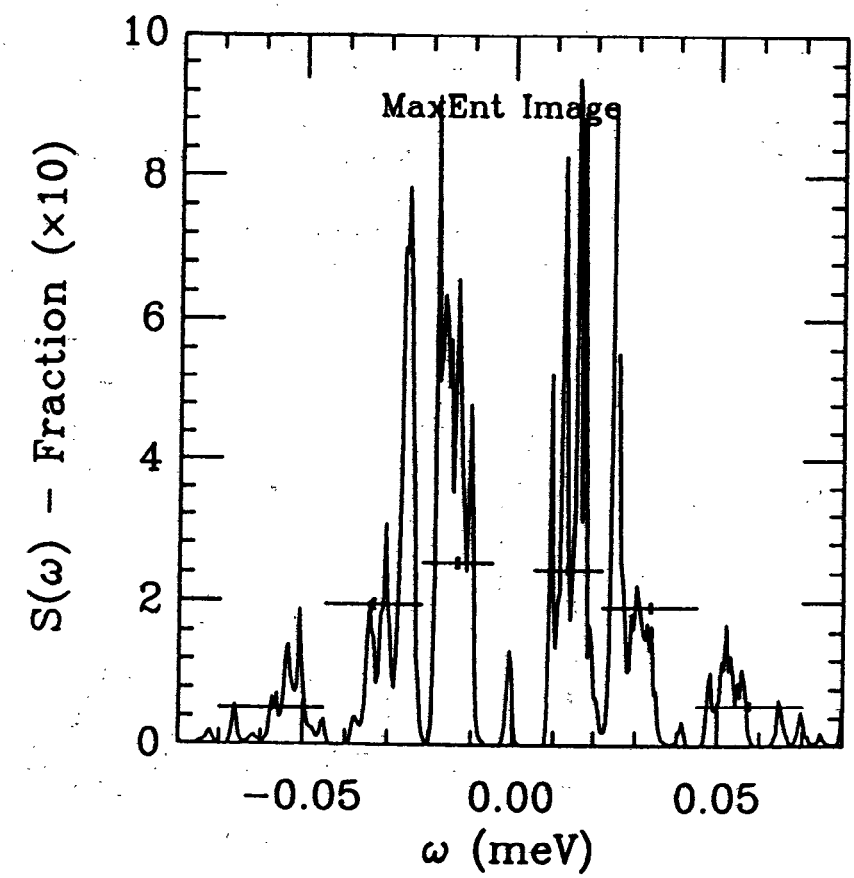

Lutidine Mica004
Counts (arb. units.)
15
10
5
0
Data IRIS
Res. Fnc.
−0.05
0.00
0.05
ω (meV)

FIGURE 8(b)


Lutidine Mica004
MaxEnt Image
S(ω) - Fraction (×10)
10
8
6
4
2
0
−0.05
0.00
0.05
ω (meV)

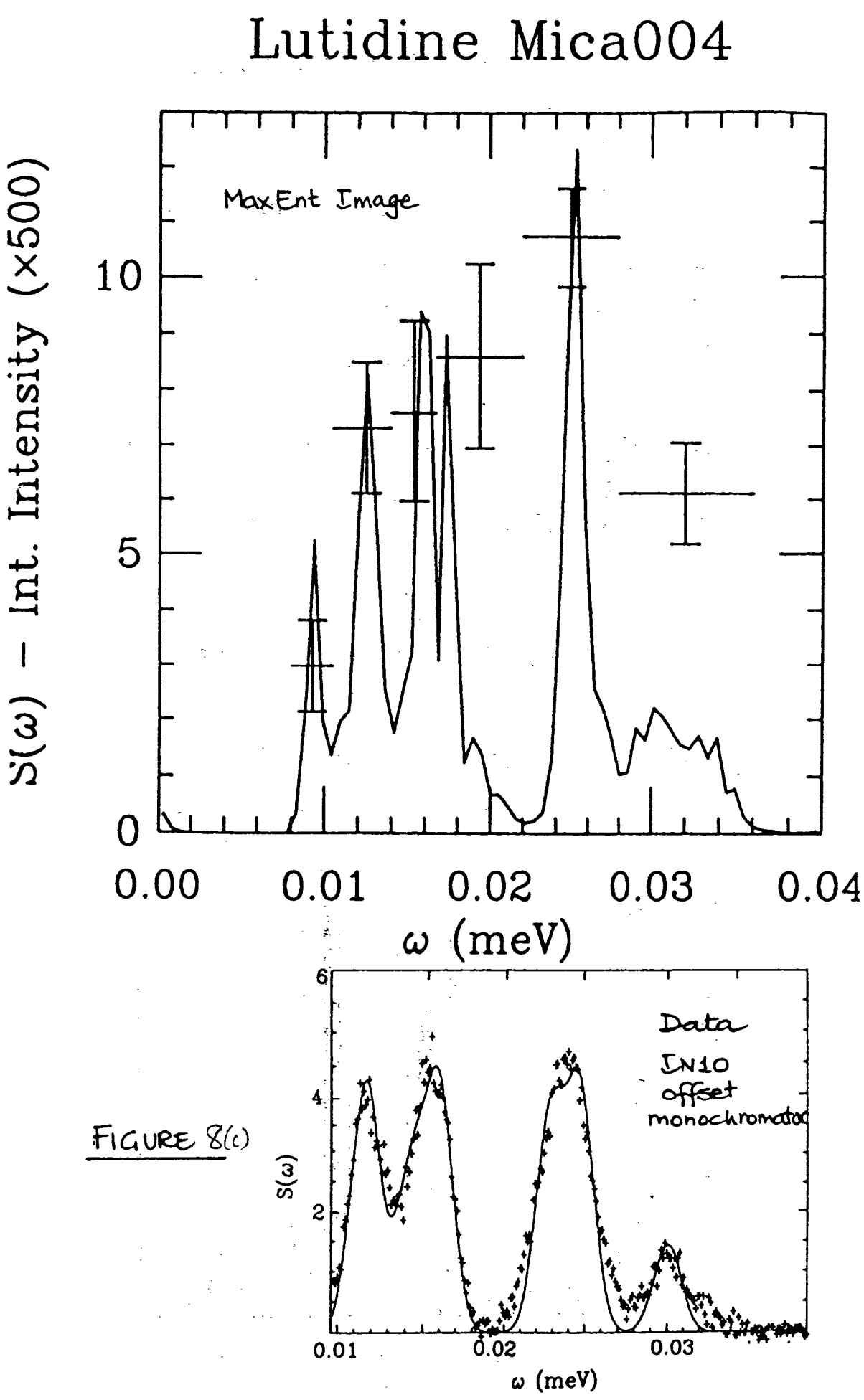

Lutidine Mica004
MaxEnt Image
S(ω) - Int. Intensity (×500)
0
5
10
0.00
0.01
0.02
0.03
0.04
ω (meV)
Data
IN10
offset
monochromato
S(ω)
2
4
6
0.01
0.02
0.03
ω (meV)

FIGURE 8(c)

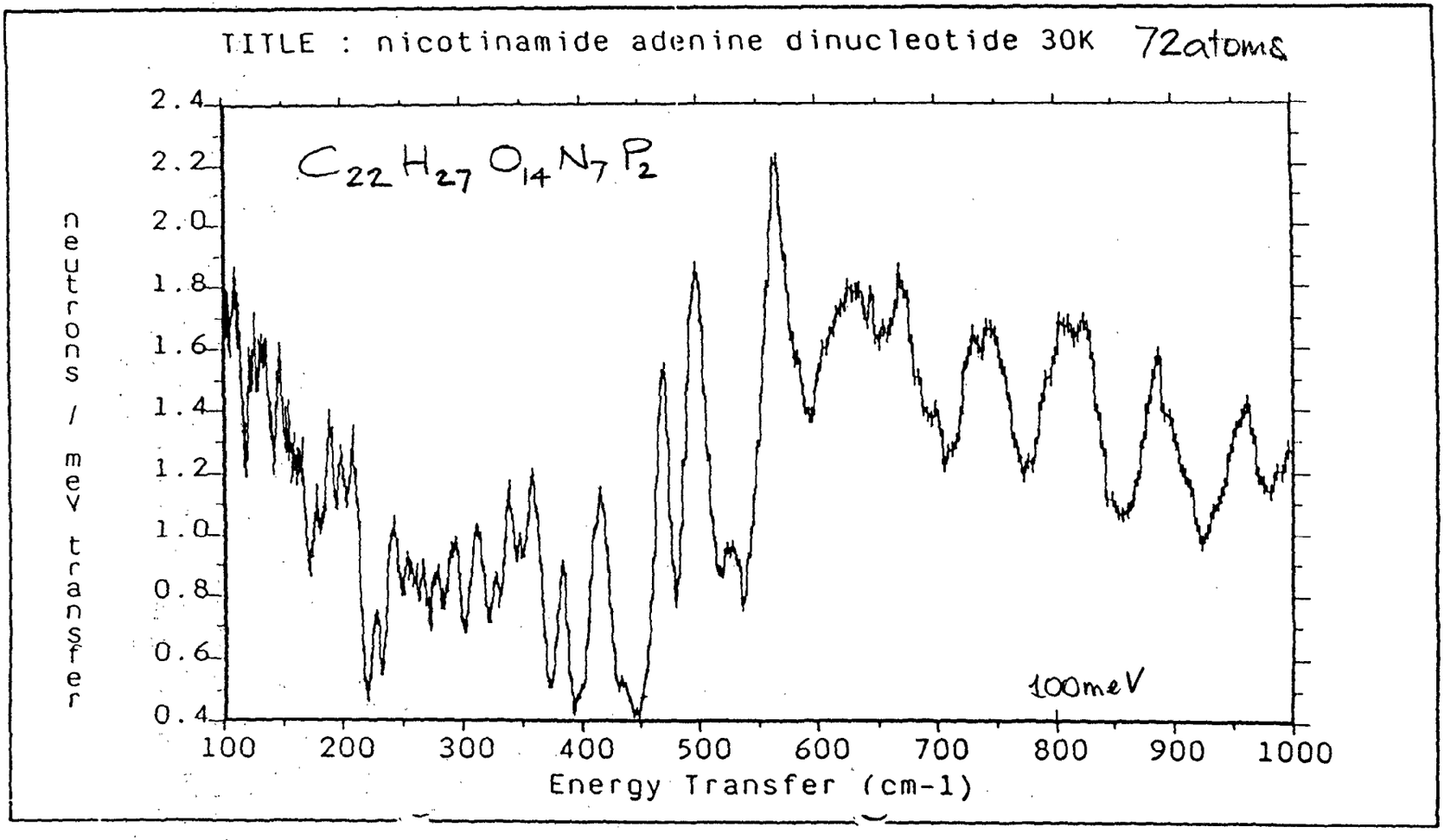

TITLE : nicotinamide adenine dinucleotide 30K 72atoms
$C_{22}H_{27}O_{14}N_7P_2$
100meV
neutrons / mev transfer
Energy Transfer (cm-1)
2.4
2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.8
0.6
0.4
100
200
300
400
500
600
700
800
900
1000

FIGURE 9