MAPS: A Chopper Spectrometer to Measure High Energy Magnetic Excitations in Single Crystals

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0. ABSTRACT

An increasing amount of the scheduled time of both HET and MARI has been spent measuring high energy magnetic excitations in single crystals. Despite the inherent limitations of current instruments, which were never optimised for such experiments, a growing number of experiments have been successfully carried out in 1-, 2- and 3-dimensional magnetic systems. The proposed spectrometer MAPS aims to dramatically increase the efficiency of these experiments and thereby to open the 100-1000meV energy transfer range in the same way that the triple axis spectrometer opened the 1-50meV range.

1. INTRODUCTION

The triple axis spectrometer has for three decades been the instrument of choice at reactors to investigate collective excitations in single crystals. It offers full flexibility in the choice of scan through (**Q**,ω)-space and, through collimation and monochromator

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changes, the instrument resolution can be adjusted to suit the problem being investigated. However, at high energy transfers the kinematic constraints placed by the requirement of small |**Q**| in magnetic experiments means that a large incident energy is needed, when the flux from a reactor is low and monochromator reflectivities are small. The small scattering angles at the monochromators and sample mean that the background from air scattering is large and a rapidly changing function of scattering angle. Single crystal experiments have occasionally been performed on time-of-flight chopper spectrometers, but because the paths in **Q** and ω are correlated, the spectrometers have almost exclusively been used with polycrystalline samples where the only **Q** dependency of the excitations is due to the magnetic form factor, or to measure excitation density of states.

Pulsed sources offer a number of advantages at high incident neutron energies over reactors, particularly a large epithermal neutron flux and low background between the pulses. However, it was not envisaged that high energy experiments in single crystals would form a large part of the experimental program at ISIS, and accordingly the principal considerations in the design of the HET spectrometer centred on its utility for measuring dispersionless excitations in polycrystalline samples. Nevertheless, experimental techniques have been developed that use the accessed volume of (**Q**,ω)-space in single crystals to advantage, and a wide range of highly succesful experiments have been performed on 1-, 2- and 3D magnetic systems [2-8].

Such experiments require considerable beam-time, and now account for a significant proportion of the time requested on HET and MARI. Accordingly, it was considered timely to request funds to build a spectrometer purpose designed to measure high energy magnetic excitations in single crystals [1], because in the light of the experience gained on HET and MARI significant optimisations can be made in the basic design that should allow a large increase in the efficiency of data collection. Advantage can be taken of the proposed flexibility of instrument configuration to allow easy incorporation of new beam components, either for test purposes (e.g. improved collimation) or as technological improvements occur (e.g. white beam polarisation). MAPS will act as a test bed for future developments of the time-of-flight chopper spectrometer both in hardware and data visualisation software.

In section 2 an outline is given of the methods used to perform single crystal experiments on HET and MARI, and the instrument developments that could be made to increase the effectiveness of the experiments are detailed with reference to experience gained on HET and MARI. Section 3 gives details of the proposed design of MAPS, and in section 4 the questions of software and data visualisation needs are briefly considered.

2. DESIGN CRITERIA

2.1 Performing single crystal experiments [1]

The most common method used with **3D systems** (e.g. Fe [2], Co [3], Cr [4]) is to arrange for a high symmetry direction of the crystal to lie along the direction of $\mathbf{k_f}$ within a chosen detector in the scattering plane (high symmetry plane)of the crystal. The path of energy transfer as a function of Q along this direction is an inverted parabola and where this parabola intersects the dispersion relation peaks in intensity will be observed (figure 1). Several detectors may be added together to improve the statistics, but as the Q resolution is degraded perpendicular to the symmetry direction, and hence perpendicular to the gradient of the dispersion relation, the peaks can remain sharp. To intersect the dispersion relation at a higher energy transfer, the incident energy is increased. To maintain the symmetry direction along $\mathbf{k_f}$ requires rotating the crystal a few degrees so that the symmetry direction is now along $\mathbf{k_f}$ in a detector with scattering angle also a few degrees different. With a series of runs at different E_i the full dispersion relation can be mapped out.

In **2D systems** the scattering is independent of the component of Q perpendicular to the 2D plane so forming rods of scattering in that direction. In some experiments (e.g. La_2NiO_4 [5]) the same technique as with 3D systems has been used, with the rods perpendicular to the scattering plane. An alternative arrangement (e.g. La_2CuO_4 [6]) has been to align the rods along $\mathbf{k_f}$ in a chosen detector, and compare the signal in adjacent detectors. The differences in the spectra arise from the different points of intersection of the detectors on the dispersion relation, and models for the dispersion relation can be tested by comparing the calculated and observed differences. In **1D systems** (e.g. $KCuF_3$ [7], $KFeS_2$ [8]) the chains are aligned along $\mathbf{k_i}$ so the scattering has azimuthal symmetry. A large solid angle of detectors can be summed with little degradation of Q along the chain. The background can be estimated from detectors in the vertical plane after the crystal has been rotated through 90 degrees because in these detecotrs the component of Q along the chains is zero.

The success of HET and MARI with single crystal experiments is due to the low background and the large detector area. The sample and detector tanks are evacuated to eliminate air scattering and so the bulk of the background is from multiple scattering within the sample. Data is gathered over a wide range of momentum and energy transfers simultaneously, so all points on a scan are gathered at once. The broad coverage in (Q,ω)-space, together with the low background, explains how the performance can be competitive with a triple axis spectrometer. but it also allows a better understanding of the behaviour of the background scattering processes. This has been crucial to the analysis of some experiments e.g. Cr, where the scattering in out-of-plane detectors, where no magnetic signal was expected, was subtracted from the in-plane detectors at the same scattering angle to leave magnetic scattering that was otherwise far from clear.

2.2 Instrument inprovements

The experience on HET and MARI has shown that there are a number of ways in which a purpose designed spectrometer could be improved to enhance the effectiveness of single crystal experiments.

- **Eliminating gaps in the detector arrays.** In the most common method of performing single crystal experiments it often occurs that in order to observe spin waves at the desired energy transfer the required detector angle corresponds to a gap in the coverage. Additionally, one can in principle construct scans by combining data from different detectors at different energy transfers (for example by integrating over an energy window in each detector a constant-E scan can be simulated). Gaps in the detector array prevents this.

- **Ensuring the same secondary flight path for different detector banks.** On HET the detectors with scattering angle 9°-29° have a 2.5m flight path, whereas those between 3°-7° degrees are at 4m. They have worse resolution as a result, and mostly have not been used in the analysis of single crystal data. In low energy experiments the resolution is acceptable, but the discontinuity in resolution between the low and higher angle banks makes comparison of the data difficult. On MARI, all detectors are at 4m from the sample and the continuity of resolution has been used, for instance with Fe, to extrapolate the multi-phonon scattering at high energy from higher scattering angles back to low scattering angle to perform a background subtraction.

- **Position sensitive detector arrays.** On HET and MARI the ^{3}He gas tubes are 30cm in length. At high energies this leads to poor **Q** resolution along their length (1.5Å^{-1} FWHH for 1eV neutrons). Position sensitive detectors would permit the resolution to be adjusted in software, both during and after the experiment. The out-of-plane detectors are generally used only for background estimates because the detector length integrates along a direction in reciprocal space that is not generally related to the crystal symmetry. It often occurs that their paths in (**Q**,ω)-space intersect the dispersion relation and so cannot be used for this purpose. Position sensitive detectors would allow scans to be constructed in software that made use of out-of-plane scattering just as much as the in-plane scattering. Scans could be constructed either to explore regions where magnetic scattering instensity is expected or to avoid magnetic intensity in order to provide an estimate of the background.

- **Increasing the total detector area.** A larger detector area at low scattering angles will greatly increase the possibility of constructing useful scans in (**Q**,ω)-space. In some experiments the gain is directly quantifiable: the count rate can improve by a factor of 6 over MARI if there is continuous coverage to 30° scattering angle (figure 2 gives an example). Coverage to this scattering angle would be an important gain, especially in low dimensional experiments for which only small samples are available.

- **Increasing the spectrometer flexibility.** Part of the success of the triple axis spectrometer comes from the ability to change collimation and monochromators in order to trade intensity for resolution. On HET and MARI the only flexibility is to change the incident energy and the chopper package. On MAPS the ability to alter the primary and secondary flight paths will permit wider intensity / resolution compromises. Adjustable apertures to control the beam size appear on triple aixs spectrometers to control the background, and these will also be a feature of MAPS.

3 SPECTROMETER SPECIFICATION

The principal spectrometer parameters are summarised in Table 1, along with those of HET and MARI.

3.1 Energy transfer range

The motivation for the proposal to build MAPS is to measure magnetic excitations at energy transfer $100\text{meV} \leq \varepsilon \leq 1000\text{meV}$, and the design of MAPS should be optimised for this range. It is also worthwhile that the MAPS design be tested at lower energies with an eye on instrumentation at proposed high power pulsed sources such as the ESS, so consideration should be made in the design for

$$5\text{meV} \leq \varepsilon \leq 1000\text{meV}$$

with $10\text{meV} \leq E_i \leq 2000\text{meV}$

so long as this does not compromise the efficiency of the spectrometer in its design energy transfer range.

The required energy resolution is that of HET/MARI or better, ie $\Delta\varepsilon/E_i \approx 2\%$ FWHH at the elastic line decreasing to $\Delta\varepsilon/E_i \approx 1\%$ at full energy transfer. The spectrometer flight paths should be flexible enough to permit optimisation of the flux for a given energy resolution.

3.2 Moderator and beamline

It is proposed that MAPS be sited on a beamline viewing an ambient temperature water moderator poisoned at 2cm depth. There is an alternative beamline which views a 77K methane moderator, which will have increased flux at E_i less than about 20meV, but the minimum moderator-chopper flight path is 12m at this site, whereas 7.5m is achievable on the proposed beamline. In the high E_i range for which MAPS is intended the flux from the moderator face is the same in either case.

3.3 Primary spectrometer

Figure 3 is a schematic plan of the MAPS spectrometer. The primary beam components will be heavily shielded modules running on rails, effectively a large optical bench. Spacer modules will be available to alter the distances between the components and the total flight path to the sample. The flight path will be evacuated. The minimum configuration (T=0 chopper, Fermi chopper, 1m collimation from chopper to sample) gives the moderator - sample distance $L_1 \approx$ 9m. The energy resolution of MAPS can be improved to 2/3 of the value for HET or MARI by allowing L_1 to increase to 15m while maintaining the sample-detector distance L_2 = 4m (and hence keeping the maximum detector solid angle - see "Secondary spectrometer" below). The minimum requirement is that the beamline can be reconfigured on a time scale at least as fast as the norm for the current changeover time between experiments at ISIS (4-5hrs) It is highly desirable that reconfiguration is fast enough to be considered routine within an experiment.

- Collimation

The beam at the sample position should be at least 6cm by 6cm, to allow the largest available samples (often 100g - 200g) to be used. There will be several motorised adjustable apertures along the beamline to permit a tightened beam when small samples are used. A soller collimator should be available after the Fermi chopper to tighten the cone of neutrons that are scattered by the chopper body.

- Choppers

MAPS will have a t=0 chopper (ie closed for the fast neutron flash), to be sited as close to the bulk shielding as possible (flight path 6.5m). A second background chopper before the Fermi chopper will be available. This will perform a coarse energy selection but will remain closed for most of the frame to attenuate fast neutrons that penetrate the Fermi chopper in its closed position.

A selection of Fermi choppers will be available suitable for high and low energy resolution configurations of the spectrometer. The design of those currently used on HET and MARI works well for $E_i \leq$ 700meV but above these energies the slats become 'grey'. An improved design using ^{10}B instead of natural boron is currently being explored.

- Monitors

The usual array of beam monitors will be required -

- in the unmonochromated beam before the coarse chopper
- after the Fermi chopper
- behind the sample

It is very important for the monitors to be highly stable. Many single crystal experiments rely on the subtraction of one large signal from another to leave a small magnetic signal.

This can only be performed with confidence if the normalisation of data sets is very accurate. Improved or alternative monitor designs will be explored. Absolute calibration of the monitors is highly desirable so that throughput of components such as choppers and collimation can be readily assessed.

3.4 Secondary spectrometer

It is proposed to have two detector banks:

1. Low angle bank with $0° < \phi \leq 30°$ covering all azimuthal angles with secondary flight path $L_2 = 4$m.
   This will require $11m^2$ of detectors (figure 4). Suitable resolution is 2cm pixelation. The bank could therefore be covered with 1m long position sensitice gas tubes with diameter 2.5cm and with resolution 1/64 of their length. One possible problem with gas tubes is the dead time. If a Bragg's law is satified at one point in a detector, then it becomes inactive along its whole length for $\approx 10\mu s$. The alternative of ZnS detectors is also being investigated.

   At high E_i the angular coverage is rather generous for seeing purely magnetic scattering e.g. at E_i=1000meV $Q = 7.6Å^{-1}$at the elastic line at 20° and $Q=9.1Å^{-1}$ at ε =500meV. However, higher angle data will be important for determining the behaviour of the multi-phonon background. Also, at lower E_i magnetic scattering will be observed in the entire bank e.g. at E_i=100meV $Q = 3.60Å^{-1}$at the elastic line at 30° and $Q=3.64Å^{-1}$ at ε=50meV.

2. (In a future expansion) a high angle bank, with $30° \leq \phi \leq 120°$ (say) with L_2=4m These would be important for studies of phonons on MAPS. They would also be useful in magnetic experiments e.g. in an experiment in Fe on MARI data up to ϕ=75° was used to understand the multi-phonon background, where the E_i used ranged up to about 800meV.

The low angle bank should have variable L_2, increasing up to 8m. With the optimum choice of chopper, this would allow, for instance, L_1 to be shortened to 10m so as to retain the same energy resolution as on HET but double the flux at the sample position. Alternatively, the flux can be maintained but the energy resolution improved to 2/3 of its HET/MARI value. The solid angle coverage will be reduced, but at L_2=8m it will still cover all $\phi < 15°$. The variable flight path can be achieved either by having removable sections beyond L_2=4m, or by mounting the detector array on rails within a SANS type vacuum vessel. In the former case, the detectors should nevertheless be mounted inside the vacuum vessel so as to eliminate the need for a large vacuum window with supporting ribs.

Space considerations restrict the maximum L_2 for the high angle bank to 4m. Depending on cost constraints, the high angle tank could be built as part of the low angle tank or as a unit to be bolted on at a future date.

4. DATA COLLECTION, COMPUTING AND SOFTWARE

4.1 Data Acquisition Electronics (DAE)

In the low angle bank MAPS will have about $10m^2$ of detectors with pixel elements of about 4 cm^2, giving about 25000 spectra, a factor of 50 greater than on MARI. With energy bins corresponding to 1/10 of the resolution ($\Delta\varepsilon/E_i \approx 1\%$) this gives a minimum requirement of 1000 time channels. Typically, on HET and MARI 3000 time channels are used. The total number of data bins will be between 25,000,000 and 75,000,000. A new Data Acquisition Electronics, DAE-II, is currently being designed at RAL, and this has sufficient power to cope with this number of histogram bins.

4.2 Visualisation and data manipulation

Significant computing power and advanced data visualisation methods will be needed to extract the full information from data gathered on MAPS, and must be considered an integral part of the spectrometer. The problem of data visualisation has yet to be tackled, but some points emerge-

- The current procedure for setting up most experiments is to produce a plot of contours of constant ε in the scattering plane of the crystal (figure 5). The reciprocal lattice of the crystal is overlaid so that the accessible ($\mathbf{Q}$,ω) points in any detector can be seen. This plot cannot indicate what is observable in the out-of-plane detectors. MAPS will access a volume of (Qx,Qy,Qz) space (at any point of which ε is determined). A generalised version of the plot is needed in which one chooses a plane in reciprocal space, fixes the third component of $\mathbf{Q}$ and plots contours of constant ε. Alternatively, a value of ε can be chosen, and contours of one component of $\mathbf{Q}$ plotted as a function of the other two (figure 2 earlier is an example of this plot, where ε was fixed at 250meV and contours of constant component of $\mathbf{Q}$ along the rods of scattering plotted on the 2D Cu-O planes).

- Data will be viewed by choosing a plane or direction in ($\mathbf{Q}$,ω)-space and integrating the counts in a resolution volume constructed from many pixels in detector element-timebin space about each point.When the algorithm that determines the resolution function in the plane or along the direction has been specified the data can be converted to S($\mathbf{Q}$,ω). (Many pixels will be needed because the counts in any one will be so few. The gain of MAPS over HET/MARI is that the plane in ($\mathbf{Q}$,ω)-space and the resolution function can be freely chosen). The form of the resolution function will in general be decided in an interactive analysis session, perhaps from a menu or a user-written library, so significant computing power will be required to perform the calculations in real time.

- Calculation of the S(**Q**,ω) data will be within a manipulation and display package. Routine manipulations that can be envisioned include, for instance, adding multi-dimensional data corresponding to symmetry related parts of reciprocal space, and adding runs or subtracting data from runs performed at different temperatures. At ISIS an in-house package, GENIE, is used to manipulate and display 1D workspaces of time-of-flight or processed data. It is an excellent toolbox, as it permits command files containing many GENIE commands to be run and can access external pre-compiled programs to transform workspaces, but to be suitable for MAPS it needs to be improved to handle multi-dimensional data sets. There is a project to write a greatly enhanced version of GENIE at ISIS which would satisfy the requirements of MAPS.

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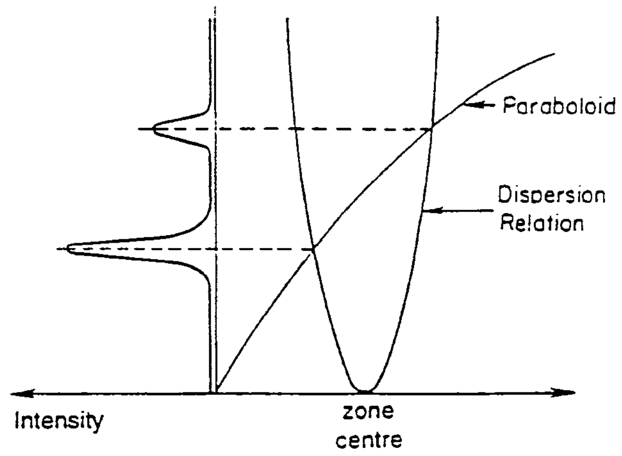
Table 1.

Spectrometer specification and comparison with HET and MARI

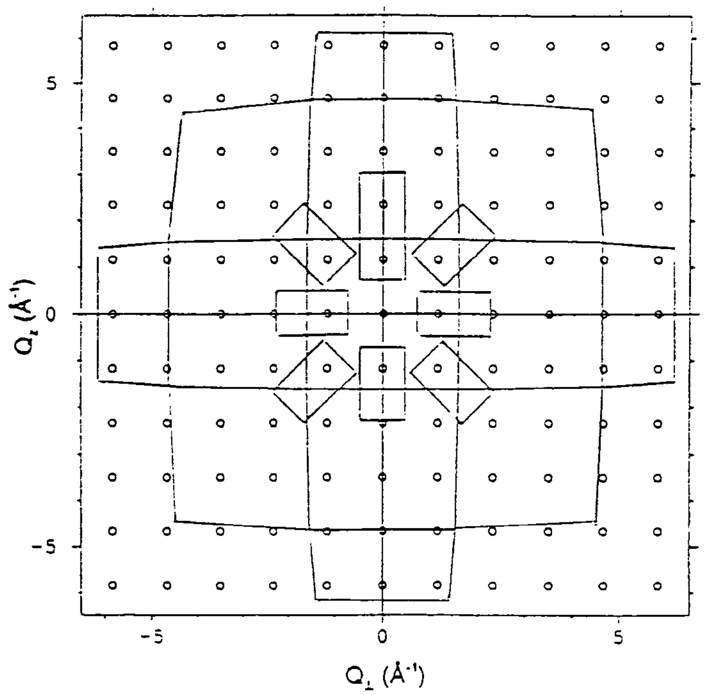
	MAPS	HET/MARI
moderator:	Ambient water (poisoned)	HET: Ambient water (poisoned) MARI: Methane 77K (poisoned)
Energy range:	10meV $\leq E_i \leq$ 2000meV 5meV $\leq \varepsilon \leq$ 1000meV	HET: 10meV $\leq E_i \leq$ 2000meV MARI: 10meV $\leq E_i \leq$ 1000meV
Resolution:	$\Delta\varepsilon / E_i \leq 2\%$ @ elastic posn.	$\Delta\varepsilon / E_i = 2\%$ @ elastic posn.
Choppers	t=0 chopper coarse chopper Fermi chopper	t=0 chopper coarse chopper (MARI) Fermi chopper
Primary flight path L_0	9m $\rightarrow$ 15m	11.8m
Beam size	6cm x 6cm (max.)	HET: 4cm x 4cm MARI: 5cm x 5cm
Secondary flight path L_2	4m $\rightarrow$ 8m	HET: 4m $3.5° \leq \lvert\phi\rvert \leq 7°$ $110° \leq \phi \leq 120°$ 2.5m $9° \leq \lvert\phi\rvert \leq 29°$ $120° \leq \phi \leq 140°$ MARI: 4m $3.5° \leq \lvert\phi\rvert \leq 135°$
Detectors and scattering angle range:		
	10atm. ^{3}He gas tubes 100cm position sensitive	10atm. ^{3}He gas tubes. 30cm long
	$\lvert\phi\rvert \leq 30°$ all azimuthal angles	HET: $3.5° \leq \lvert\phi\rvert \leq 7°$; $9° \leq \lvert\phi\rvert \leq 29°$ MARI: $3.5° \leq \lvert\phi\rvert \leq 15°$
	$30° \rightarrow 120°$ plane array	HET: high angle bank MARI: $15° \leq \lvert\phi\rvert \leq 135°$

Fig. 1

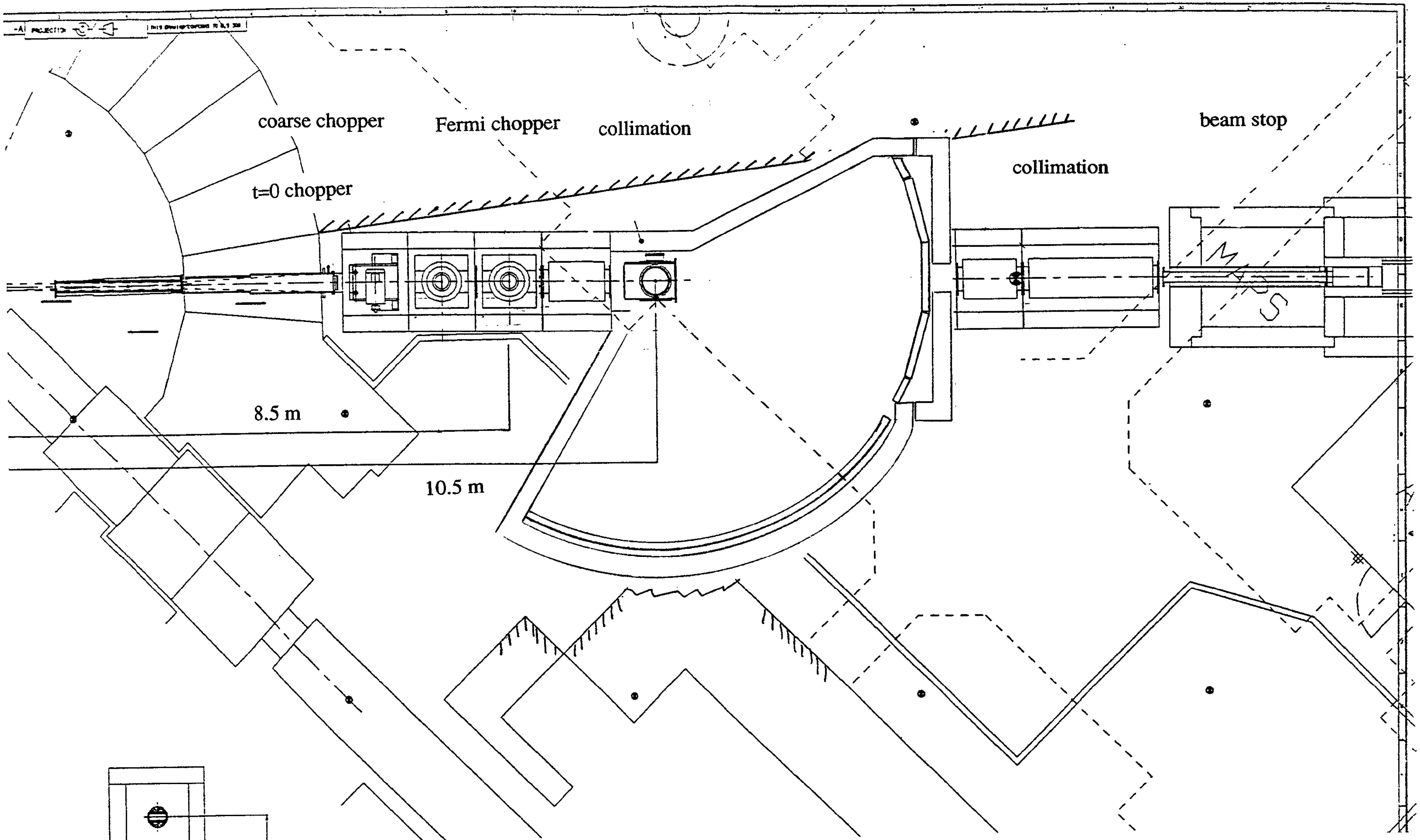


A section through the paraboloid for a detector which accepts K_f that lie along a symmetry direction of a crystal. At those energy transfers where the paraboloid intersects the dispersion relation peaks in scattered intensity are expected.

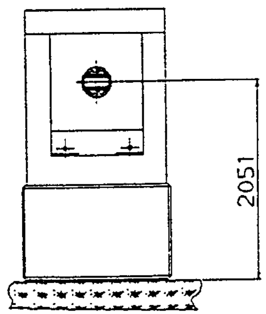
Fig. 2



The relative solid angles of the low angle detectore on MARI and the proposed spectrometer MAPS as viewed along the incident neutron pateh. The momentum transfer scale is set by assuming E_i=650 meV and an energy transfer of 250 meV, approximately the condictions required to observe the zone boundary magnon in $La_{1.85}Ba_{0.15}CuO_4$. The circles mark the zone centres of this material when the 2D axis is aligned along K_i. The number of centres enclosed by the detector areas of MARI and MAPS give an idea of the relative data collection rates for the two instruments



An example MAPS configuration



BEAM CROSS-SECTION

Fig. 3

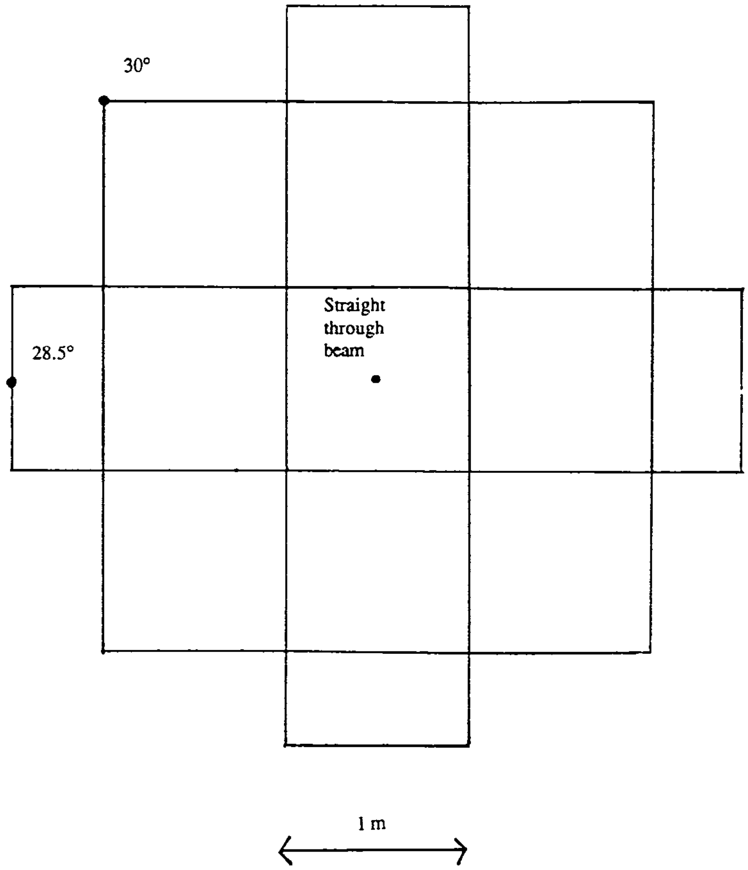


Fig. 4

MAPS low angle detector bank. The scattering angles are shown at the extremities of the bank.

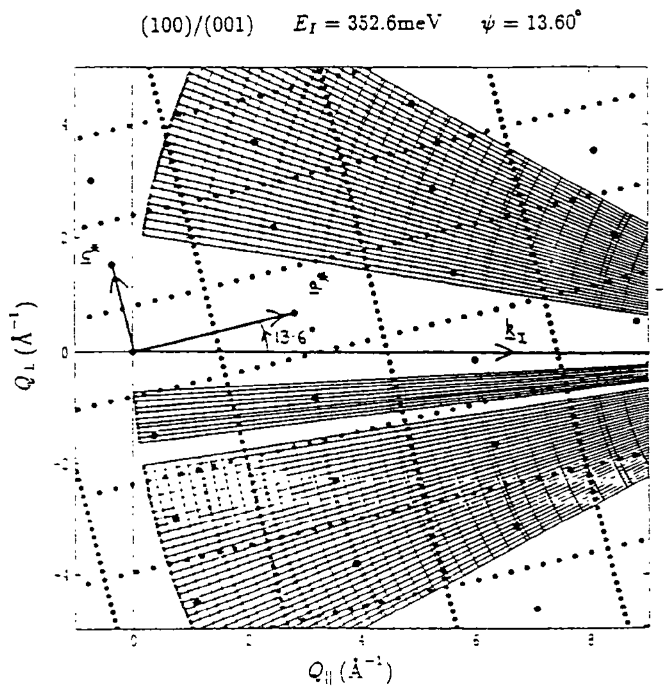


Fig. 5

A plot of the accesible region of (Q,ω)-space in the (100)-(001) plane of h.c.p. cobalt with $\mathbf{a}^*$ at 13.6° to $\mathbf{k}_i$ and E_i=350meV on HET. The radial lines correspond to the locus of vector Q intercepted by different detectors and the circular contours are lines of equal energy transfer in 10meV intervals (50meV for the double lines). The spots mark reciprocal lattice points. The crystal is oriented so that the $\mathbf{a}^*$ symmetry direction through (1,0,-1) lies approximately along $\mathbf{k}_f$ for one of the central low angle detectors.