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FIRST RESULTS FROM THE UK-JAPANESE SPECTROMETER MARI

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ABSTRACT

The commissioning and preliminary scientific results from the MARI spectrometer at ISIS are reviewed.

I. INTRODUCTION

The MARI spectrometer is funded by the Japanese Ministry of Education, Science and Culture (Monbusho) as part of the UK-Japan Collaboration in neutron scattering. This collaboration, initiated in 1986, was the creation of the late Professor Yoshikazu Ishikawa. The name of the spectrometer, MARI, is the Japanese for 'Truth' and also the name of his daughter. It is appropriate that Professor Ishikawa's pivotal role in the UK-Japanese collaboration should be remembered in this way. The spectrometer was inaugurated by his widow, Mrs Hiroko Ishikawa, in a moving ceremony at the Rutherford Appleton Laboratory on 16 November 1990.

Figure 1. Dr Paul Williams (Director RAL), Mrs Hiroko Ishikawa and Sir Mark Richimond (Chairman SERC) at the MARI Inauguration.

The design concept (see Figure 2) is based on the highly successful HET spectrometer at ISIS and shares much of its technology. A 30 cm rotating nimonic chopper is used to attenuate fast neutrons produced at the moment when the protons hit the target. Subsequently, a monochromatic beam is produced by phasing a high speed Fermi chopper to the pulsed source. A wide variety of choppers (interchangeable with HET) are available with characteristics optimised for incident energies from 20 to 2000 meV. The beam at the sample position is 50 x 50 mm^2 with an intrinsic resolution of 1%. The vertical detector array is divided into a low angle bank and eight higher angle windows which together give an extensive and continuous angular coverage from 3° to 135°. The low angle bank (3 -12°) has a complement of 128 high pressure helium detectors in an octagonal array. The higher angle windows have three strips each containing 32 detectors arranged where appropriate on the Debye-Scherrer cone. Currently the full low angle array and the central strip of each window are installed. The highest angle window (120-135°) has its full complement of 96 detectors. Further detectors will be obtained as funds become available.

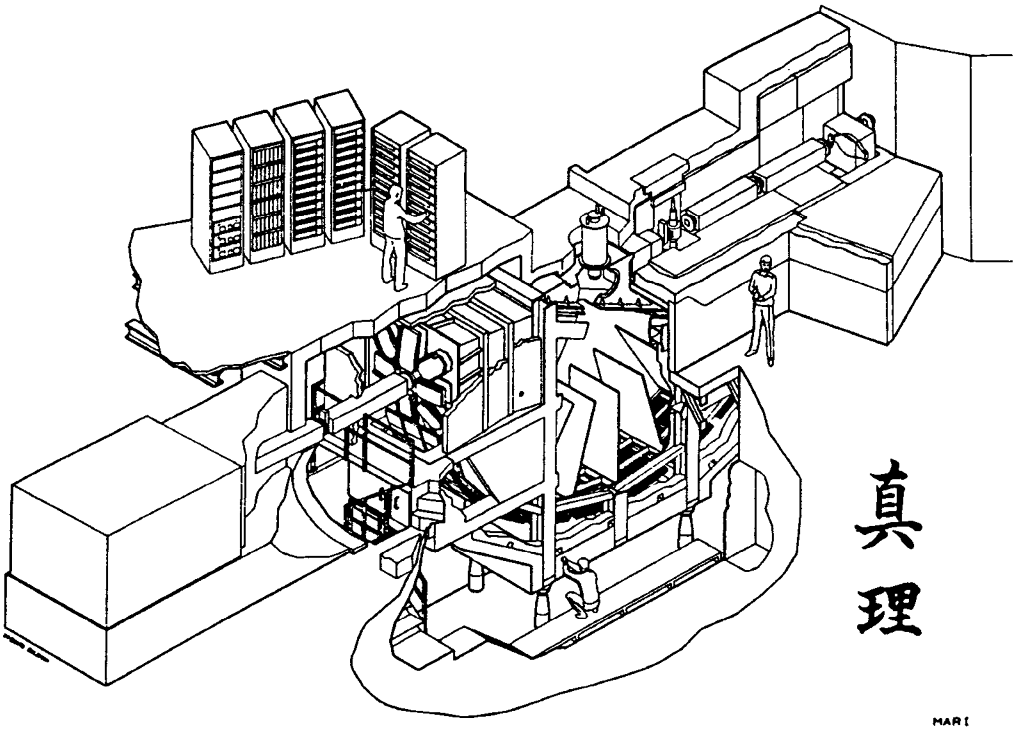


Figure 2. Schematic layout of the MARI spectrometer

After completion of the installation late in 1989, a hectic three-cycle commissioning phase began. Initial backgrounds were soon reduced to the levels attained on HET and the characteristics of MARI were thoroughly explored. This phase has been highly successful, due to the dedication and hard work of the excellent MARI team, and the spectrometer was made available for scheduling for the latter half of 1990. Proposals were considered by three Experiment Selection Panels (ESPs): Liquids and Amorphous Systems, Excitations and Momentum Distributions, and Molecular Spectroscopy, reflecting the broad appeal of the latest ISIS spectrometer. The ESPs showed excellent judgement in selected a broad and balanced programme for an already significantly oversubscribed instrument. The first six months of the experimental programme have borne out the fact that MARI is a world class instrument, destined to make a significant impact in condensed matter science.

II. EARLY RESULTS

The advantages of having MARI view the 100 K liquid methane moderator were confirmed. The monochromatic lineshape is significantly improved in the 50 -150 meV region and incident energies down to 20 meV are now possible with good flux. Such low incident energies provide excellent absolute resolution for studying low energy excitations such as the crystal field transitions in doped superconductors. The pairing mechanism in these ceramic superconductors has been explored by studying the temperature dependence of the linewidths of these transitions, Figures 3(a) and 3(b).

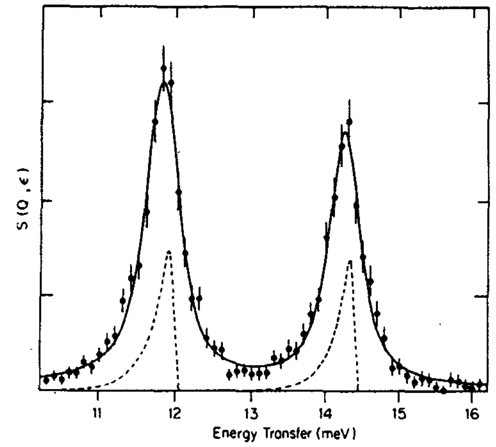


Figure 3(a). Γ_4 and Γ_2 transitions in Tm:$YBa_2Cu_3O_{7-x}$, measured at 20K on MARI. The dashed line represents the instrument resolution.

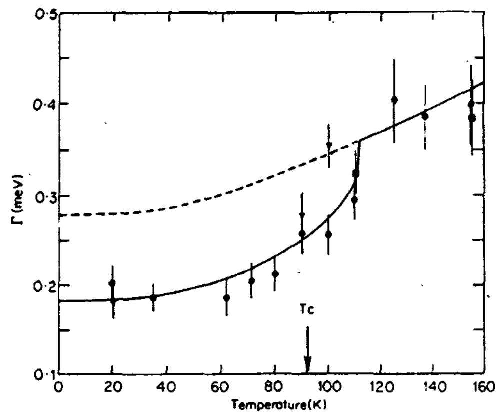


Figure 3(b). Variation of linewidth with temperature of the Γ_4 transition in Tm:$YBa_2Cu_3O_{7-x}$.

The advantage of the wide and continuous angular cover available on MARI is well illustrated by a commissioning experiment on helium. Final state interactions in Bose condensed ^{4}He provide a severe test of our understanding of this simple fluid. These manifest themselves as an oscillatory behaviour in the constant Q width of the dynamical structure factor S(Q,ε) and have been observed by triple-axis spectroscopy, but with poor resolution. Previous measurements using the high angle bank on HET required a different incident energy for each Q. Although the resolution was excellent, there was some difficulty in interpolating to constant Q from the limited available angular range and since many energies were required, statistics were poor for any given data set. A single measurement on MARI using an incident energy of 250 meV provided an S(Q,ε) surface (Figure 4(a)) from which constant Q data ranging from 4 to 16 $Å^{-1}$ have been obtained with high accuracy, in which the oscillatory behaviour can clearly be observed (Figure 4(b)).

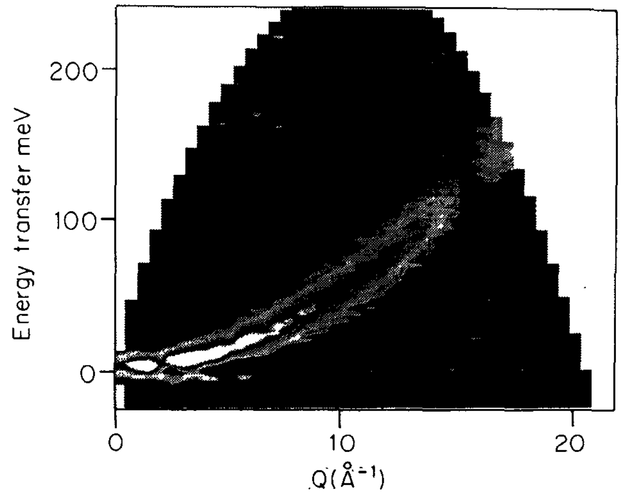


Figure 4(a). The S(Q,ε) surface for He at 1.5K accessible with 250 meV neutrons.

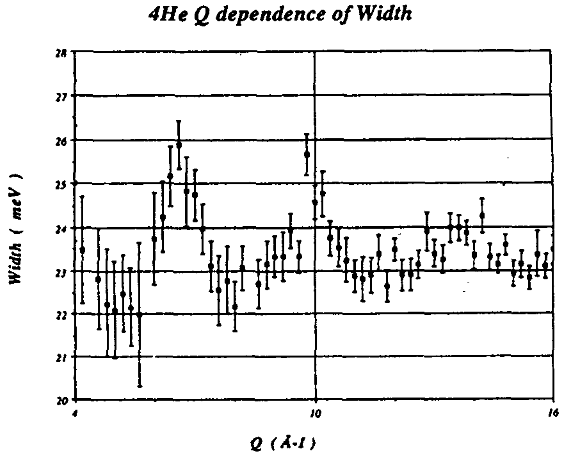


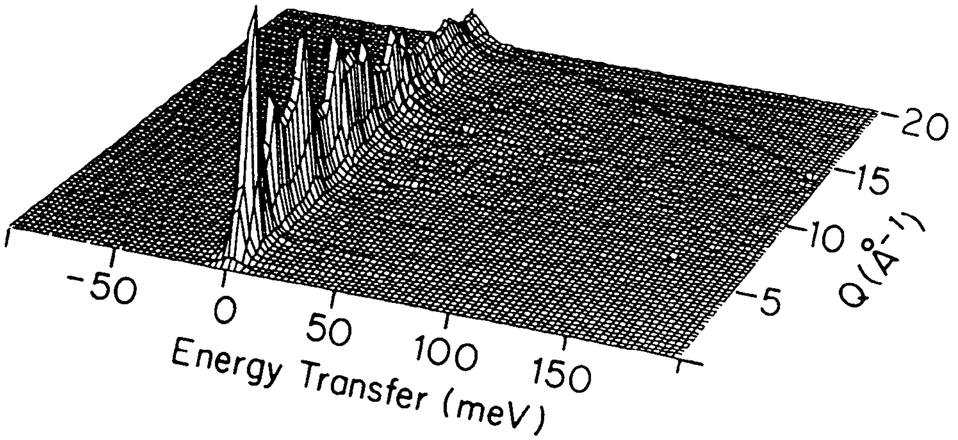
Figure 4(b). Oscillations in the width of the structure factor in He at 1.5K.

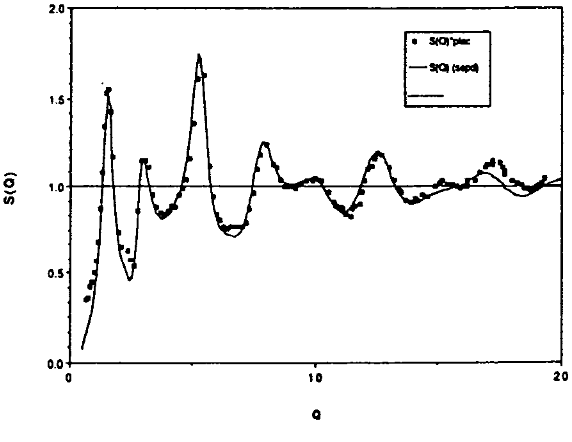
III. RANDOM NETWORK GLASSES

In the absence of a regular lattice, the dynamical structure factor gives unique insight into the correlations between structure and dynamics in glassy materials. Pioneering studies at Argonne National Laboratory's IPNS facility have until now represented the state of the art in this field. The superiority of MARI in resolution and intensity should provide high quality data which may prompt new extensions of the theory and aid a better microscopic description of non-crystalline materials.

During commissioning, a preliminary $S(Q,\epsilon)$ measurement was made on the tetrahedral oxide glass SiO_2. These data, obtained in only a day, are shown in Figure 5(a) on an absolute scale. Integration over energy at constant momentum transfer gives the static structure factor S(Q) and a good check of the quality of the data through this zeroth sum rule, Figure 5(b). The average vibrational density of states obtained from these data are also shown, in Figure 5(c). Fine structure is seen around 50 and 150 meV, suggesting the existence of the saturated non-bridging oxygen on the cluster surface. These results bode well for future high statistics experiments which will probe the Q dependence of these excitations and lead to a sophisticated discussion of the microscopic behaviour of such random network glasses.

Figure 5(a). The $S(Q,\epsilon)$ surface for SiO_2.





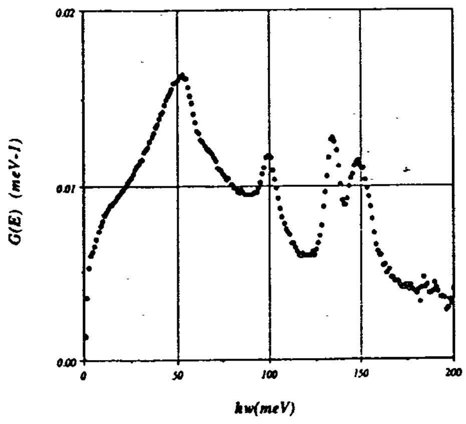


Figure 5(b). S(Q) for SiO_2.

Figure 5(c). Density of states of $G(\epsilon)$for SiO_2.

IV. LOCAL MODES IN LUH

MARI looks certain to have a significant impact on many aspects of vibrational spectroscopy including the study of the local modes in metal hydrides. Until now, these studies have been constrained as both HET and TFXA offer only a limited Q range at any one spectrometer setting. MARI has the momentum transfer range to follow accurately the rise and fall of intensity due to the Debye–Waller factor and hence to help determine the assignment of observed features. In rare earth – hydrogen systems such as $LuH_{0.1}$, the effects of the hexagonal lattice anisotropy on short range order and the vibrational spectrum of hydrogen can now be studied. Figure 6 gives data from MARI and indicates the more limited range available on HET and TFXA.

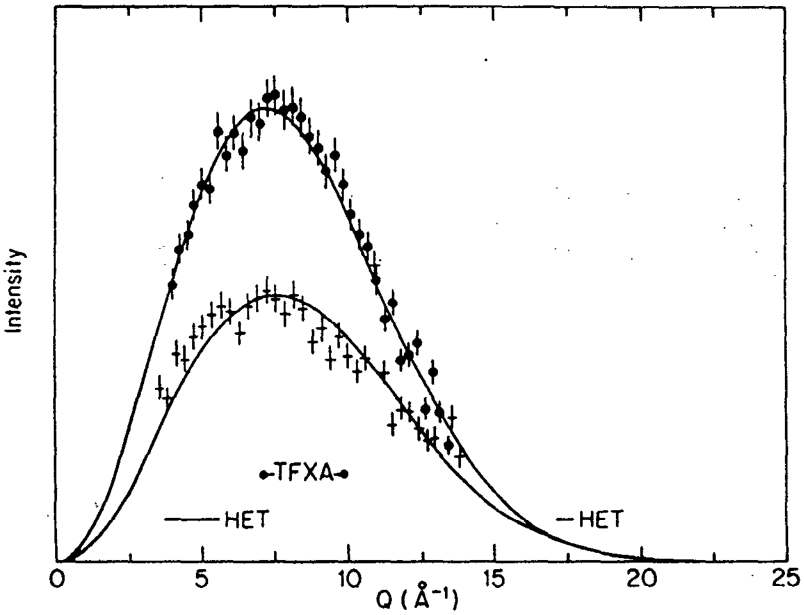


Figure 6. The variation of intensity with Q for the 100 meV and 134 meV modes in $LuH_{0.1}$. The more restricted range of both HET and TFXA are indicated.

V. ANTIFERROMAGNETISM

The potential of time–of–flight spectrometers for the study of excitations in low dimensional systems has been well illustrated by the first MARI experiment on single crystals. The covalent chains of iron–centered, edge–sharing sulphur tetrahedra in $KFeS_2$ order antiferromagnetically in one dimension. Earlier magnetic measurements implied that the spin–wave spectrum would be very steep. This was indeed found to be the case with the spin wave dispersion extending to more than 200 meV, Figure 7. The one dimensional nature of the problem means that all of the detectors in the MARI low angle bank can be summed together. The entire dispersion surface was mapped out using several incident energies in only a few days.

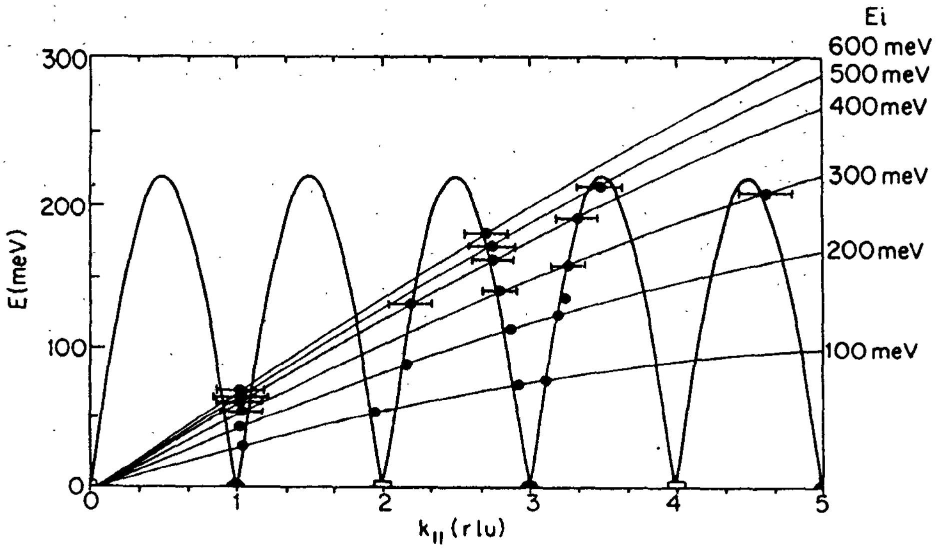


Figure 7. The dispersion of the antiferromagnetic spin waves in the quasi–one–dimensional system $KFeS_2$. For a crystal of volume 3 cm^3, each scan was collected in 1 mA-hr.

In the two-dimensional S = 1 antiferromagnet La_2NiO_4, well defined spin waves were observed to energies above 100 meV (Figure 8). With the appropriate crystal orientation, the two dimensional nature of the scattering could be exploited to great effect by directing the rod-like scattering into the horizontal bank. These detectors were summed to give the signal and simultaneously the vertical part of the array could be summed to give a measure of background.

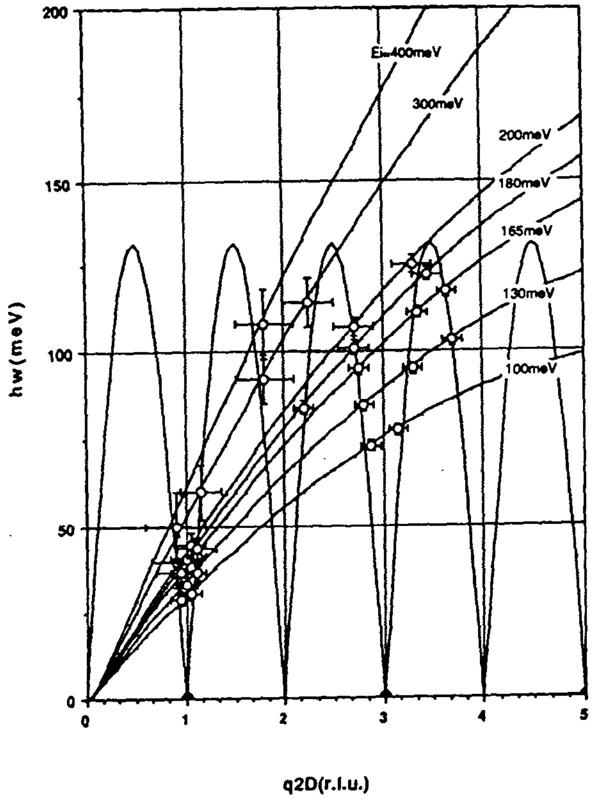


Figure 8. The dispersion of the antiferromagnetic spin waves in the two dimensional system La_2NiO_4.

Acknowledgement

We would like to thank all the support staff at ISIS for their excellent contributions to the MARI project.

Q(N.Watanabe): What was the major difference in resolution between HET and MARI for measuring crystal field line broadening in Tm 1-2-3 compound? I think there is no difference in moderator pulse width for E_i~25 meV.
Was there any advance in scientific results with MARI compared to HET?

A(A.D.Taylor): At low energies, the resolution is comparable. The large solid angle at low angles on MARI provides a significant intensity increase, however, on MARI. At this time, HET was fully scheduled: available for test experiments.

Q(R.Pynn): What type of detectors do you use on MARI?

A(A.D.Taylor): 2.5cm diameter, 30cm long 10atm, ^{3}He from Reuter-Stokes.

Q(R.Pynn): How does MARI compare with IN4 & IN1?

A(A.D.Taylor): IN4 always—will always win—if all you care about is intensity. For resolution, MARI is competitive down to, say, 20 meV. If you can use IN1, do so. If not (because of energy, resolution, S/N) use HET / MARI. For 1 and 2D systems, MARI is very effective even at 50 meV.

Q(H.Tietze): Does the vertical scattering plane of MARI have any influence on the resolution and performance of the instrument with respect to an assumed horizontal scattering plane?

A(A.D.Taylor): No basic influence on resolution, however, worse background from top in lighting and sample environment, which is in general designed vertical for horizontal scattering plane.