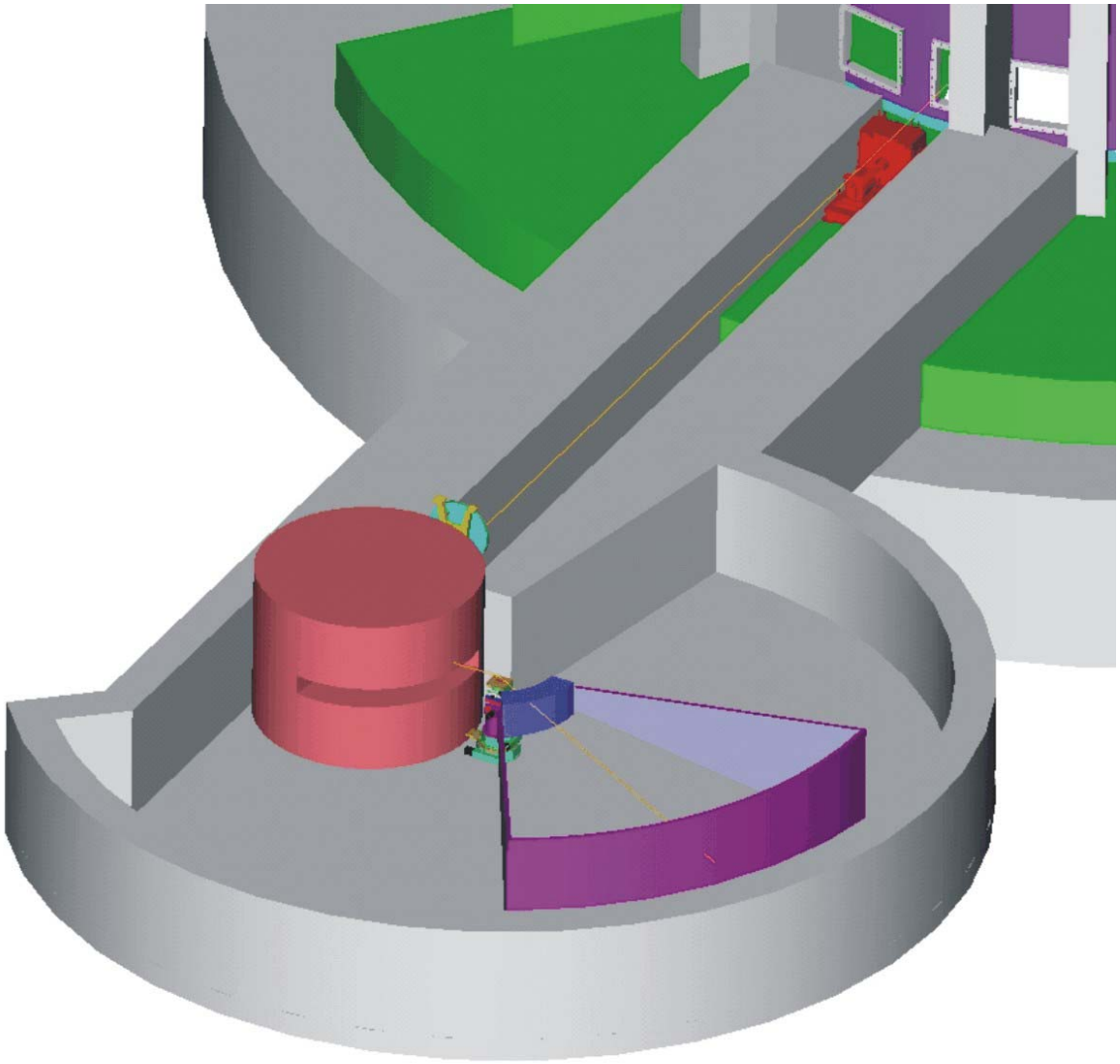
HYSPEC: A Crystal-Time-of-Flight Hybrid Spectrometer for the Spallation Neutron Source



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EXECUTIVE SUMMARY:

The study of phase transitions and novel ordered phases remains at the forefront of condensed-matter research. In studies of superconductivity, magnetism, ferroelectricity, colossal magnetoresistance, charge order, etc., one is interested in determining how each type of order occurs, including how and why it arises from the disordered state. The energy scale for excitations that have an impact on ordering is typically on the order of a few to tens of meV; this is the same energy scale for excitations that impact transport properties. The intensities and energy resolutions obtainable with thermal neutrons are ideal for such inelastic studies. At the same time, one needs to be able to detect and monitor the relevant order parameter through elastic diffraction measurements, which frequently involves measurements of superlattice peaks that may be extremely weak. Often one wishes to study correlations that have not achieved long-range order, in which case one must measure diffuse scattering and be able to discriminate elastic from inelastic contributions. In many cases understanding the order of interest requires studying how it is modified or induced by an extreme environment, such as a strong magnetic field, very low temperature, or high pressure; this is best accomplished by decoupling the sample environment from the detector system. In order to exploit the full power of neutron scattering to distinguish magnetic from nuclear scattering, it is necessary to implement polarization analysis, with polarization sensitivity in both the incident and scattered beams. Finally, in order to discover new phenomena, and to not simply characterize phenomena that have been discovered at other facilities, one has to be able to work with small crystals, thus requiring that a spectrometer focus as much flux as possible into a small spot size.

Up to now, the combination of requirements described above has been met only by triple-axis spectrometers at steady-state sources. Here we present the case that by combining time-of-flight spectroscopy with Bragg focusing optics and the enhanced flux of the pulsed beam that will be available at the SNS, one can build an instrument that not only satisfies our needs, but which will also significantly exceed the performance of the best triple-axis spectrometers at the best steady-state sources. The instrument that we propose, HYSPEC, fills a niche of dramatic importance to exciting areas of condensed-matter physics, one that is not competitively covered by any of the other SNS instruments that have been considered to date.

HYSPEC will be positioned on beam line 15, which looks at a coupled, supercritical H_2 moderator. To transport the beam from the moderator to the sample the instrument will have a straight, 21.8 m long, m=3 supermirror guide that will incorporate T_0, frame overlap and order suppressing choppers along its length and, near its downstream end, a counter rotating chopper pair that will serve to define the neutron burst width and incident energy. A short distance downstream of the counter-rotating chopper pair, the monochromatic beam will impinge on either a pyrolytic graphite or (for polarized beam

studies) Heusler alloy vertical focusing crystal mounted in a rotating drum shield. Because vertical focusing by crystals is extremely efficient, the 4(w) x 15 (h) cm^2 beam exiting the guide will be reduced to area as small as 4(w) x 2(h) cm^2 thus maximizing the flux at the sample position. A series of collimators and beam definers will be placed immediately upstream of the sample and a set of radial collimators in the detector bank just downstream of the sample. In the polarization analysis mode the radial collimator would be replaced by a set of 19 Fe-Si supermirror benders that would make it possible to measure simultaneously both spin-flip and non-spin-flip processes. Detection of the sample scattered neutrons will be done by 188 ^3He position-sensitive detectors housed in a moveable detector bank 4.5 m from the sample that will cover a horizontal angular range of 60° and a vertical range of 15°.

HYSPEC's flux-on-sample was compared to the other planned inelastic instruments CNCS, ARCS and HRCS (which have overlapping energy ranges and resolutions) using the MCSTAS Monte Carlo simulation program. For HYSPEC, the maximum flux on sample was found to be 1.1 x 10^7 n/cm^2-sec at 15 meV with a 3% energy resolution. This is a factor of two more than the maximum flux of CNCS, which is at 5 meV, and about ten times more than maximum flux of HRCS and ARCS, which is around 100 meV. This high sample flux combined with TOF analysis and the wide angular acceptance of the detector array means that a given scattered neutron spectrum will be collected in an order of magnitude less time than on existing single crystal sample instruments.

The easy adaptability of HYSPEC to polarization analysis is unique among the suite of SNS inelastic instruments. To produce a focused, polarized beam the PG crystal is simply replaced by a vertical focusing Heusler alloy crystal. Analysis of the polarization of the scattered neutrons will be done with a set of 19 broad-spectrum Fe-Si supermirror benders downstream of the sample: a well-established and extremely reliable technology. Because the sample-to-detector distance is large, the spin-flip and non-spin-flip parts of the scattering will fall on different groups of detectors and can thus be measured simultaneously.

Finally, it should be noted that an SNS Inelastic Neutron Scattering Workshop held on 11/1/99 at ANL [SNS Document: IS-1.1.8.2-8004-MM-A-00] recommended that a spectrometer with almost identical characteristics to HYSPEC be viewed as a "potential "day-one" instrument".

HYSPEC: A Crystal-Time-of-Flight Hybrid Spectrometer for the Spallation Neutron Source

1. INTRODUCTION

This document lays out a proposal by the Instrument Development Team (IDT) composed of scientists from leading US Universities and National Laboratories to design and build a conceptually new high-flux inelastic neutron spectrometer at the pulsed Spallation Neutron Source (SNS) at Oak Ridge. This instrument is intended to supply users of the SNS, and scientific community represented by the IDT members in particular, with a platform for ground-breaking investigations of the low-energy atomic-scale dynamical properties of crystalline solids. It is also planned that it will be equipped with a polarization analysis capability, therefore becoming the first polarized beam spectrometer in the SNS instrument suite, and the first successful polarized beam inelastic instrument at a pulsed spallation source in general.

The proposed instrument is designed primarily for inelastic and elastic neutron spectroscopy of single crystals. In fact, the most informative neutron scattering studies of the dynamical properties of solids nearly always require single crystal samples, and they are almost invariably flux-limited. In addition, in measurements with polarization analysis the available flux is further reduced through selection of the particular neutron polarization, which puts even more stringent limits on the feasibility of a particular experiment. To date, these investigations have mostly been carried out on crystal spectrometers at high-flux reactors, which usually employ focusing Bragg optics to concentrate the neutron beam on a typically small sample. Construction of the high-luminosity spallation neutron source at Oak Ridge, which will provide intense pulsed neutron beams with time-averaged fluxes equal to those at medium-flux reactors, opens entirely new opportunities for single crystal neutron spectroscopy. Drawing upon experience acquired during decades of studies with both crystal and time-of-flight (TOF) spectrometers, the IDT has developed a conceptual design for a focused-beam, hybrid time-of-flight instrument with crystal monochromator for the SNS called HYSPEC (an acronym for hybrid spectrometer). The proposed instrument has a potential to collect data more than an order of magnitude faster than existing steady-source spectrometers over a wide range of energy transfer ($\hbar\omega$) and momentum transfer ($\boldsymbol{Q}$) space, and will transform the way that data in the inelastic single crystal spectroscopy is collected. HYSPEC is optimized to provide the highest neutron flux on sample at a good-to-moderate energy resolution in thermal neutron energy range. By providing flux on sample several times higher than other inelastic instruments currently previewed for the SNS, the proposed instrument will indeed allow unique ground-breaking measurements, and will ultimately make polarized beam studies at a pulsed spallation source a realistic possibility.

Even though the polarized beam option was not considered at the time, a spectrometer with performance characteristics similar to those of HYSPEC was identified as one of the potential "day-one" inelastic instruments for the SNS at the Inelastic Neutron Scattering Workshop[1] organized by the SNS at Oak Ridge in 1999. It was included as such in the Workshop recommendations, along with six other instruments, four of which are

[1] Report on the SNS Inelastic Neutron Scattering Workshop, SNS Document IS-1.1.8.2-8004-MM-A-00 (2000).

currently approved, and three have already been funded and are being constructed. This proposal is a request by the IDT for SNS approval of HYSPEC as one of the instruments for construction, and for the assignment of a beamline 15 served by a coupled, 20 K supercritical hydrogen moderator to the proposed spectrometer.

2. SCIENTIFIC OBJECTIVES

Much of our current understanding of atomic-scale structure and the dynamical properties of solids and liquids was gained by virtue of neutron scattering studies. Inelastic neutron spectroscopy provided physicists with an unprecedented, detailed access to phonon dispersions, magnetic excitation spectra, soft-modes and critical dynamics at phase transitions, unrivaled by other experimental techniques. Because the neutron only interacts very weakly with matter, it is practically a non-perturbing probe of the matter's inner structure and dynamics, not sensitive to charges and surface layers. Therefore, unlike techniques where the photon electric field, or charged particles (eg electrons, muons), which significantly modify local electronic environment, are used, neutron spectroscopy allows determination of the intrinsic, un-perturbed physical properties of materials. By the same virtue, the neutron is a highly penetrating and non-destructive probe, allowing the investigation of microscopic properties of bulk materials (and not just their surface layers), studies of samples embedded in a complex sample environment, such as cryostats, magnets, pressure cells, etc. In fact, ability to accept a variety of devices creating extreme sample environment was one of the key factors that determined tremendous success of the modern reactor-based neutron spectrometers. Finally, proper manipulation with neutron spin quantum number using various beam-polarizing devices gave an unrivaled opportunity to separate the structural and magnetic phenomena on the atomic scale. Incorporating and optimizing these unique features of the neutron scattering technique in design of a neutron spectrometer is extremely important.

The discovery of new materials and novel unexpected phenomena along with advent of new experimental techniques invariably lead to major advances in condensed matter science. One example is the discovery of strongly correlated electron systems, from heavy-fermions to high-T_c superconductors to manganites exhibiting giant magneto-resistance effects, which has added a new class of materials to those with which we are familiar. They presented new types of macroscopic behavior with a strong potential for future technological applications, which require detailed understanding on the microscopic level. The neutron scattering studies which are indispensable for such understanding, represent a significant experimental challenge. First of all, new materials are usually only available in small quantities. Secondly, the scattering intensities associated with the interesting features in the electronic structure are often intrinsically small. Finally, because several contributions to the microscopic electronic Hamiltonian in these systems are of equal importance for determining their macroscopic bulk properties, direct discrimination between the magnetic scattering by electronic spin and structural/vibrational scattering by the means of polarization analysis is vital.

The goal of the present IDT was to design an instrument which, using the unique, high-flux, pulsed neutron beam at the SNS, would meet the challenge posed for neutron inelastic spectroscopy by the new scientific discoveries and the breakthroughs in the

synthesis of the new materials. There are an extremely large number of problems that are currently at the forefront of condensed matter physics and demonstrate the need for an instrument with the unique capabilities of HYSPEC. Because of space limitations we describe some of these in Appendix. They are intended to be illustrative; there could be many more!

3. THE HYSPEC SPECTROMETER

Our primary objective in formulating the design of the HYSPEC spectrometer was that it delivers the highest possible monochromatic flux to few-cm-sized samples over a broad range of thermal and sub-thermal neutron energies [5,90] meV.

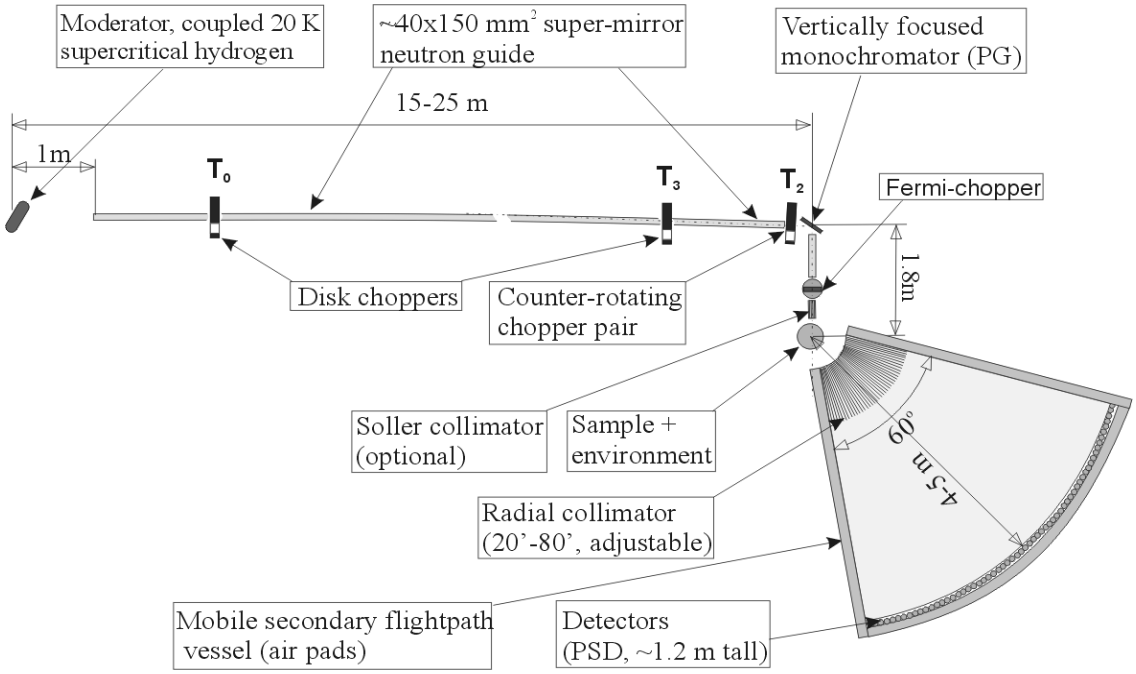


Figure 1 Schematic showing HYSPEC's principle elements.

Additionally, we wanted to develop a design that would (i) minimize beam-related background such as that arising from sample environments, (ii) provide reasonably good (and easily adjustable) energy and scattering vector resolution ($\delta E/E \sim 0.02$ - 0.15; $\delta Q/Q \sim 0.005$ - 0.1), (iii) be readily adapted to polarization analysis, (iv) permit rapid alignment of samples and easy installation of specialized sample environments and (v) allow for straightforward, direct, on-line monitoring and analysis of incoming data. Ultimately these criteria led us to the direct geometry, hybrid concept shown schematically in Fig. 1 and in a 3-D view on the cover. The instrument main design parameters are summarized in Table I.

Table I. Instrument Parameters

Moderator	Coupled, 20K, supercritical H_2
Incident energy	5 - 90 meV

Energy resolution	0.02 <ΔE/E <0.15 (for elastic scattering) depending on neutron energy and rotation rates of choppers
Q resolution	ΔQ/Q ~ 0.005 - 0.1
Primary flight path	Guide with expander and compressor sections 15-25 m long
Secondary flight path	4.5 m
Energy defining choppers	Counter-rotating disk and Fermi choppers. Maximum rotation rate 300Hz
Frame-overlap/order suppressor choppers	Disk choppers. Maximum rotation rate 60 Hz
Flux focusing crystal	Segmeted, vertically curved PG and fluorinated mica. Heusler for polarized beam
Sample position	1.8 m from crystal
Beam size at sample (optimally focused)	4 (w) x 2 (h) cm^2
Detectors	188 2.5cm diameter position sensitive ^{3}He tubes. Horizontal pixel resolution 20 min. Vertical pixel resolution 20 min. Horizontal array acceptance: 60°. Vertical array acceptance: ±7.5°
Sample environment	Will accept all standard sample environment equipment

3.1 Primary spectrometer (Monochromator)

Incident neutron energy will be defined by the time of flight in the primary spectrometer. Reflection from the monochromator crystal in most cases will only serve to focus the beam on sample. In addition, the monochromator will serve in place of the pulse-shaping chopper, cutting the unwanted high-energy tail of the spectral distribution of the incident neutrons, as shown in the Figure 2. Primary spectrometer is envisioned as consisting of a 21.8 meter long guide with a center section composed of 40 mm wide by 150 mm high, supermirror-coated, modules and with 40 mm wide expander sections at each end, Figure 1. Whether the guide will be straight or slightly curved is still an open question that will ultimately be resolved by detailed shielding studies. Placed at intervals along the guide would be three disk choppers; the T_0 and frame-overlap choppers - rotating at either the source frequency or a sub-multiple, and a counter-rotating disk chopper pair, rotating at integral multiples of the source frequency. At the guide's downstream end we plan to place a rotating drum shield containing a vertical-focusing crystal, a beam stop (or, if a straight guide is used, a "get lost" pipe), a vertically tapered guide, a Fermi chopper and a Soller collimator. Attached to the shield - and moving with it – would be a conventional sample rotation stage mounted on air pads, with a 2-axes goniometer capable of supporting large, off-center loads. In such an arrangement, neutron energies are determined both by the beam exit angle (as defined by the crystal

and the in-shield collimator), and by the phase of rotation of the counter-rotating chopper pair and/or the Fermi chopper relative to the source. Apart from its energy defining function, the crystal - together with the section of vertically tapered supermirror guide in the drum shield – also has the important function of efficiently focusing the beam at the sample position.

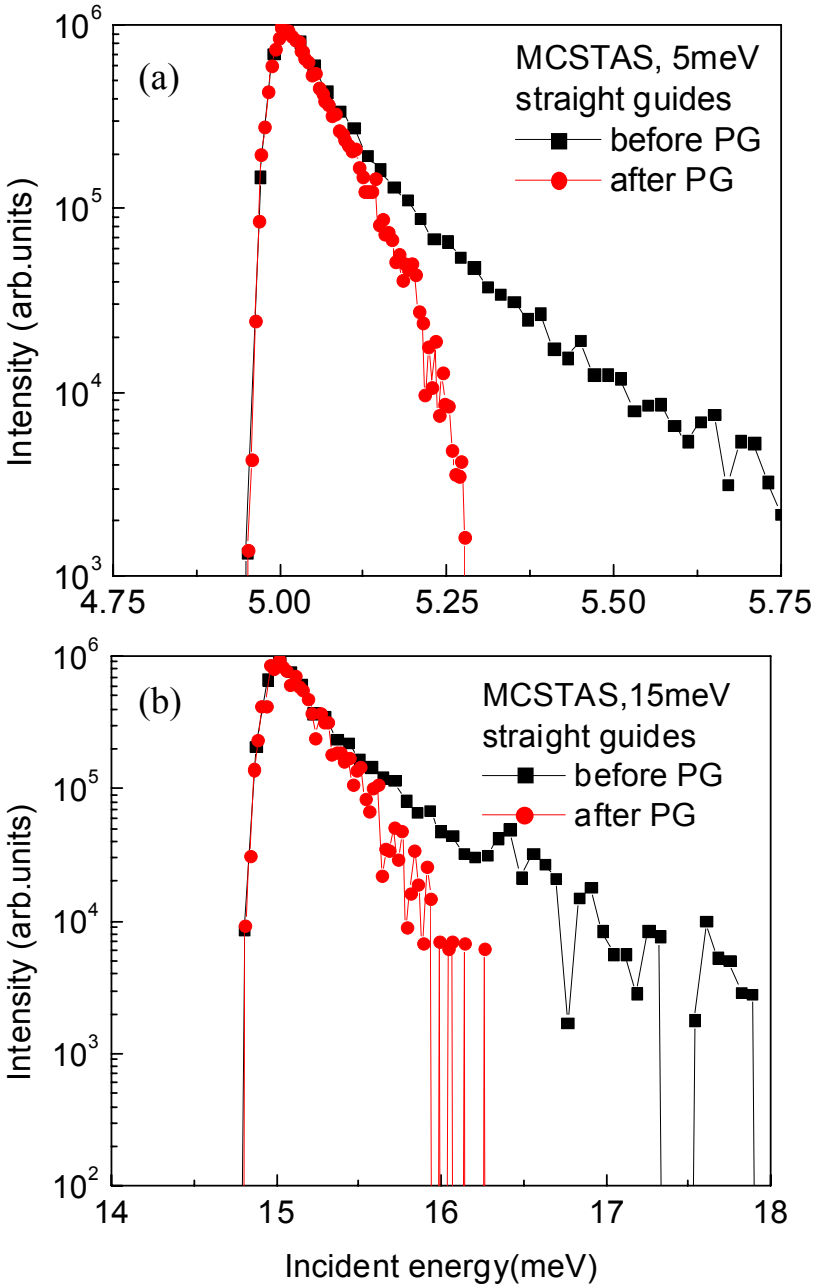


Figure 2. Incident neutron spectrum before and after reflection from a PG monochromator; (a) E_i = 5 meV; (b) E_i = 15 meV.

Each chopper has a unique role in defining the monochromatic incident beam. The T_0 chopper blocks the gamma rays and high energy neutrons emitted in the earliest

part of the source pulse. The frame overlap chopper (T_3) assures that unwanted, higher order (shorter wavelength) neutrons are removed. Also, its rotation rate can be reduced when necessary to block alternate source pulses in cases where the scattered energy spectrum is so broad that spectral overlap becomes a problem. In the main, medium-resolution, high-data-collection-rate mode of operation, the counter-rotating disk choppers T_2 (together with the crystal) will define both the energy of the beam and the spectral and time width of the pulse at the sample position, the latter being the main factor in determining the energy resolution of the time-of-flight analyzing section. In those cases where higher energy resolution is needed, it is envisioned that the time width of the sample pulse would be further reduced by the addition of a Fermi chopper (T_4). This chopper would have short, straight slots and a vertical axis of rotation and be located in the shield immediately upstream of the collimator. Like the counter-rotating disk choppers, it would rotate at integral multiples of the source frequency.

For the focusing crystal, we propose using (for non-polarized applications involving incident neutron energies of 5 meV and higher) individual pyrolytic graphite (PG) plates attached to a segmented GMI-type holder. This arrangement will provide optimal vertical focusing over the entire 5-90 meV neutron energy range. Because a relatively broad horizontal mosaic (probably 1.0^{o}) will be needed for optimum performance, we plan to use sets of three ZYA grade PG plates stacked with spacers to slightly offset the angles between them. This so-called "fanned arrangement" has been demonstrated to expand the horizontal mosaic but leaves the vertical mosaic (and thus the vertical focusing properties of the crystal) unchanged. At incident neutron energies below 5 meV, where high monochromator scattering angles hamper measurements because of the excessively tight resolution introduced by the PG crystal in a near backscattering geometry and space restrictions, we envision substituting an equivalent crystal composed of fanned plates of fluorinated synthetic mica.

3.2 Sample Stage

As emphasized in the Scientific Objectives Section, it is important for many of the areas of research for which HYSPEC is intended that specialized sample environments can be easily and quickly installed. Because the collimation upstream and downstream of the sample will restrict the horizontal field of view of the detectors to the sample area alone, the HYSPEC sample axis will not have to be part of the instrument vacuum system as in other planned inelastic instruments. Thus we expect to be able to employ conventional cryostats, magnets, furnaces, pressure cells, etc. without creating any significant, sample- environment-related background problems. This is another unique feature of HYSPEC.

3.3 Secondary Spectrometer (Analyzer/Detector)

Scattered neutron energy analysis would be done by time-of-flight alone. We propose using a 4.5 meter radius array made up of 188 one-dimensional, position-sensitive, tube-type ^{3}He detectors 2.5 cm in diameter and 1.28 meters long, centered at the sample position. A set of radial collimators (with horizontal angular acceptances of 20, 40, and 60 minutes) interposed between the sample and detectors would restrict their horizontal field of view to the sample area alone. Individual horizontal and vertical pixel

resolutions are envisioned as being respectively 20 and 20 minutes of arc. Additionally, the array would span a 60 degree arc horizontally and have a vertical acceptance of ± 7.5 degrees with respect to the scattering plane. It would be mounted on air pads on a "tanzboden" so that it could both move with the drum shield and be rotated about the sample axis to permit accurate positioning with respect to the monochromatic beam incident on the sample. The detector area would most likely be gas filled and located in a fixed, well shielded housing.

4. POLARIZATION ANALYSIS

One of the particularly attractive features of the HYSPEC concept is that it can quickly and easily be adapted to polarization analysis. All that is required is that the focusing crystal used for non-polarized studies be replaced by a crystal that both focuses and polarizes the monochromatic beam incident on the sample and that polarization analyzers be installed between the sample and detectors. In the immediately following paragraphs we briefly describe the crystal we propose using for polarized applications. The selection of a polarization analyzer will be addressed in paragraphs that follow.

4.1 The Polarizing Crystal

Polarizing crystals are crystals of ferromagnetic materials with low index Bragg peaks in which the nuclear and magnetic contributions to the coherent scattering add for one of the two neutron spin states and subtract and cancel for the other when the atomic magnetic moments are fully aligned by an external magnetic field. Of those investigated to date, the consensus view is that the Heusler alloy crystal Cu_2MnAl is the best choice in terms of both reflectivity and polarizing efficiency. Detailed studies - such as those made, for example, by A. Freund et al [Physica 120B, 86 (1983)] - have shown (i) that well annealed Cu_2MnAl crystals have (111) Bragg reflecting efficiencies that can approach those expected for ideally imperfect crystals, (ii) that the polarization of the reflected neutrons is in excess of 95% in crystals in which the Mn moments are fully aligned in an external field (in this case produced by permanent magnets) and (iii) that there is no significant loss of polarization when individual crystal plates are mounted so as to form a cylindrically-curved, vertical focusing array.

We are aware that Cu_2MnAl crystals, like all polarizing crystals, are not without problems. One is that the procurement of crystals of good quality has been difficult in the past. We have been advised, however, that they can now be obtained from the ILL. A second is that the nuclear and magnetic contributions to the (222) Bragg reflection are not well-matched and second order contamination of the reflected beam can significantly reduce the polarizing efficiency. Fortunately this is ruled out as a potential problem for HYSPEC because the upstream choppers will deliver a higher-order-free, monochromatic beam to the crystal. And third, there is the issue of parasitic Bragg reflections which are known to have a non-negligible impact on both the polarizing efficiency and reflectivity and are likely to impair performance at certain neutron energies. The time-honored way to side-step this difficulty is simply to chose a crystal orientation that gives optimal performance at the energy at which the spectrometer will most often be used, in this case 15 meV, and incorporate messages into the operating software that warn experimenters about operating at those incident neutron energies where the polarizing efficiency and/or

reflectivity is significantly affected by parasitic reflections. Since this will somewhat limit experimental flexibility, it is our intention to explore the feasibility of fabricating the individual plates in the vertical focusing array from stacks of 0.5 mm thick wafers cut from Cu_2MnAl single crystals. There are two potential advantages of this so-called "composite crystal" approach. One is that composite wafer stacks are not as spatially coherent as monolithic crystals; this reduces parasitic reflections. The other is that introducing mosaic into the wafers by single-axis deformation before they are bonded together creates a highly anisotropic mosaic that improves the reflectivity without impacting the vertical focusing.

4.2 The Polarization Analyzers

Signal and background are typically of comparable intensity in polarization analysis measurements. Translated into practical terms, this means that definitive determination of the signal polarization will only be possible if both the polarizer and polarization analyzer have polarizing efficiencies in the 0.80–0.95 range. Heusler alloy polarizer crystals, which produce polarizations on the order of 0.90–0.95, easily meet this standard over the neutron energy range in which HYSPEC will operate. Because they are of comparable efficiency and are both well-tested and maintenance-free, our choice for analysis of the polarization of the scattered neutron beam is the supermirror-bender polarization analyzer. Reduced to essentials, this type of analyzer is nothing more than a short, curved multi-channel guide with magnetically-aligned, polarization-selective Fe-Si supermirror films on the channel walls. Because the angles of total reflection of the + and – spin states differ by more than a factor of three in such films, neutrons of one spin state tend to follow the curved channels while those of the other continue in their original direction. The incident beam is thus divided into divergent beams of opposite polarization. When the beam is well-collimated and the bender optimally curved and tilted, polarization analyzing efficiencies in the 0.80-0.95 range are easily achieved. Moreover, at sufficient analyzer-to-detector distances the two beams become spatially separated and both polarizations can be observed at the same time [S.H. Lee and C.F. Majkrzak; J. Neutron Res. 7, 131 (1999)].

To make the most effective use of the extended angular acceptance of the HYSPEC detector array, an equivalent array of bender analyzers would need to be installed, each directly downstream of a 20 minute angular acceptance collimator. Like all such analyzers presently in use, we envision that the bender channels would be formed by single crystal Si wafers with surface coatings of Fe-Si supermirror film. For HYSPEC the wafers would be 0.25 mm thick, 5 cm long and 14 cm high. To perform optimally at 15 meV, they would be horizontally bent to a radius of about 5 meters. Packed in groups of 80 into 3 cm exterior width thin-walled aluminum alloy containers, the wafer packs would form 2 cm wide, 80 channel benders. Room temperature single crystal Si, it should be noted, is sufficiently transparent to neutrons in the energy range of interest that scattering and absorption losses would be of the order of 10 percent. Permanent magnets would be used to align the Fe moments in the Fe-Si films. Assuming the closest possible packing of the containers along an arc 55 cm from the sample axis, 19 such bender analyzers could be positioned within the (60 degree horizontal and 15 degree vertical) solid angle subtended by the detector array.

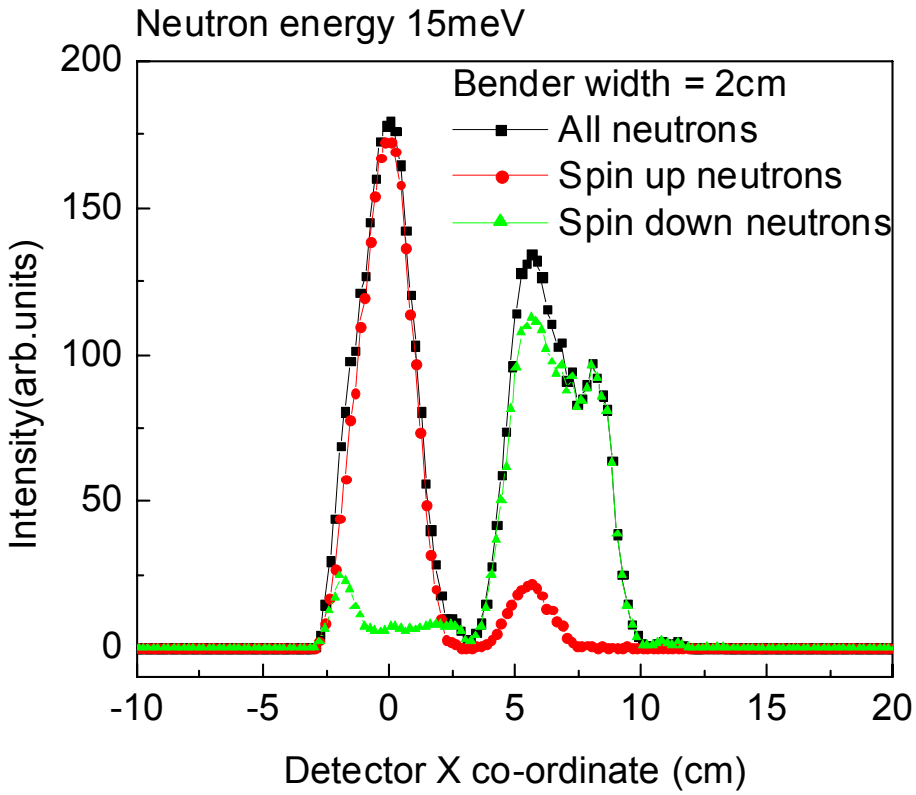


Figure 3. Spatial distribution of two neutron polarizations on the detector as created by a single supermirror-bender

From the Monte Carlo (MC) simulation shown in Figure 3, it is evident that the spatial profiles of the two oppositely polarized beams would be completely separated at the detector bank 4.5 meters from the sample axis. It is also evident in the figure that the undeflected beam will spread over two (2.5 cm diameter) detectors and the oppositely polarized (somewhat wider) beam following the curved channels will fall on the adjacent three detectors. Considering that each bender analyzer unit would be centered on a group of 10 detectors, 50 percent of the detector array would be actively collecting data in this arrangement (which is very nearly optimum from the standpoint of maintaining adequate spatial separation of the beams from adjacent analyzers). Note that although each of the 19 analyzers is limited to a specific scattering angle, rotation of the detector bank around the sample axis will make it possible to cover - sequentially - the full scattered neutron angular range. No more than ten rotational steps of the detector would be needed to monitor both the flipped and unflipped spin intensities over the full 60 degree angular range covered by the detector array. It is also clear from the Figure 3 that the flipping ratio measured in some detectors is higher than that in the others (in which the peak of the "wrong" polarization occurs). Therefore, if the signal is sufficiently strong, it is possible to enhance the polarization sensitivity by restricting the counting to the detectors with the highest flipping ratios. This is equivalent to tightening the beam collimation after the bender-polarizer, but could be done by the experiment analysis software without

redoing the actual measurement. Such flexibility is another attractive feature of the supermirror-bender polarization analysis scheme proposed for HYSPEC.

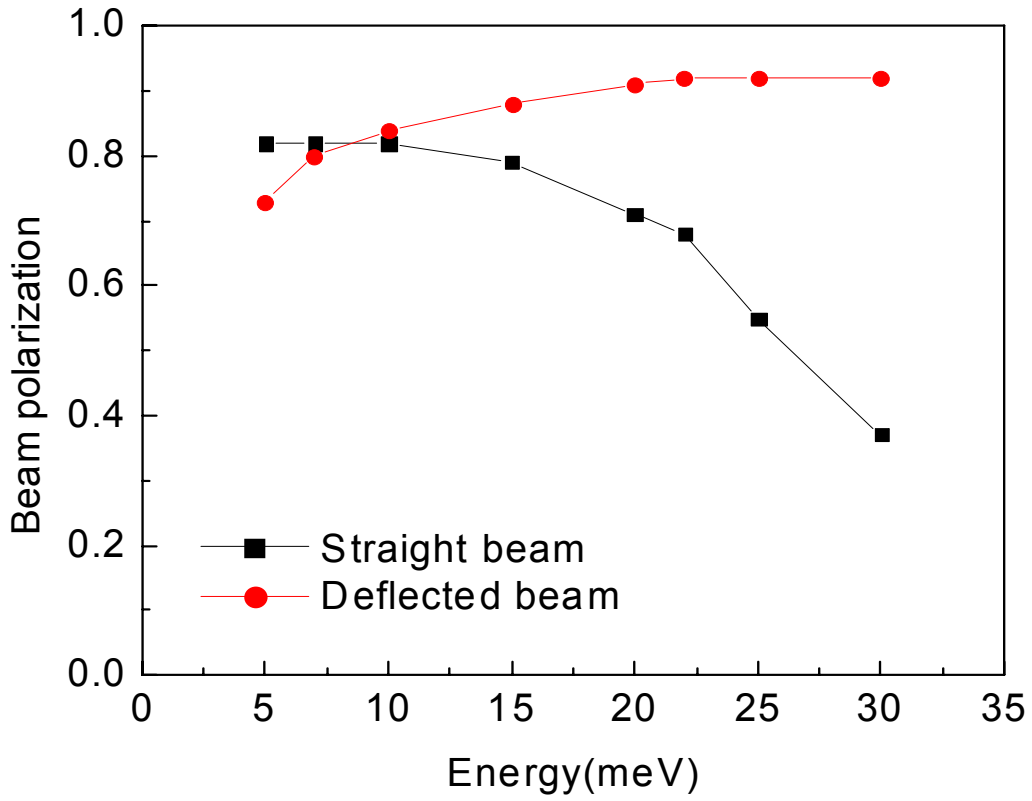


Figure 4. Average beam polarization in each of the two beams

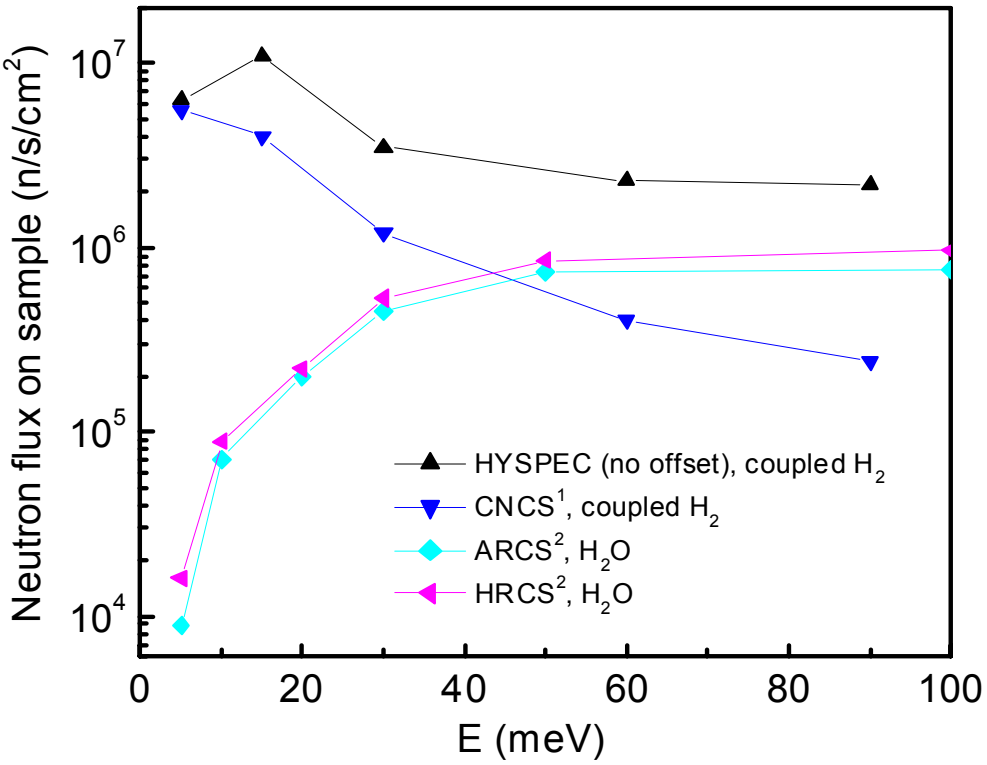
Figure 4 shows a MC simulation of bender analyzer performance over a range of energies on either side of 15 meV. From this it is evident that benders designed for optimum polarizing efficiency at 15 meV will operate with little loss of efficiency over an energy range extending from roughly 8 to 18 meV. A second set of essentially identical bender analyzers configured for optimum performance at 5 meV would cover the energy range from about 3 to 8 meV with about the same efficiency, making it possible to span the entire scattered neutron range from 3 to 18 meV.

Probably the most attractive feature of using bender analyzers in a time-of-flight energy analysing system like HYSPEC's is the capability of monitoring *both* scattered neutron spin states simultaneously over a relatively broad scattered neutron energy range. But there are other attractions as well: bender analyzers are extremely stable and, as noted above, once built require little or no attention and are completely maintenance free.

5. PERFORMANCE

Of the many special features of the instrument, the most important from the viewpoint of efficient use of source neutrons is the utilization of the superior focusing properties of curved crystals to concentrate the monochromatic flux on sample.

To quantify the advantage of the HYSPEC concept in this respect, a number of MCSTAS, Monte-Carlo-based, flux-on-sample simulations were made to compare HYSPEC's flux-on-sample with that of other proposed SNS inelastic instruments. As is evident in Figure 4, despite the finite reflectivity of the PG crystal, its superior focusing properties concentrate the monochromatic flux more effectively than converging guides over the greatest part of the energy range in which HYSPEC will operate. For the moderate resolution, single crystal sample studies for which it is planned, the HYSPEC approach is markedly superior.



[1]CNCS model based on "Optimization...",J.V.Pearce et al.

[2]G.Granroth, Private communication

Figure 5. Calculated flux on sample for HYSPEC and other inelastic spectrometers planned for the SNS

5.1 Moderator Choice

Monte Carlo simulations have also shown that to perform optimally over the incident neutron energy range of interest (5-90 meV), a spectrometer of this type needs to be located on a beamline served by a coupled, 20 K, supercritical H_2 moderator. Our first choice would be Beamline 15. Because optimum performance requires that the monochromator and analyzer energy resolutions be reasonably well matched, the length of the monochromating section should not be much greater than 20 meters. It is also importantant that it ends at a place on the SNS experimental floor where there is

sufficient space to accommodate both the rotating drum shield and the relatively extended time-of-flight analyzer and its associated shielding. Extending the instrument to an outside building would lead to a detrimental mismatch between the monochromating and analyzing resolutions and an unacceptable reduction of monochromatic flux at the sample position.

6. ADVANTAGES OF THE HYSPEC CONCEPT

Apart from the special features described above, HYSPEC has many other attractive properties:

• Incident neutron energy selection would be primarily by time-of-flight as would analysis of scattered neutron energies. Thus full advantage is taken of the pulsed nature of the beam.

• Only two components - the counter-rotating chopper pair and the Fermi chopper – would operate at high rotation speeds. Both would be of conventional design and would operate well within the limits imposed by the tensile strengths of the available materials.

• Moving the sample out of the direct beam and placing shielding between it and the detectors is expected to reduce beam-related background to a minimum.

• Near-forward sample scattering will be accessible to investigation.

• Depending on the type of measurement, the spectrometer operation could be optimized for either a wide-angular-acceptance or Q-selective mode.

• Apart from its focusing properties, the crystal also has the important effect of substantially reducing the (source produced) spectral asymmetry of the beam incident on the sample: the energy resolution function will be more symmetrical than on other instruments on coupled moderators.

• A relatively wide primary beam (4 cm) will be available thus making it possible to fully illuminate large as well as small samples.

• A broad and continuous range of incident neutron energies ($5 \leq E_i \leq 90$ meV) will be accessible without order contamination.

• Energy resolution can be incrementally varied simply by changing the rotation rate of the counter-rotating and /or Fermi choppers.

• Changing the collimation of the incident monochromatic beam or the angular acceptance at the analyzer (or both) is all that is needed to vary the longitudinal Q-resolution.

• Various kinds of monochromating crystals with a variety of focussing options could be employed such as, for example, two-dimensional focussing, asymmetrically-cut, perfect crystal focussing, etc.

7. FUTURE CONSIDERATIONS

Yet to be addressed in detail are two design issues. The first and most important relates to keeping the background to a minimum: a choice will have to be made between a curved guide with a beam stop inside the monochromator drum shield and a straight guide with a “get-lost” pipe with the beam stop a considerable distance from the data collecting area. The second issue is purely mechanical: the large (and heavy) analyzer of the spectrometer will have to be rigidly coupled to the large (and heavy) monochromator

drum shield and move with it when the incident energy is changed. Additionally, the detector array will have to rotate in a precise and reproducible fashion about the sample axis. This, however, does not seem to pose any conceptual difficulty, as similar task is successfully accomplished in the design of several existing modern neutron spectrometers, such as FOCUS at Swiss Spallation Neutron Source SINQ, or a Spin-Echo machine recently commissioned at NIST Center for Neutron Research.

There is also a location constraint that the Committee will need to take into consideration: because the proposed secondary flight-path is sufficiently long (4.5 m), and in addition the sample axis is offset from the primary beam (1.8 m), the combined drum shield, analyzer and associated analyzer shielding will require an ample amount of space on the experimental floor and limit the choice of beam lines.

8. INSTRUMENT DEVELOPMENT TEAM

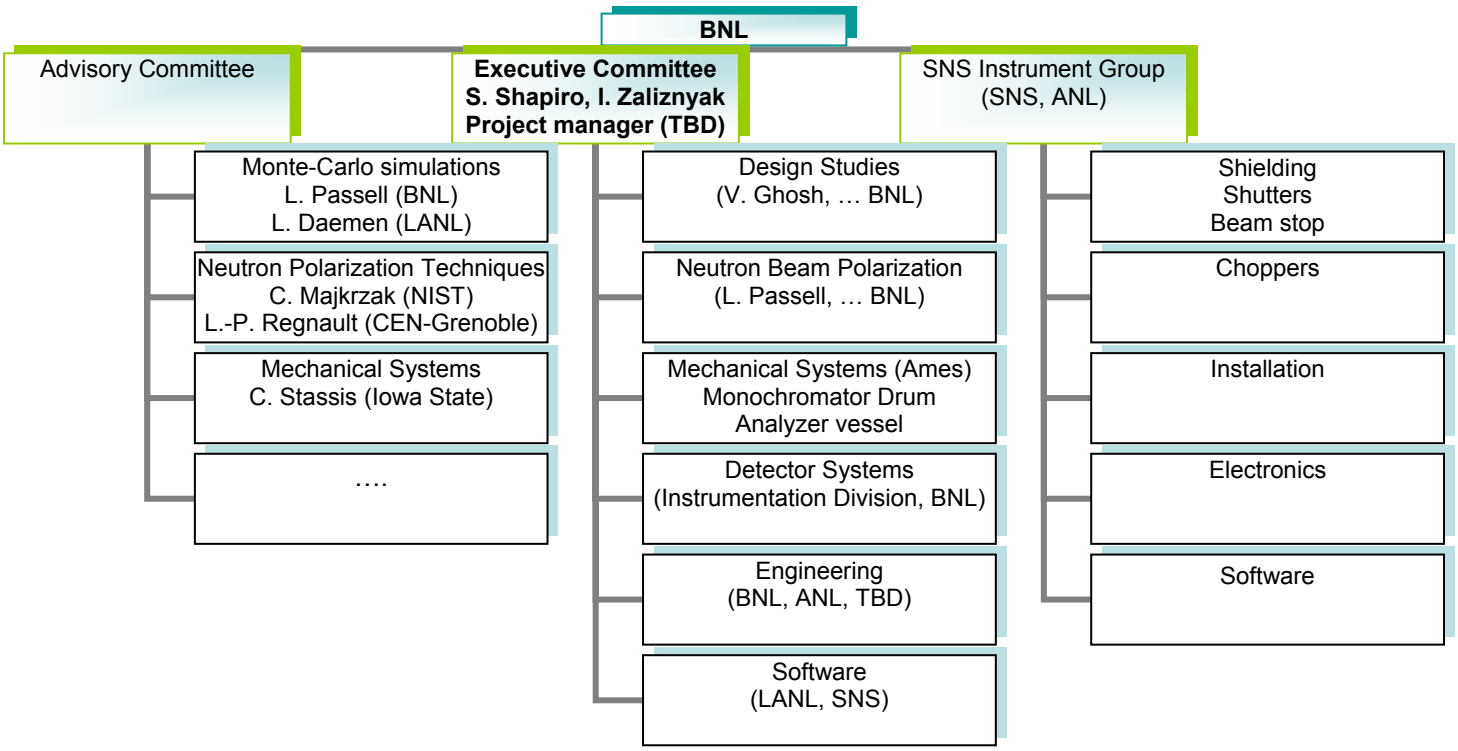
The members of the IDT are listed below and come from a variety of national and international universities and laboratories. All have, however, a common interest in investigation of low energy, atomic-scale dynamical properties of crystalline solids. Many are relatively senior members of the condensed matter community with well-established scientific credentials such as Tranquada, Lander, Shirane, Rhyne, etc. who expect to use HYSPEC to continue their research programs. The IDT also includes younger faculty members such as Lee, Greven, Kiryukin, Hirota who are still in the process of building their scientific programs and will use HYSPEC as one of their important research tools. Other members of the IDT have particular areas of expertise, which are vital to the project. For example, Majkrzak and Regnault are leading experts in polarized neutron beams, the Ames group (Stassis) has extensive experience in building large mechanical structures, and Daemen is a leader in Monte Carlo simulations and has extensive experience in software development. It also goes without saying that there will have to be close interaction with the SNS Instrument team and Abernathy is currently has that connection.

8.1 Members of the IDT and their Institutional Affiliations

I. Zaliznyak, co-PI	BNL
S. M. Shapiro, co-PI	BNL
G. Shirane	BNL
J. Tranquada	BNL
L. Passell	BNL
D. Abernathy	SNS
L. Daemon	Los Alamos
M. Greven	Stanford
B. Gaulin	McMaster
K. Hirota	ISSP
V. Kiryukhin	Rutgers
G. Lander	EITU
Y. Lee	MIT
C.Majkrzak	NIST

S. Nagler	ORNL
R. Osborn	ANL
L. P. Regnault	CEN-Grenoble
J. Rhyne	U. Missouri
C. Stassis	Ames/Iowa St.
A. Zheludev	ORNL

8.2 Project Current Organization Chart



9. ESTIMATED BUDGET

The HYSPEC estimated budget is presented in Table II below. It is necessarily tentative and was arrived at after consultations with the SNS Instrument team following a visit to Argonne in November 2001. Some items which would be common for other instruments such as shielding, detectors, choppers, etc were estimated from the HRCS Instrument Summary for IDT prepared by G. Ganroth (July 8, 2002). These numbers include overhead and contingency. We estimate that within a realistic funding profile the instrument could be built in 5 years from onset of funding. This is consistent with most instruments already funded, such as the CNCS.

Table II. HYSPEC Estimated Budget

Primary Flight Path			
	Choppers		1400
	Incident beam shielding		2300
	Supermirror guides		500
	Beam monitors		100
		SUB	4300
Monochromator			
	Shielding		1000
	Crystals and holder		400
	Collimators		50
		SUB	1450
Sample stage			
	Goniometer, sample table		160
	Ancillary equipment		250
		SUB	410
Analyzer, detector			
	Collimator sets		200
	Polarization benders		1000
	PSD, electonics mounts		900
	Beam Controls-DAQ		140
	Detector vessel		1500
	Beam stop/get lost pipe		200
		SUB	3740
	CAPITAL TOTAL		9900
	LABOR (1/3 Capital)		3300
	TOTAL		13200

Appendix A. SCIENCE EXAMPLES

A.1 Complex Alloys and Nanomolecules

The proposed HYSPEC spectrometer is a world-class instrument with polarization analysis capabilities, a feature that makes it a unique instrument among those to be installed at SNS. Actually, the capability of using polarized neutrons is essential in practically any detailed study of the magnetic properties of condensed matter systems. Such studies extend from the determination of the magnetic form factor to a detailed analysis of magneto-vibrational scattering studies, such studies provide invaluable information about the static magnetic properties and, most importantly, the interactions between magnetic and other excitations in condensed matter systems. Currently though, only for some of the reflectometers previewed for the SNS a simple polarized neutron capability is envisaged. Polarization analysis capability of the instrument is of particular importance for the study of complex phases. These phases exhibit complex arrangements of various interacting degrees of freedom (such as spin, orbital moment, charge, etc.), with fascinating physical properties, some of which may be exploited in emerging technologies ranging from high-T_c superconductivity and spin valves to photonic switches and quantum computing. The well-known examples of such systems, presently under intense experimental and theoretical scrutiny, are the various phases of superconducting cuprates and the magnetoresistive manganites.

Among the complex systems whose physical properties can be exploited in emerging technologies and which therefore have recently attracted considerable attention are the magnetic molecules and the intermetallic compounds of $R_5(Si_xGe_{1-x})_4$ family.

A.1.1 $(Si_xGe_{1-x})_4$ Intermetallic Compounds

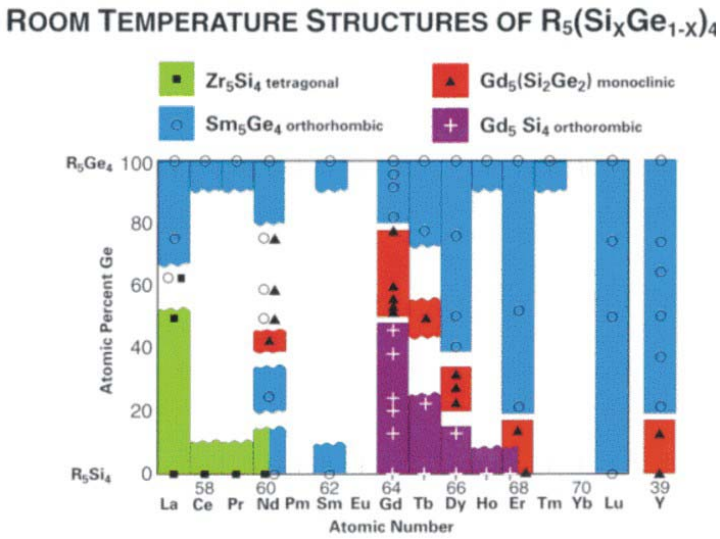


Figure 2.1. The homogeneity ranges and structure types observed in various $R_5(Si_xGe_{1-x})_4$ systems at room temperature

This family was discovered over thirty years ago[2], and has recently opened up an unprecedented opportunity to solve the century-old problem of the intrinsic relationships between the chemical composition, atomic structure and properties of metallic materials. These compounds exist in nearly every R-Si-Ge system, and the distribution and room temperature crystallography of the $R_5(Si_xGe_{1-x})_4$ phases is illustrated in Figure 2.1.

The crystal structures of the $R_5(Si_xGe_{1-x})_4$ phases consist of

[2] F. Holtzberg, R.J. Gambino, T.R. McGuire, J. Phys. Chem. Solids **28**, 2283 (1967).

36 atoms per unit cell distributed among 6 to 9 independent crystallographic sites.[3] These alloys exhibit a number of diverse and unique properties[4] associated with both their naturally layered crystal structures and the combined magnetic-crystallographic transformations at low temperatures, driven by a reversible breaking and reforming of specific covalent Si(Ge)-Si(Ge) bonds (Fig. 2.2, left). The crystal structures change via a martensitic-like collective shear movement of sub-nanometer thick slabs by as much as 1.1 Å and are easily affected by magnetic field, temperature, and pressure. The transitions are accompanied by a giant magnetocaloric effect, a colossal magnetostriction, and a giant magnetoresistance, suggesting many possible technological applications in sensors and energy-transforming devices through intelligent manipulation of the phase transformation by composition, applied fields, temperature, and pressure.

For the most part, identical crystallographic transition also occurs at high temperature in the paramagnetic state (Fig. 2.2, right) when the alloy stoichiometries are near critical (i.e. those where compositional variations induce a crystallographic phase change). X-ray powder diffraction measurements indicate that both the low-temperature magnetically ordered and the high-temperature magnetically disordered orthorhombic Gd_5Si_4-type phases are the same. Preliminary first principles calculations[5] indicate, on the other hand, that the high temperature phase transition may be an order-disorder transformation, during which Si and Ge atoms are redistributed among their respective lattice sites. This crystallographic phase change has a far-reaching effect on the magnetic properties of the material, as illustrated in Fig. 2.3.

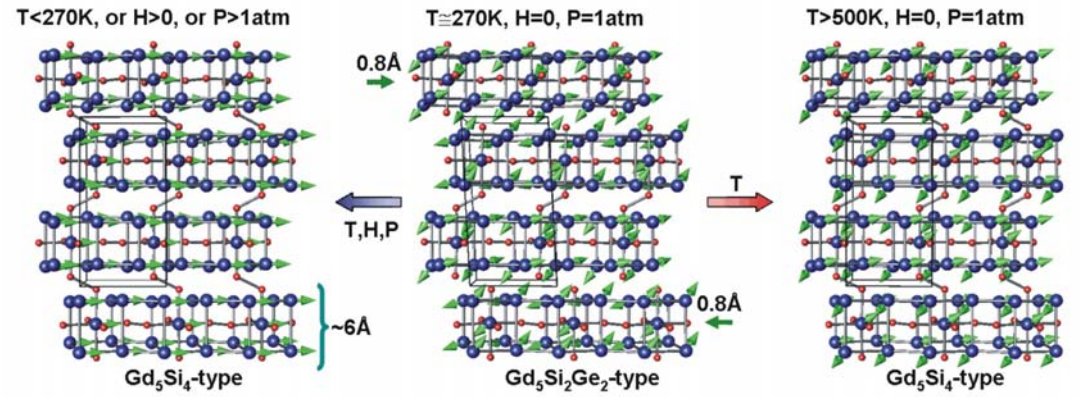


Figure 2.2. The schematic of the coupled magnetic-crystallographic transformation in the $Gd_5(Si_2Ge_2)$ compound, which near ~270 K may be triggered by any of the three thermodynamic variables: temperature, magnetic field and pressure (left). The schematic of the crystallographic-only temperature induced transformation, which occurs between ~500 and 800 K (right). Gd atoms are indicated using large blue spheres, and Si(Ge) atoms are shown as small red spheres. One of the sub-nanometer thick slabs is highlighted by a bracket. Green arrows indicate the directions in which the slabs move during crystallographic phase changes.

[3] V. K. Pecharsky, K.A. Gschneidner, Jr., J. Alloys Compd. **260**, 98 (1997); W. Choe, V.K. Pecharsky, A.O. Pecharsky, K.A. Gschneidner, Jr., V.G. Young, Jr., G.J. Miller, Phys. Rev. Lett. **84**, 4617 (2000).
[4] V.K. Pecharsky and K.A. Gschneidner, Jr., Adv. Mater. **13**, 683 (2001).
[5] V.K. Pecharsky, G. Samolyuk, V.P. Antropov, A.O. Pecharsky, and K.A. Gschneidner, Jr., J. Solid State. Chem. (Submitted; Presented at the 23rd Rare Earth Research Conference, UC Davis, CA, July 13-18, 2002).

Only systematic neutron scattering studies as a function of composition temperature, magnetic field, and pressure can provide a detailed description of the various phases of these systems, the magnetic-crystallographic transformations and their origin. For such detailed studies, the polarization analysis capability of HYSPEC will be an invaluable tool, since it is essential to establish the origin (magnetic or nuclear) of the large number of peaks observed in the elastic and inelastic neutron scattering studies of these materials.

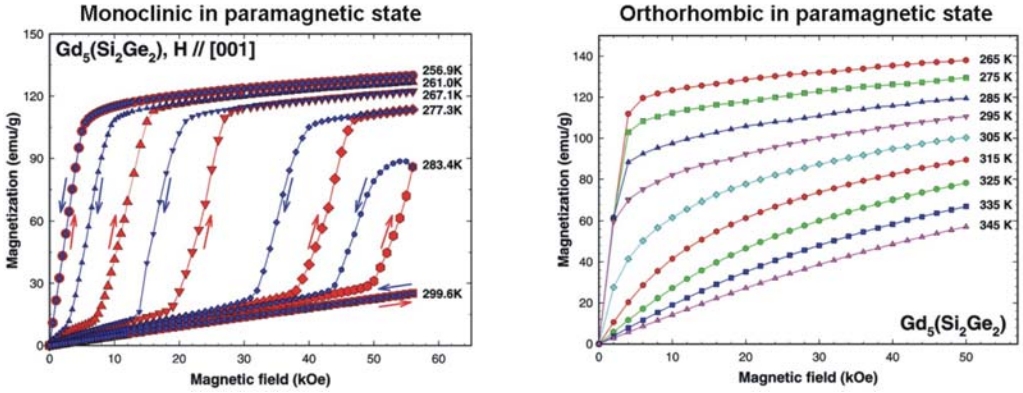


Figure 2.3. The magnetic behavior of two different polymorphs of the $Gd_5(Si_2Ge_2)$ compound. The monoclinic $Gd_5(Si_2Ge_2)$-type structure with half of the interslab bonds undergoes a first order magnetic-martensitic transformation during isothermal magnetization (left). The orthorhombic Gd_5Si_4-type structure with all interslab bonds displays a conventional second order paramagnetic-ferromagnetic transformation.

Such detailed studies combined with first principle calculations are essential for a fundamental understanding of the fascinating properties of these compounds (such as the magnetocaloric effect). The proposed instrument at the high intensity SNS facility is ideal for such detailed investigations.

A.1.2 Nanosize Magnetic Molecules

The study of systems consisting of magnetic molecules is presently a rapidly developing field within nanomagnetism. These are crystalline materials are composed of identical molecules which can be manipulated to obtain a large variety of spin designs, and can be used as prototypes for the study of fundamental problems of magnetism at the nanoscale level (such as quantum tunneling and quantum coherence). An understanding of these phenomena on a fundamental level may help the development of important technologies from spin valves to quantum computing.

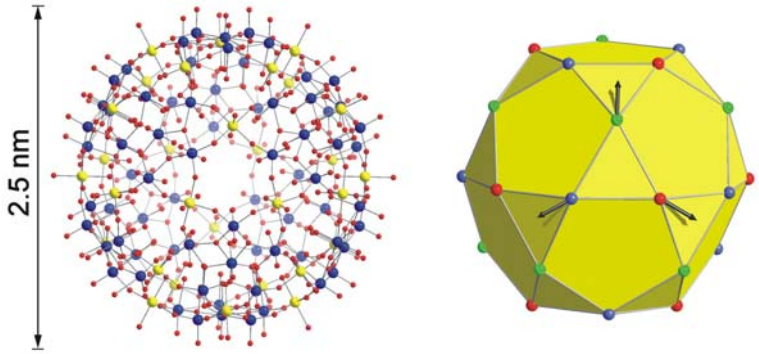


Figure 2.4. Structure of the $\{Mo_{72}Fe_{30}\}$ molecule (left; Mo: blue, Fe: yellow, O: red) and the inscribed Fe_{30} icosidodecahedron (right; the classical ground state is characterized by three spin vector subsets [blue, green, and red Fe centers]). In certain targeted derivatives, all or a part of the thirty Fe(III) centers will be replaced by different di- and tri-valent paramagnetic ions.

The more recently synthesized systems consist of magnetic

molecules containing a number of 3d or 4f magnetic ions connected by organic or inorganic linking atoms. The advantage of these systems over conventional nanosized magnetic particles is that they can be synthesized as crystals, and this allows a systematic study of their properties by neutron scattering techniques. Since the size as well as well as the spin of the magnetic ions can be controlled to some extent these systems are ideal to study the transition between atomic and nanoscale magnetic phenomena as well as the range from S = 1/2 quantum spins (e.g., Cu and V ions) to essentially classical spins (e.g. Fe^{3+} (s=5/2), and Tb^{3+} (J=6) ions). Systems with magnetic molecules containing as few as two magnetic ions are presently available. To date systems with magnetic molecules containing as many as 30 magnetic ions have been synthesized. An example of such a system ($Mo_{72}Fe_{30}$) is illustrated in Fig. 2.4.

To date, only a few neutron scattering experiments have been performed on these systems, mainly because of inadequate energy resolutions over different energy regions. For complete understanding of the dynamical properties of these systems, it is necessary to perform elastic and inelastic neutron scattering experiments as a function of both temperature (down to approximately 10 mK) and magnetic field (up to approximately 10 Tesla). The observed magnetic energy levels can be compared directly with the results of theoretical calculations and can be used for the determination of the parameters used in the model Hamiltonian. Of particular interest here is the determination of the anisotropy parameter, which is presumably due to the spin-orbit interaction and which may be estimated theoretically. Particularly challenging is the determination of the (small) zero-field energy splittings of the ground state at low temperatures. Neutron scattering studies in a magnetic field will also allow us to observe the evolution of the ground state of the system as a function of the magnetic field. This will provide important information about the onset of magnetic level crossings. Again, the polarization analysis capability of the HYSPEC spectrometer will be an invaluable in the study of these interesting systems.

In summary, the detailed study of complex systems, which are being synthesized so that they exhibit physical properties of potential use in emerging technologies, is a challenging problem for which the use of the polarization analysis capabilities of the HYSPEC spectrometer will be invaluable. Still, for a detailed study of problems of this complexity, data obtained in the HYSPEC spectrometer must be combined with the information obtained in complementary instruments at SNS, such as the proposed High Energy Resolution Chopper spectrometer and the backscattering instrument.[6]

A.2 Functional Materials

Fundamental understanding of the microscopic processes governing physical phenomena in the functional materials is key to the technological progress. HYSPEC is an indispensable experimental tool needed to obtain such understanding.

A.2.1 Nanoscale Features of Functional Materials

The drastically enhanced electronic and magnetic responses occurring in complex functional materials as a result of inhomogeneity on a microscopic length scale have recently attracted very significant attention. Both fundamental properties, and enhanced

[6] The author is grateful to Professors Gschneidner, Jr., Pecharsky, and Luban, and Dr. Kögerler of the Ames Laboratory for providing information for this brief summary.

materials characteristics potentially useful for industrial applications are currently being actively investigated. During the last 5 years, it became clear, for example, that nanoscale inhomogeneities are intrinsic to a number of important oxide systems, such as superconducting cuprates[7], magnetoresistive manganites[8], and lead-based relaxor ferroelectrics.[9] Also, understanding and controlling the giant structural, optical, and magnetic responses of inhomogeneous materials holds great potential for the creation of technologically advanced consumer products.

Some of the most important systems discussed above exhibit nanoscale magnetic inhomogeneities. In magnetoresistive manganites, for example, nanoscale magnetic inhomogeneities play the crucial role in the so-called Colossal Magnetoresistance (CMR) effect.[10] While it is known that nanoscale magnetic regions exist in these materials, the structural and magnetic properties of these regions remain largely uncharacterized.[10,11] To understand the physics of the CMR effect, and to guide future work for the synthesis of materials with enhanced magnetoresistive properties, it is crucial to determine the magnetic and structural properties of the inhomogeneous states realized in these materials.

Another class of experimental systems that has recently attracted significant attention are doped magnetic semiconductors, such as Mn-doped GaAs and InAs, and Co-doped ZnO and TiO_2.[12,13] The recent claims[13] of synthesis of room-temperature magnetic semiconductors have generated a significant controversy. It is becoming clear now, that in many cases the prepared systems are actually very fine mixtures of non-magnetic semiconductors (e.g., GaAs or InAs) and ferromagnetic metals (e.g., MnAs or MnSb). Physical properties drastically evolve when the mixed systems vary from complete solid solution to macroscopically phase-separated materials. [14] Some of these systems exhibit very large values of magnetoresistance.[15] A number of these materials can now be synthesised in the bulk form. In order to understand this intriguing behavior, it is essential to characterize the microscopic properties of these materials, with special attention paid to the nanoscale magnetism. In this case, as well as in the CMR manganites discussed

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[7] Lang K.M., Madhavan V., Hoffman J.E., Hudson E.W., Eisaki H., Uchida S., Davis J.C., Nature 415, 412 (2002)

[8] E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. 344, 1 (2001).

[9] P. M. Gehring, S.-E. Park, and G. Shirane, Phys. Rev. Lett. 84, 5216 (2000)

[10] P. Dai, J. A. Fernandez-Baca, N. Wakabayashi, E. W. Plummer, Y. Tomioka, and Y. Tokura, Phys. Rev. Lett. 85, 2553 (2000); C. P. Adams, J. W. Lynn, Y. M. Mukovskii, A. A. Arsenov, and D. A. Shulyatev, Phys. Rev. Lett. 85, 3954 (2000); T. Y. Koo, V. Kiryukhin, P. A. Sharma, J. P. Hill, and S-W. Cheong, Phys. Rev. B 64, 220405(R) (2001)

[11] V. Kiryukhin, T. Y. Koo, A. Borissov, Y. J. Kim, C. S. Nelson, J. P. Hill, D. Gibbs, and S-W. Cheong, Phys. Rev. B 65, 094421 (2002)

[12] T. Hayashi, Tanaka M, Nishinaga T, Shimada H, Tsuchiya H, Otuka Y., J. Cryst. Growth 175, 1063 (1997)

[13] M. L. Reed, El-Masry N. A., Stadelmaier H. H. , Ritums M. K., Reed M. J., Parker C. A., Roberts J. C., Bedair S. M., Appl. Phys. Lett. 79, 3473 (2001); Y. Matsumoto, Murakami M, Shono T, Hasegawa T, Fukumura T, Kawasaki M, Ahmet P, Chikyow T, Koshihara S, Koinuma H., Science 291, 854 (2001)

[14] Y. Shon, Young Hae Kwon, Deuk Young Kim, Xiangjun Fan, Fu D, Tae Won Kang, Jap. J. Appl. Phys. 40, 5304 (2001); S-W. Cheong, unpublished. See, also, "Structural Inorganic Chemistry" A. F. Wells, Fifth edition, (Oxford University Press, 1984), and "Physics of Ferromagnetism" S. Chikazumi, (Oxford University Press, 1997)

[15] H. Akinaga, Mizuguchin M, Ono K, Oshima M., Appl. Phys. Lett. 76, 357 (2000)

above, characterization of both static and dynamic magnetic properties will be of great importance. In particular, dynamic magnetic properties of the nanoscale regions are expected to be significantly different from the corresponding bulk systems. In fact, drastic changes in the lattice dynamics properties have recently been observed in relaxor ferroelectrics,[9] which can be considered a non-magnetic analogue to the systems described here.

Polarized neutrons will, undoubtedly, be one of the most effective tools to study the nanoscale magnetism in these, and related, systems. A very important factor underlying the usefulness of this probe is that magnetic signals from systems inhomogeneous on nanoscale are spread both in the reciprocal space, and in energy, and are, therefore, weak. In addition, nonmagnetic background is typically strongly enhanced in such systems. With the proposed spectrometer (HYSPEC) it would be possible to successfully separate diffuse magnetic signals from the background, such as multiple Bragg scattering, phonons, and incoherent scattering due to isotopic disorder, and the polarization analysis will be of crucial importance.

A.2.2 Anomalous Phonon Behavior in Functional Materials

It has long been known that structural phase transformations occurring in materials such as ferroelectrics are driven by a lattice vibrational mode whose frequency tends to zero at the transformation temperature. This so-called 'soft' mode usually occurs at a well defined wavevector in a particular optic or acoustic phonon branch and is frequently accompanied by a 'central' peak, i.e. divergent elastic scattering. The anomalies associated with the soft mode result in very anisotropic dispersion curves. Recent work on ferroelectics like PZT and PMN[9] and on shape memory alloys such as Ni-Al, Ti-Pd, and Ni_2MnGa have demonstrated the need for good wave-vector resolution and for minimization of the extraneous elastic scattering from the sample environment. It also makes clear that single crystals are necessary. Most often these crystals are grown explicitly for neutron scattering experiments and tend to be small. Focussing of the incident beam is therefore vital to assure the maximum number of neutrons impinging on the sample; a requirement met by the proposed HYSPEC instrument.

A.3 Strongly Correlated Electron Systems

Strongly correlated electron systems, from heavy-Fermions to high-T_c to materials exhibiting giant magneto-resistance effects, has added a new class of materials to those with which we are familiar. Of crucial significance in these materials are the role of electron correlation energies and the interaction of the lattice with the spin degrees of freedom. Unlike the BCS superconductors in which vibrational degrees of freedom (the phonons) play a key role in mediating superconductivity, the new phenomena are classified as having a range of interactions, with the dominant one often difficult to identify.

Neutron inelastic scattering is the single most important probe for unraveling the energetics and spatial dependencies of these interactions. At the same time, we have to be able to separate interactions that are electronic from those that are vibrational. This requires polarized neutrons, and very often polarization analysis. We may demonstrate

the argument by invoking the pioneering work by Axe et al.[16] on the vibrational spectra of Nb_3Sn above and below T_c. These experiments, performed in the 1970s, used unpolarized neutrons. Because superconductivity in BCS materials are dominated by vibrational energies there has never been any doubt that the measurements were of phonons. However, if we think today of the superconductivity of UPd_2Al_3, of high-T_c materials, that question is much harder to answer. In both UPd_2Al_3 [17] and high-T_c materials[18] full polarization analysis has proved of enormous value in separating the electronic and lattice effects.

We need to continue to develop the technology of polarized beams and polarization analysis (PA), so that these experiments can extend over a wider range of energy than presently available and not be so demanding in terms of intensity. Very large crystals are presently required to map out excitations in full PA ,[19] which means that the technique cannot be generally used. An instrument at the SNS will therefore open a completely new field of endeavor. Not just to be able to perform PA on selected excitations, but, more importantly, to construct $S(\boldsymbol{Q},\omega)$ maps of spin-flip and non-spin-flip intensity.

Furthermore, efforts in three-dimensional polarization analysis, now done almost exclusively with the spherical neutron polarimeter at the ILL, can often make unambiguous identification of magnetic configurations[20] and needs to be implemented on a wider scale. Its extension into inelastic scattering is just beginning.[21] Many surprises in our understanding of magnetism, and in defining precise interactions, will emerge with the use of polarized neutrons.

A.4 Quantum Critical Points

A quantum critical point is a phase transition in which the temperature at which long-range order is established is suppressed to zero by quantum disorder. This may be induced either externally, e.g. with the application of pressure or magnetic field, or internally through either compositional variations or by increasing disorder. Close to a quantum critical point, the energy scale of critical fluctuations is determined by the temperature which leads to highly unusual scaling of the critical dynamics. Universal scaling in Δ/T has been observed by inelastic neutron scattering in at least two systems believed to be close to quantum critical points, $UCu_{5-x}Pd_x$ and $CeCu_{6-x}Au_x$. Such experiments are challenging because they require measurements over a wide range of energies. Thus far, the most complete data have been obtained on time-of-flight spectrometers at both reactor and pulsed neutron sources. However, the need for large samples has necessitated the use of systems in which quantum criticality is induced by sample non-stoichiometry rather than by an extreme sample environment. Theoretically, it is important to reproduce this scaling behavior in high-quality single crystals that are stoichiometric so that atomic disorder can be discounted as a contributory factor. For instance, the most reliable examples of quantum phase transitions are in systems such as

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[16] J. D. Axe and G. Shirane, Phys. Rev. Lett. **30**, 214; Phys. Rev. B **8**, 1965 (1973)
[17] N. Bernhoeft et al., Phys. Rev. Letters **81**, 4244 (1998)
[18] H. Fong et al., Phys. Rev. B **61**, 14773-14786 (2000)
[19] R. Caciuffo et al., Phys. Rev. B **59**, 13892 (1999)
[20] S. H.Lee et al., Phys. Rev. B **60**, 10405 (1999); A. Hiess et al., Phys. Rev. B **64**, 134413 (2001)
[21] See article by Caciuffo et al. in ILL Annual report 2001

$CePd_2Si_2$ and $ZrZn_2$, occuring at pressures of 28 kbar and 21 kbar respectively. There is also evidence that metamagnetic transitions in some heavy fermion compounds - commonly occurring in fields of greater than 10 T -may be associated with quantum phase transitions. The ability to utilize extreme sample environments on HYSPEC will be especially important in investigating quantum phase transitions. Also, the focusing properties of HYSPEC will be vitally important because of limited sample sizes and volumes.

A.5 Correlated Phases in Many-Electron Systems

The ground states of many-electron systems with spin, orbital and/or charge/valence degeneracy often exhibit non-trivial arrangements of these degrees of freedom which lead to many fascinating physical properties. Recently much attention has been paid to exploring the charge and/or orbitally ordered phases in transition metal oxides such as the superconducting cuprates and magnetoresistive manganites.[22] Understanding the nature and origin of these phases is of both fundamental importance and technological interest. Neutron scattering is the most informative and least perturbing experimental way of providing information on the correlated structural distortions and/or magnetic correlations in the charge/orbital/spin-ordered phases. Among the unique features inherent to the neutron scattering approach is that it provides both good energy and wavevector resolution, which permits accurate discrimination between the static and dynamic correlations and allows separate characterization of the corresponding correlation ranges. Examples where this is of crucial importance are the recent observations of striped liquid phases in the doped layered nickelates, incommensurate dynamical correlation in the cuprate superconductors and characterization of dynamical slow-down in the course of the spin-glass freezing transition in the half-doped cobaltate $La_{1.5}Sr_{0.5}CoO_4$.[23]

A spectrometer, optimized for studies of the short- and long-range static and dynamic correlations in single crystals should be designed to have both a small elastic and negligible inelastic background, a symmetric and well-reproduced resolution function (permitting a straightforward and reliable (de)convolution procedure) and have a polarization analysis capability to distinguish between structural and magnetic scattering. Equally important, it should provide complete and continuous coverage of a significant volume in the reciprocal space of the crystal with both energy and wavevector resolution easily adaptable to the type of the problem studied. Finally, it is essential that such an instrument be easily equipped with a variety of sample environments since definitive insight into the nature of many-electron correlated phases is often obtained by studying how they are affected by changes in external magnetic field, temperature, pressure, etc. As will be evident, the proposed HYSPEC spectrometer is designed to both provide superior performance and to address these concerns.

A.6 Quantum Magnetism

Amazing progress has been made in the field of quantum low-dimensional magnetism in the past twenty years, almost entirely driven by inelastic neutron scattering experiments.

[22] J. Zaanen, Science 286, 251 (1999)
[23] I. Zaliznyak, J. P. Hill, J. Tranquada, R. Erwin, Y. Moritomo, Phys. Rev. Lett. 85, 4353 (2000)

The simplest fundamental model systems are by now very well understood. Today the challenge is to learn about more complex phenomena such as the effects of disorder and impurity substitution, spin-lattice interactions and one- and two-dimensional to three-dimensional crossover effects.

One example of a new research direction in quantum magnetism is the problem of *random-exchange* spin chains. Consider the case of a *S=1/2* quantum spin chain with nearest-neighbor Heisenberg exchange interactions of random magnitude. In the simplest model, a given percentage x of the bonds are of magnitude J_1, and the remaining (1-x) bonds correspond to a coupling strength J_2, the strong and weak bonds being distributed randomly. For a fixed x the limiting cases are easy to understand. For $J_1<<J_2$ the chain breaks up into an ensemble of finite-sized clusters: dimers, trimers, tetramers, etc. Each cluster has a *discrete* excitation spectrum, that in a neutron experiment appears as a series of sharp excitations at well-defined energies. For $J_1=J_2$ the situation is entirely different. In a *uniform* spin chain such as this, the spectrum is an exotic continuum of states with no sharp excitations and a strong wave vector dependence. The question is: how does the spectrum look in the intermediate case of $J_1<\sim J_2$?

To date, the answer is not known, even theoretically. The main reason is that an experimental realization of a model suitable for inelastic neutron experiments has not been available until the recent preparation of the new model compound $BaCu_2(Si_xGe_{1-x})_2O_7$. Solid solutions with $0<x<1$ are characterized by susceptibility curves that can be fitted to a Bonner-Fisher law with an intermediate value of J and are expected to be realizations of the S=1/2 random-exchange model. Inelastic neutron scattering experiments on $BaCu_2(Si_xGe_{1-x})_2O_7$ single crystal samples are expected to provide unique insight into the role of disorder in quantum magnets.

Previous work on the x=1 system was carried out on thermal and cold triple-axis instruments at reactor sources as well as on the MARI time of flight instrument at ISIS. It is clear from these measurements that none of these instruments will be fully adequate for the study of the solid solution $BaCu_2(Si_xGe_{1-x})_2O_7$. First of all, the excitation bandwidth for x close to unity is expected to be of the order of 150 meV and using a spallation source instrument becomes mandatory if one wants to cover the relevant energy range. At the same time, the small sample size (the largest existing crystals of $BaCu_2SiGeO_7$ are about $4x4x10mm^3$) will require the ability to focus the incident beam to a small spot size; a major feature of HYSPEC. Moreover, the weak and diffuse nature of continuum scattering requires a very low and well-controlled instrument background. Finally, to gain a detailed understanding of the spin dynamics, polarized neutron experiments will be needed. All of these needs will be satisfied by the proposed HYSPEC instrument.

A.7 Advanced Polarization Analysis using TAS and TOF spectrometers

Much of the present condensed matter research is devoted to understanding the physics of "strongly correlated electronic systems". This class of systems encompasses a large variety of physical situations: un-conventional "d-wave" superconductivity in high-T_c superconductors (LSCO, YBCO...) or spin-ladder cuprates ($Sr_{14}Cu_{42}O_{41}$…), un-conventional "p-wave" superconductivity (Sr_2RuO_4, UGe_2, $CePd_2Al_2$...), charge-ordered materials (NaV_2O_5, $Sr_{14}Cu_{24}O_{41}$, $LaNiO_{4+y}$...), spin-Peierls materials ($CuGeO_3$, FeOCl, $Pb[Cu(SO_4)(OH)_2]$...), exotic spin dynamics in quantum-chain systems (J1-J2 systems,

alternating chains, Haldane-gap chains, dimer systems, magnetization-plateau systems...), highly frustrated systems (kagome, J_1-J_2-J_3 honeycomb, pyrochlore...), etc.

The physics which is involved in all these systems may be understood from the competition of different degrees of freedom: spin, orbit, charge and lattice distortion. The understanding of mechanisms at the origin of exotic properties is only possible if one can determine accurately and unequivocally the wave vector and energy dependencies not only of the "standard" correlation functions (i.e. those coupling identical degrees of freedom : <spin-spin>, <orbit-orbit>, <displacement-displacement>...), but also the "hybrid" correlations functions coupling different degrees of freedom (<spin-orbit>, <spin-displacement>, <orbit-displacement>...). To our knowledge, the only way to solve this problem is to measure the magnetic-magnetic (MM), nuclear-nuclear (NN) and magnetic-nuclear (interference) terms using the inelastic polarized neutron scattering in conjunction with a powerful polarization analysis technique.

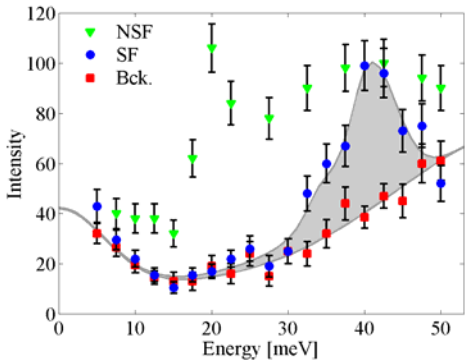

Figure 2.5: Energy scan through the antiferro-magnetic zone center at **Q**=(1.5,0.5,1.7): non-spin-flip (green circles) and spin-flip (blue circles) intensity, and background position **Q**=(1.8,0.6,1.7) (red squares) for an incident polarisation parallel to **Q** (spin-flip channel). The energy dependence of the background is due to the increased counting time at higher incident energies. The shaded area illustrates the magnetic signal peaking at the resonance position of 41 meV.

The best known polarization analysis technique is the so-called "longitudinal polarization analysis" (LPA) method invented long time ago by Shull, Moon, Riste and Kohler.[24] The LPA is invaluable to obtain separately the magnetic and structural contributions, or the various magnetic components at a given scattering vector. We have recently used the LPA on IN22 at ILL to determine directly the magnetic excitation spectrum in the high-Tc compound $YBCO_{6.85}$ (Fig.1)[25] or $Sr_{14}Cu_{24}O_{41}$ (unpublished results). The advantage of using a polarized beam versus an unpolarized one is clearly demonstrated in Fig. 2.5. The magnetic response is superposed to a series of more or less well-controled structural features (phonons, braggons, spurions...) which render the determination highly uncertain. The same scan performed with a polarized beam allows a very precise determination of the magnetic contributions (in particular the resonant peak at 41 meV and the IC dynamic correlations existing between 25 meV and 41 meV). The "normal" method would have consisted in performing constant-energy scans at different energies to determine both the signal and the background.

Some years ago (in the early 80's), the LPA was successfully used on IN12 to demonstrate the relevance of soliton-type non-linear excitations in the quasi-1D antiferromagnet TMMC.[26] In particular, the use of polarization analysis was fundamental

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to determine univocally and separately the contribution $S^{||}(\boldsymbol{Q},\omega)$, directly related to the solitons, and $S^{\perp}(\boldsymbol{Q},\omega)$, related to the inter-soliton spins, Fig. 2.6. With un-polarized neutrons, the method to separate both contributions would have consisted in measuring the Q-dependence of the response, which is problematic if one contribution is much smaller than the other. A recent example is measurement of the SF and NSF contributions in $CuGeO_3$, a prototype spin-Peierls system, which allowed a spectacular determination of all the magnetic (bound states, continuum…) and phonon modes, and the double-gap structure of the magnetic excitation spectrum.

The LPA method can be transposed to TOF machine. At least there is no conceptual interdiction, although the practical realization may not be simple. As important it could appear, the LPA method is still a relatively "old" and "rustic" method. Indeed, LPA only recovers one part of the information. Because the incident polarization direction is selected by applying a magnetic field on the sample, only a single projection of the polarization vector after scattering can be determined. In the last decade an important effort has been made at ILL to develop a new polarization analysis technique, offering the possibility to measure all three components of the final neutron polarization (CRYOPAD concept).[27] The "spherical neutron polarimetry" (SNP) is a new alternative to LPA. It has demonstrated strong capabilities in diffraction, in particular to resolve non-trivial magnetic structures.[28] By accessing the "off-diagonal" components of the polarization tensor (i.e. P_{xy},P_{xz},P_{yz}...), the method allows in pinciple to determine the so-called "inelastic nuclear-magnetic interference" (INMI) terms.[29] SNP is a very elegant but difficult method to determine very-small magnetic or structural **inelastic** contributions through their interferences with strong ones. Although the method has not yet been demonstrated for INS, the SNP could bring new information for, e.g., the relevance of orbital correlations, "hidden-order", "orbital-currents" in High-T_c cuprates or in heavy-fermion materials (URu_2Si_2, etc). This field of investigation is new for INS and almost all remains to be done.

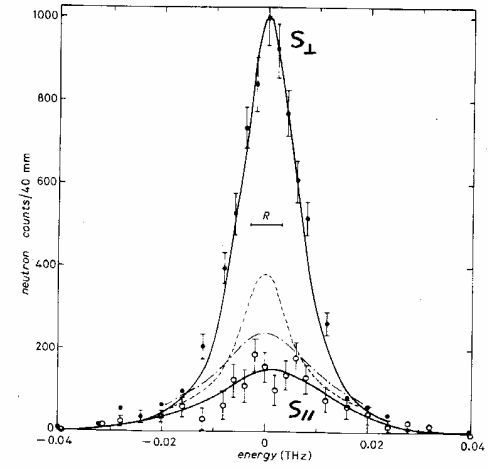

Figure 2.6: Energy spectra of magnetic fluctuations observed in directions parallel and perpendicular to the external field at **Q**=(1.4,0,1.006). R represents the instrumental resolution. The full lines are theoretical prediction of the flipping model.

HYSPEC offers a unique opportunity to develop new polarization analysis techniques at a pulsed spallation source. Of course, a broad-band/broad-angle CRYOPAD adapted to TOF spectrometry has not yet been built (although there is a project and some ideas at ILL). On the other hand, the flexibility of HYSPEC which owes to it "hybrid" nature, will probably make the "mono-detector/narrow beam" SNP method easily adaptable.

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