NEUTRON THREE-AXIS SPECTROMETRY AT THE ADVENT OF 21ST CENTURY

JIRI KULDA
Institut Laue-Langevin
BP 156, 38042 Grenoble Cedex 9, France
E-mail : kulda@ill.fr

Received January 5, 2006

The implementation of multiplexing techniques combined with advances in neutron optics make the neutron three-axis spectrometers (TAS) an efficient tool to map inelastic response from single crystals over momentum transfer ranges comparable to the size of a single Brillouin zone. Thanks to recent progress in polarization techniques such experiments can be combined relatively easily with neutron polarization analysis, which does not only provide unambiguous separation of response corresponding to structural and magnetic degrees of freedom, but permits a quantitative analysis of the magnetic response anisotropy, often of crucial importance to test theoretical predictions. In the forthcoming decade we therefore expect a further development of the complementary use, rather than competition, of the reactor-based TAS's with time-of-flight (TOF) instruments for single crystal spectroscopy at the existing (ISIS) as well as at the newly built (SNS, J-PARK) pulsed sources.

KEYWORDS : Neutron Inelastic Scattering, Three-axis Spectrometers, Elementary Excitations

1. INTRODUCTION

Historically, neutron inelastic scattering studies using three-axis spectrometers (TAS) are synonymous of measuring dispersion relations of elementary excitations along high-symmetry directions in crystalline solids. At present such kind of experiments yields more and more place to studies of other objects like continuum modes in low-dimensional quantum spin systems or fluctuations related to ordering processes, both on short and long ranges. Although other experimental techniques, like NMR or muon spin rotation, can provide valuable information on the same systems, neutron inelastic scattering remains the method yielding the most complete information on the role of space and time correlations and their interplay in the behavior of condensed matter systems. Moreover, neutrons couple with comparable strength to both the structural and magnetic degrees of freedom and the two scattering components can be quite cleanly separated using polarized neutron techniques.

The evolution of the scientific case of neutron spectroscopy in the 90's has implied profound changes in the design of the TAS instruments. While good energy resolution has kept its importance, the resolution in momentum transfer became most frequently traded off for enhanced luminosity. Large, monochromatically focusing crystal arrays used in the design of monochromators and analyzers bring about an increase in data collection rate between 1 and 2 orders of magnitude, permitting to detect weak effects and/or to work with single crystal samples having volumes down to 10 mm^3. The use of neutron polarization analysis continues to be penalized by losses of 1 to 2 orders of magnitude in neutron count-rate as compared to unpolarized work. Nevertheless the progress in focusing and polarization techniques has brought about a qualitative step in the sensitivity of the polarized set-ups on an absolute scale: a state-of-the-art polarized neutron TAS now offers a sensitivity comparable to that of an unpolarized TAS at the same source in the 80's and 90's.

These general trends are well illustrated by a recent study of the stripe ordering in $La_{5/3}Sr_{1/3}NiO_4$ [1]. As often in the case of contemporary TAS experiments, the attention is not focused on the steep dispersion of the collective excitations (magnons) of the Ni magnetic moments but on the diffuse scattering due to the fluctuations across the domain boundaries separating the stripes. Thanks to the upgrade of the ILL IN8 thermal TAS [2], consisting in increase of the beam-tube diameter and in implementation of a large doubly focusing PG 002 monochromator (together with a matching horizontally focusing PG 002 analyzer), it is possible to collect, within a few hours and with a relatively small sample ($V = 0.8\ cm^3$), a complete map of the inelastic response over a $\boldsymbol{Q}$, E range corresponding to the magnetic Brillouin zone (Figure 1a). The measurement

time of the order of hours (to be compared to days on the same instrument in its state of the early 90's) permits repeated acquisition of such maps at different energy transfers and/or temperatures or other external parameters. A similar progress in the polarized neutron TAS (ILL IN20) [3] permits to perform a complementary experiment using polarization analysis to demonstrate that this part of response is due to spin-flip scattering only and, hence, to prove unambiguously the spin nature of the ridges of diffuse scattering (Figure 1b).

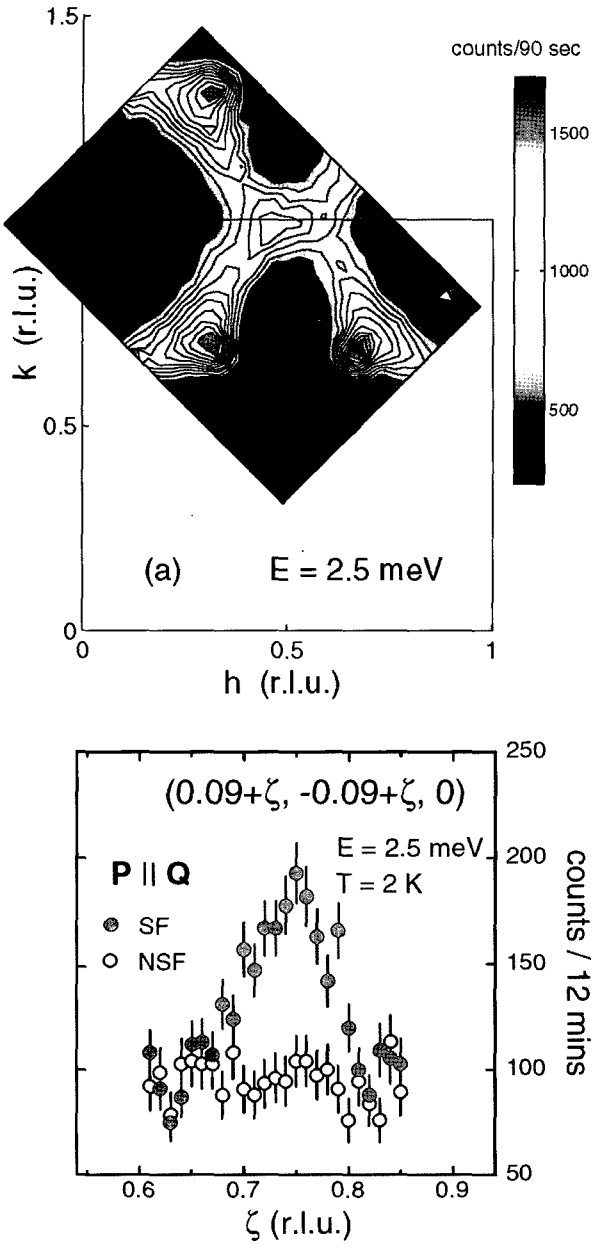

Fig. 1. Map of the Inelastic Response of LSNO (a) and Polarized Neutron Data (b) Demonstrating the Spin Nature of the Ridges Along the <ξξ0> directions [1]

Experiments of this type do not rely any more on the traditional $\boldsymbol{Q}$ = const. scanning mode, which necessitated a combination of sample and analyzer arm movement upon each scan step, and can therefore be accelerated by spectrometer multiplexing: a larger number of separate analyzer/detector channels can collect data simultaneously at different momentum/energy transfers at a given fixed orientation of the sample with respect to the incident beam. An instrument movement anologous to a classical ω-scan can then be used to sweep the inelastic response over a surface in the $\boldsymbol{Q}$, E space. The pioneering implementations of this approach - the E2 flat cone spectrometer at HMI Berlin [4] and the RITA set-up at Riso [5] - are followed by new projects pursuing an optimized design.

As an example, Figure 2 displays a sketch of the *FlatCone* device [6], presently in commissioning phase at the ILL Grenoble: an array of 31 discrete analyzer/detector

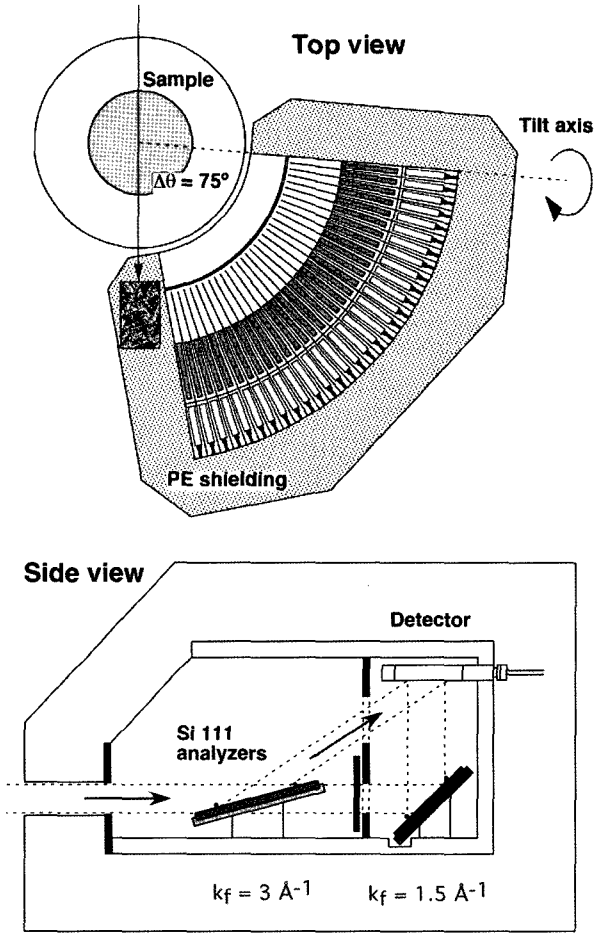

Fig. 2. Schematic Sketches of the *FlatCone* Multianalyzer Setup; the Arrows in the Bottom Part Represent the Neutron Flight Path, Corresponding to the k_f= 3 Å^{-1} Analyzer Used in the Present Experiments

channels covers an angular range of 75° with the possibility to choose between final neutron wave-numbers of 1.5 $Å^{-1}$ and 3 $Å^{-1}$. Moreover, the flat-cone geometry with a tilt angle range of –10° to +20° permits to cover the momentum range of one Brillouin zone in the vertical direction. Fig. 3 displays an example of room-temperature data from a PZN–8%PT (lead-zinc niobate doped by lead titanate) relaxor ferroelectric exhibiting the so-called "waterfall" phonon dispersion anomaly, collected in the course of the *FlatCone* commissioning experiments. The frames delimit the ranges explored in a previous IN8 experiment [7], using a similar experimental setup with Si 111 monochromator and analyzer crystals. The time needed to collect the map data with the same statistics using the step-by-step procedure of IN8 was 16 hours compared to 3.3 hours with the *FlatCone* on IN20 (having a nominally lower monochromatic flux).

Since last decade the scientific case of single crystal spectroscopy has been addressed also by direct geometry TOF spectrometers at pulsed sources, offering the possibility of simultaneous registration of the signal over large areas in the $\boldsymbol{Q}$, E space. This new approach has received a wide attention of the neutron scattering community; however, the flux limitations of the present and nearest future spallation sources do not allow it to become an equivalent alternative to the TAS technique at steady state sources. Rather, the experience from recent years provides evidence for the benefits of a complementary use of both techniques: in case of investigation of a new material the initial exploratory phase on TOF machine (visualization of quantum spin fluctuations in the 1D spin-Peierls compound $CuGeO_3$ on MARI or discovery of incommensurate antiferromagnetic fluctuations in YBCO on HET, both at ISIS) had to be followed by extensive and detailed TAS studies. The reason for this is that the large "pixel power" (product of pixel numbers of their large solid-angle detectors with the number of energy channels of the TOF instruments) cannot always compensate the inherently low incident flux of the existing instruments at pulsed sources.

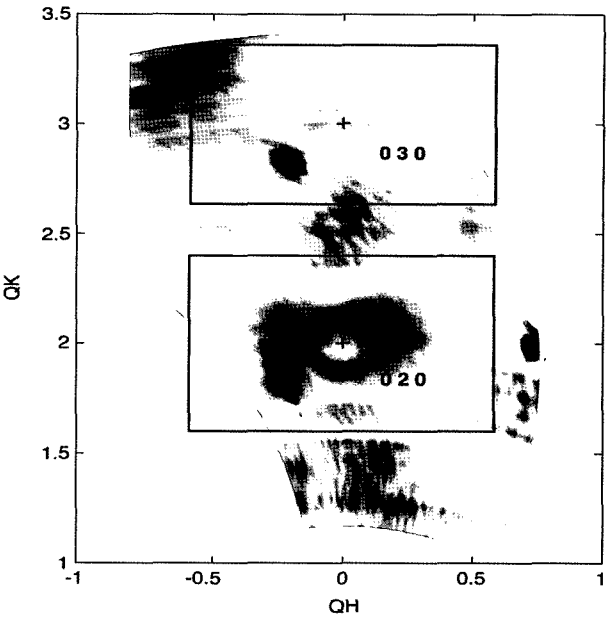

Fig. 3. The IN20 *FlatCone* Constant Energy Map ($\Delta E = 5.5$ meV, $k_f = 3$ $Å^{-1}$) of the Inelastic Response in a PZN-8%PT Relaxor Ferroelectric Crystal in the 020 and 030 Brillouin Zones at and $T = 300$ K; the Black Frames Delimit Areas Mapped in a Previous IN8C Experiment (cf. text)

To assess the relative performances of the TAS and TOF techniques, let us consider a model case, where the $S(\boldsymbol{Q}, E)$ is to be explored over a square area in momentum space of a 0.9 $Å^{-1}$ side with neutrons having a fixed final wave number of $k_f = 4.1$ $Å^{-1}$, corresponding to a solid angle interval of about 0.05 sterradians. The energy range of interest would be 10 - 30 meV, representing about 30% of the maximum incident neutron energy. Table 1 summarizes the relevant quantities for two TAS-based (IN8, IN8 Flat Cone at the ILL) and for two TOF based (MERLIN at ISIS

Table 1. Comparison of the TAS and TOF Performance for a Model Case Described in the Text (based on instrument parameters given on the ILL, ISIS and SNS official web pages)

	IN8	IN8 Flat Cone	MERLIN	ARCS
Incident flux [$cm^{-2}s^{-1}$]	$5\ 10^8$	$5\ 10^8$	$2\ 10^5$	$8\ 10^5$
Pixel number	1	31	50000	12800
Solid angle k_f [sterrad]	0.0033	0.033	3.2	0.25
Relevance factor Q	1	0.25	0.015	0.2
$\Delta E/E_i$	0.07	0.07	1	1
Relevance factor E	1	1	0.3	0.3
Figure of merit	**1**	**2.5**	**0.025**	**0.1**

and ARCS at SNS) setups.

The departure point is the typical TAS resolution volume (single pixel) given for IN8, with a performance limited by its tiny size but always having a full relevance. The solid angle and energy intervals for the other setups, whenever exceeding the above-defined size of the relevant S($\boldsymbol{Q}$, E) range, are scaled down to this value by the corresponding relevance factors. The overall instrument performance is then characterized by the product of the incident flux with detector solid angle, covered energy range (relative to maximum incident neutron energy E_i) and the relevance factors, given in the bottom line of Table 1; the pixel number is given just for illustration.

The resulting figures of merit, significantly more favorable for the TAS instruments, demonstrate that for experiments exploring the S($\boldsymbol{Q}$, E) topology over ranges not exceeding a single Brillouin zone the steady state source instruments based on the TAS technique (without insisting on the number of *three*) keep their importance. As an important corollary to this rather general statement, we may conclude that well-optimized TAS instruments at steady-state neutron sources, providing fluxes only by a factor 3 - 10 times smaller than the ILL high-flux reactor, stay competitive for many interesting inelastic scattering applications.

ACKNOWLEDGEMENT

The author is indebted to many of his colleagues both at the ILL and at other places for helpful discussions.

REFERENCES

[1] A.T. Boothroyd, P.G. Freeman, D. Prabhakaran, A. Hiess, M. Enderle, J. Kulda, and F. Altorfer, *Phys. Rev. Lett.* 91, 257201, (2003).

[2] A. Hiess, R. Currat, J. Saroun, F.F. Bermejo, *Physica B* 276-278, 91 (2000).

[3] J. Kulda, J. Saroun, P. Courtois, M. Enderle, M. Thomas, P. Flores, *Appl. Phys.* A74 [Suppl.] S246-S248, (2002).

[4] R. Born, D. Hohlwein, *Z. Phys. B – Condensed Matter* 74, 547 (1989).

[5] K. Lefman, D.F. McMorrow, H.M. Ronnow, K. Nielsen, K.N. Clausen, B. Lake, G. Aeppli, *Physica B* 283, 343 (2000).

[6] M. Kempa, B. Janousova, J. Saroun, P. Flores, M. Boehm, F. Demmel and J. Kulda, Physica B (2006), Proceedings of ICNS'2006 Sydney, accepted for publication.

[7] J. Hlinka, S. Kamba, J. Petzelt, J. Kulda, C.A. Randall, S.J. Zhang, Phys. Rev. Lett. 91 (2003) 107602