

C11. PANTHER - Polarisation ANalysis with THERmal neutrons

P. P. Deen, T. Fennell, H. Schober, A. Orecchini, S. Rols, K. A. Andersen, J. R. Stewart (The ISIS Facility, UK)

Abstract

PANTHER will build on the success of IN4, the world's most intense time-of-flight (ToF) spectrometer. A large position-sensitive detector (PSD) will improve data collection rates significantly, the background will be greatly reduced, and it will incorporate features indispensable for magnetic studies (small angles, polarisation analysis, high magnetic field devices). The new instrument will enable rapid surveys of (Q, ω) space, as well as more detailed studies in fields ranging from magnetism to the structural excitations - phonon densities of states, dispersion of collective modes and molecular vibrations – that govern the behaviour of many important physico-chemical systems.

Scientific case - magnetism

Magnetism underpins society, from information technology to electromechanical devices. It is central to the growing spintronics industry, and is likely to play a key role in quantum computing. In order to understand and ultimately exploit the materials on which such devices are based it is necessary to understand their magnetic structure and excitations, often beyond simple classical models. Seemingly exotic phenomena – for example the unusual magnetic order displayed by heavy-fermion systems, unconventional superconductors and magnetically frustrated compounds – are often essential in rationalising bulk properties of. Neutrons provide the most incisive tool to study such behaviour, directly probing the magnetic spin state of the compound in question giving us spatial and temporal information. The magnetic excitation spectrum in many of these compounds, particularly strongly correlated electron systems based on transition metals, sits firmly in the thermal neutron range, 5 - 100 meV.

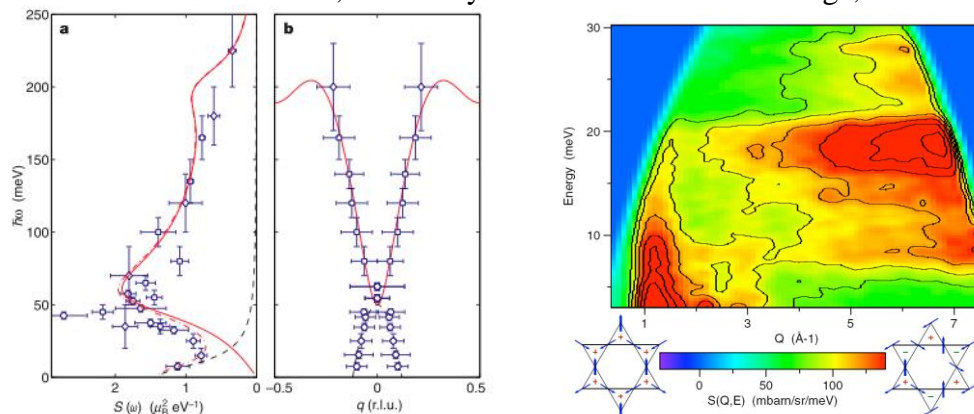


Figure 1: (a) Experimental results for integrated magnetic scattering (i) and dispersion (ii) of the excitations in the high T_c superconducting cuprate $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ [6], (b) Total $S(Q, \omega)$ of deuterium jarosite measured with $E_i = 40\text{meV}$ [3]

There are two, complementary forms of neutron spectroscopy most commonly brought to bear on such systems: triple-axis techniques that focus in on specific points in (Q, ω) space, and ToF methods more suited to surveys of (Q, ω) , and the mapping of more diffuse features. For many ‘exotic’ magnetic materials, the latter technique – particularly employing large-area detectors - is crucial in distinguishing broad weak continua from background, as in low dimensional quantum antiferromagnets [1] or spin liquids [2, 3]. Unravelling complex

dependence of scattering intensity on Q requires large reciprocal space coverage, for example in the emergent cluster ground-state in ZnCr_2O_4 [4] or strongly correlated electron systems such as $\text{La}_{5/3}\text{Sr}_{1/3}\text{NiO}_4$ [5]. Figure 1 shows two examples of (a) magnetic scattering and dispersion of magnetic excitations in the superconducting cuprates $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ [6], and (b) the magnetic scattering from the exotic spin liquid state in deuterium jarosite at low Q coexisting with high intensity phonon scattering at high Q , [3]. Detailed mapping of $S(Q,\omega)$ using ToF techniques reveals the well known resonance in high temperature superconductors [7], found in the thermal regime at $E = 41$ and 34 meV for $\text{YBa}_2\text{Cu}_3\text{O}_7$.

The power of neutron scattering for investigating magnetism is greatly extended by the possibility of more sophisticated forms of polarization analysis – particularly XYZ PA [8], which allows the unambiguous separation of magnetic from nuclear and spin incoherent cross sections. Concomitant advances in crystal growth and data analysis software mean that such studies will become the routine first step in understanding the excitations of new materials. Particular examples where this has proven essential are the recent studies of the spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$ on the cold-neutron time-of-flight instrument D7 at ILL where it allowed the extraction of subtle features of the diffuse scattering [9]. An equivalent instrument in the thermal range has not yet been developed since supermirrors used to polarise neutrons in the cold wavelength regime are highly inefficient for thermal or sub-thermal neutrons. No large area polarized neutron scattering spectrometer exists, although such an instrument is now feasible using ^3He filter polarization analysis recently developed at ILL [10, 11].

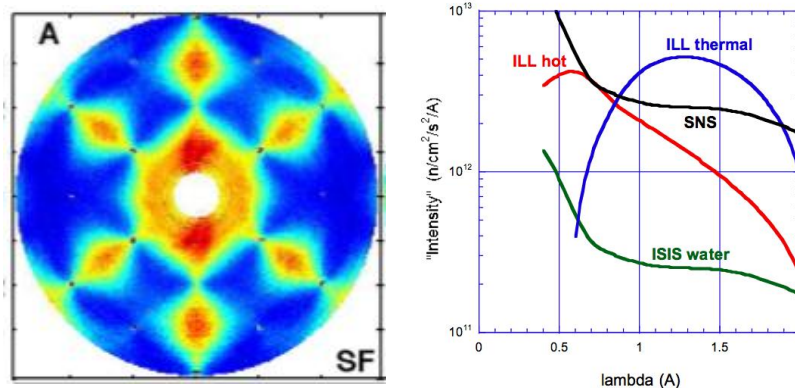


Figure 2 (left): Magnetic scattering from the spin ice $\text{Ho}_2\text{Ti}_2\text{O}_7$, [10]. **Figure 3**: Comparison of the intensities expected for a thermal ToF spectrometer at a reactor source with equivalent instruments on a 'hot' neutron guide, at ISIS and at the SNS.

Scientific case - atomic and molecular spectroscopy

ToF neutron spectroscopy is already a well-established technique to elucidate the dynamics of crystals and molecules. However, it has recently become significantly more incisive. One reason for this is the increasing use of powerful DFT based simulation techniques which allow subtle features in the vibrational spectrum to be related to dynamical coupling of phonon modes with other degree of freedom of the system [12]. Another is the technical development of instruments, with focussing optics, and large area detectors (PSD)). At the same time, there is an ever-increasing range of new materials, provided in part by developments of methods of chemical synthesis, with a broad spectrum of physical properties: multiferroics, high- T_c superconductors, nano-organized molecules, inverse expansion material, thermoelectric material, and guest-host molecular systems. An example comes from the nano-carbon molecular zoo showing an impressive diversity: carbon nanotubes, fullerenes and graphene are attracting tremendous interest based both on potential applications and the opportunities to explore fundamental principles such as the influence of dimensionality or

confinement on physical properties. Here, the IN4 spectrometer has proved especially effective on account of its high flux and wide (Q,ω) range. For example, the molecular excitations (rotations and translations) of H_2 molecules strongly confined inside C_{60} cages [13] were observed. The mobility of C_{60} confined in single-walled nanotubes has been studied on IN4 down to low temperature [14], revealing the vibration and the rotation of the fullerene peas. These experiments were performed on 100 mg and 450 mg of material respectively.

The large (Q,ω) range in the thermal region is of further importance for the study of collective dynamics in liquids, glasses and biological molecules. The determination of the dispersion curves in these systems, which is a subject of rising interest, requires a fine-tuned combination of thermal incident energies and accessible small scattering angles. The information provided by the dispersion curves needs often to be complemented by knowledge of the density of vibrational states, requiring measurement of the DOS up to thermal and higher energies. Such information is of interest to a great number of systems where the dynamic behaviour of water, for instance, plays a major role. Examples range from proteins to clays, from proton transport membranes for fuel cells to filters for water purification.

Instrument

The present version of IN4 suffers from well identified problems: significant background, no PSDs at small angle and a degraded resolution at high energy transfer in neutron energy loss. PANTHER will be a ToF spectrometer designed to remediate these issues while keeping the flux at the level of the present IN4 machine at least. PANTHER will fill a (Q,ω) range that will be intermediate between RAMSES (IN6) and IN1-LAGRANGE, fed by an intense thermal neutron spectrum, $0.8 < \lambda < 4 \text{ \AA}$. It will be designed to accept advanced sample environments (broad (P,T) range, large magnets).

The project will also take advantage of the ILL's ^3He program and will be complemented by the programme to upgrade significantly the suite of cryomagnets. Simulations [11] revealed that in the spectral window of a crystal monochromator Fermi chopper thermal ToF instrument, such as IN4, the implementation of a PASTIS insert can match a chopper spectrometer on a MW power short pulse spallation source, **Figure 3**. The PASTIS insert uses nuclear-spin-polarized ^3He gas for PA. The PASTIS insert has been conceived and designed to enable neutron PA techniques on thermal time-of-flight (TOF) spectrometers. The feasibility of the device for large-solid-angle PA of thermal neutrons has now been demonstrated through tests on IN3 at ILL. In the case of magnetic scattering it will be important to allow scattering at very low angles, where the magnetic scattering cross section is greatest, and this will require the cell to be very close to the transmitted beam. Preliminary work at ILL to improve on quartz cells currently used are very promising.

The price for such a spectrometer is expected to be in the range 2-4 M€.

References

- [1] S. Notbohm *et al*, Phys. Rev. Lett., 98 (2007) 027403 [2] M. A. de Vries *et al* Phys. Rev. Lett., submitted (2009). [3] B. Fåk, *et al* EPL, 81 (2008) 17006 [4] S.-H. Lee, *et al*, Nature, 418 (2002) 856 [5] A. T. Boothroyd, *et al*, Phys. Rev. Lett., 91 (2003) 257201 [6] J. M. Tranquada, *et al* Nature, 429 (2004) 534 [7] S. M. Hayden *et al*, Nature, 429 (2004) 531 [8] J.R.Stewart, *et al* J. Appl. Cryst., 42 (2009) 69 [9] T. Fennell Science Express Reports, Published online September 3 2009; 10.1126/science.1177582 [10] J. Stewart, *et al* Physica B, 385-386, (2006) 1142 [11] J. Stewart, J. Neut. Res., 15 (2007) 95 [12], M. Zbiri *et al*. Phys. Rev. B, **79**, 064511, [13] A. J. Horsewill *et al*, Phys. Rev. Lett, 102, 013001 (2009), [14] S. Rols *et al.*, Phys. Rev. Lett, 101 (2008) 065507