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TOSCA neutron spectrometer: The final configuration

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Abstract. The forward and backward scattering parts (phase II) of the new neutron spectrometer, TOSCA, replacing the old TFXA and TOSCA (phase I), have been successfully installed at the ISIS pulsed neutron source. The results show a significant enhancement in the counting rate due to the larger detector area. The improved resolution (to 1.5%–3% of the energy transfer) as compared with the previous instruments (TXFA: 2.5%–3.5%; TOSCA-I: 2%–3.5%) has been achieved by increasing the primary flight path from 12 m to 17 m. A chopper has been added in order to avoid neutron frame overlap and to reduce the fast neutron background. Additional diffraction capability will be installed in the near future. An example of the high resolution that is routinely available is provided by the spectrum of potassium hydrogen phtalate at 20 K.

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The second and final version of a high-resolution crystal analyser spectrometer, TOSCA, has been built, installed and operated at the ISIS pulsed neutron source (Rutherford Appleton Laboratory, Chilton, Didcot, Oxfordshire, UK) as a joint project of Consiglio Nazionale delle Ricerche (Italy), University of Kent at Canterbury (UK) and Rutherford Appleton Laboratory.

TOSCA is mainly intended for molecular vibrational spectroscopy but will also have some general diffraction capability with elastic detectors in forward and backward scattering in order to provide good coverage of the momentum transfer, Q, space (5–350 nm^{-1}). This is equivalent to the 0.02–2 nm range in d-plane spacing.

The instrument makes it possible to measure inelastic neutron scattering in a large energy-transfer range, $\hbar\omega =$ 5–1000 meV, with a relative energy-resolution of about $\Delta\omega/\omega = 1.5\%$–$3\%$ in the whole interval of $\hbar\omega$.

It has replaced both the old TFXA [1], removed in spring 1998, and subsequently a prototypical version of TOSCA (TOSCA-I [2, 3]), which was incorporated into the final version of TOSCA in spring 2000. The advantage of TOSCA over its two predecessors is improved detected flux and energy resolution. As far as the intensity is concerned TOSCA exhibits an overall gain factor of 6.3 over TFXA and 1.9 over TOSCA-I, mainly obtained through a larger detector bank area.

All three spectrometers were set to view the same water moderator, which was kept at room temperature and was poisoned at a depth of 20 mm by a thin gadolinium foil. The two earlier spectrometers, which were placed at 12 m from the neutron moderator, included two and ten back-scattering detector modules, respectively, and showed a $\Delta\omega/\omega$ of 2.5%–3.5% and 2%–3.5%, respectively (as reported in Fig. 1). Resolution calculations were experimentally tested [4]. TOSCA is sited 17 m from the water moderator and makes use of ten detector banks: five viewing forward scattering ($\simeq 47.50°$) and five viewing backward scattering ($\simeq 132.30°$) (see Fig. 2 for a schematic of TOSCA). Each bank contains 13 ^{3}He squashed tubes, the same thickness

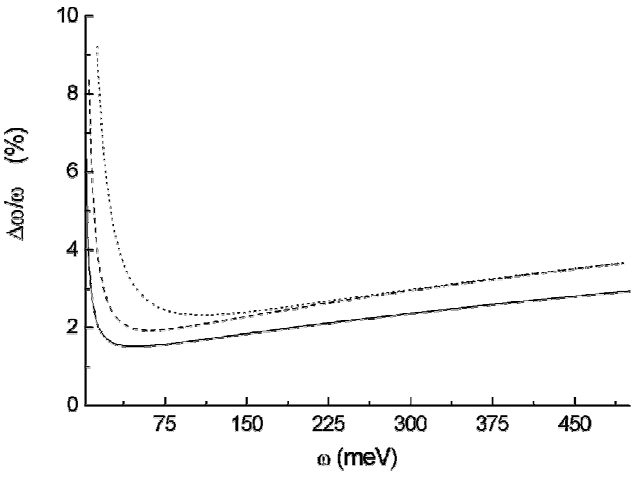

Fig. 1. Comparison of the energy-resolution functions for TFXA (*dashed line*), TOSCA-I (*dotted line*) and TOSCA (*solid line*)

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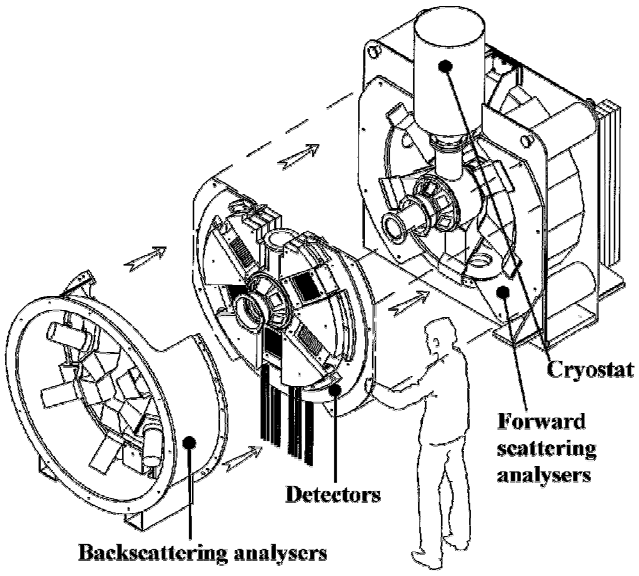

Fig. 2. View of TOSCA showing detector banks for forward and backward scattering

as in TOSCA-I (2.5 mm), but much thinner than on TFXA (6.0 mm). However, they are much longer than on TOSCA-I (from 100 mm to 250 mm) in order to compensate for the larger distance from the neutron source, but at the same time to maintain the spectrometer sensitivity. Other changes were made to the TOSCA beam-line in order to enhance the neutron flux and to reduce the instrumental background, which on TOSCA is flat and structureless, but presently slightly larger than on TFXA. The beam cross-section is now $40 \times 40\,\mathrm{mm}^2$ (was $20 \times 50\,\mathrm{mm}^2$ in TOSCA-I), and a Nimonic chopper has been installed to avoid frame overlap at the sample position and to remove fast neutrons normally associated with open-geometry machines. The whole instrument is buried in shielding composed of borated plastic of various sorts, B_4C, steel and cadmium plates. A three-head, closed-cycle refrigerator, embedded in the spectrometer, allows measurements in the range between 10 K and room temperature. A computer-controlled 24-position sample changer, based on a robust conveyor-belt system, is currently available for solid and liquid systems.

The spectrometer is of the inverted-geometry type [5] and in this respect is similar to older instruments operating at various neutron sources: ZING-P (Argonne, USA) [6], WNR at LANSCE (Los Alamos, USA) [7], CAT [8] and LAM-D [9] at KENS (Ibaraki, Japan) and NERA-PR at FNLP (Dubna, Russia) [10]. Scattered neutrons are detected at a fixed energy (between 4.1 meV and 3.5 meV) by a set of pyrolytic graphite analysers with a thickness of 2 mm and a mosaic spread of 2.5°. They are oriented in such a way as to make use of the (002) Bragg plane, which implies an interplanar distance of 0.3354 nm. The incident neutron energy is determined from the measured total time of flight, t, through the following kinematic relation:

$$t = \frac{L_0}{v_0} + \frac{L_1}{v_1}, \tag{1}$$

where L_0, L_1, v_0 and v_1 are incident (0) and scattered (1) flight-path lengths and neutron velocities, respectively. In TOSCA typical values for the neutron flight-paths are: $L_0 = 17.00$ m, $L_1 = 0.55$–0.70 m. Before neutron detection, 75-mm-thick beryllium filters, cooled to below 35 K by single-stage, closed-cycle He refrigerators, are used to suppress high-order harmonics reflected by the graphite analysers. Since the uncertainty of L_1 dominates the instrument energy resolution [5], a two-dimensional focusing of t was arranged for the detectors, in order to compensate for the errors in L_1 with the opposite errors in v_1 [5]. This configuration has been obtained in TOSCA by arranging the detector banks in a circular geometry around the beam axis and aligning the sample and each detector tube in two parallel planes, again parallel to the plane of the respective analyser (see Fig. 2). In addition, to further improve the energy resolution, the Marx principle [1] was also applied by positioning the ^{3}He tubes perpendicular to the scattering plane.

In TOSCA, owing to the low values of v_1 (and hence of final neutron momentum), Q rapidly increases with ω along a narrow stripe in the (Q, ω) kinematic space (see Fig. 3). It starts at $\omega = 0$ from $Q = 15$–$21\,\mathrm{nm}^{-1}$, then it increases approximately as $(2m_n\omega/h)^{1/2}$. However, this spectrometer will be used mainly for studying undispersed modes in solids (molecular vibrations [11], proton local modes in metal hydrides [12], etc.) or phonon densities of states [13], where the Q-dependence is not usually crucial. In TOSCA, the presence of forward and backward scattering detector banks will allow, for low values of energy transfer, the exploration of two close, but still different, (Q, ω) trajectories at the same time (as shown in Fig. 3).

An example of the high-resolution measurements possible with TOSCA is provided here by two spectra from polycrystalline potassium hydrogen phtalate (KAP). This substance is a typical example of an acid salt with a short asymmetrical hydrogen bond (0.2546 nm). Its dynamics has been extensively studied in the past through optical spectroscopy, both in the energy range of the lattice excitations (0–35 meV) [14] and in the internal mode region (35–450 meV) [15], including the vibrations of the acid phtalate ions and the stretching modes of the OH—O bonds. For this reason KAP is an ideal sample for evaluating the instrument performance in vibrational molecular spectroscopy. Two KAP samples were meas-

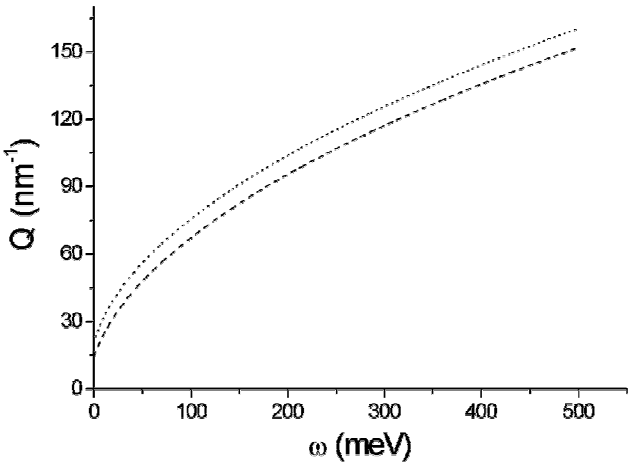

Fig. 3. Momentum transfer accessible with TOSCA in forward (*dashed line*) and backward (*dotted line*) scattering as a function of the energy transfer

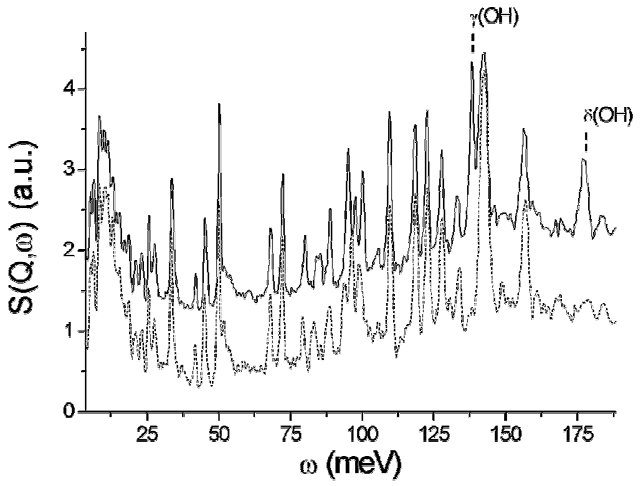

Fig. 4. TOSCA inelastic spectra from potassium hydrogen phtalate at 20 K. The fully protonated sample is shown as a *solid line*, while the *dashed line* represents a selectively deuterated one. Missing peaks clearly indicate the two hydrogen bond modes

ured with TOSCA at a temperature of $T = 20$ K: one was fully protonated, $K[C_6H_4(COOH)COO]$, while the other was selectively deuterated, $K[C_6H_4(COOD)COO]$. Their spectra are reported in Fig. 4, after a sample-can contribution subtraction and a preliminary normalisation. The high resolution of the vibrational peaks is clearly visible. In addition, by simply comparing the two patterns, two major excitations (missing in the selectively deuterated sample) are easily identified: one at 138.7 meV and the other at 177.2 meV. They can be associated with the γ(OH) and δ(OH) modes of the hydrogen bond, respectively, confirming the assignment proposed by B. Orel et al. [15]. It is possible to conclude that the new spectrometer TOSCA in its final configuration has shown excellent performance during its commissioning on the ISIS spallation neutron source, with very good energy-transfer resolution and a good signal-to-noise ratio. It has the potential to make a significant contribution to neutron vibrational spectroscopy, providing measurements with energy range and resolution almost comparable with the traditional optical (infrared and Raman) techniques.

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