

Neutron quasi-elastic scattering in materials research

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Abstract. The paper reviews the quasi-elastic neutron scattering activities at the Bhabha Atomic Research Centre and demonstrates how this technique gives information on rates and geometries of stochastic motions in liquids and solids.

Keywords. Neutron quasi-elastic scattering; stochastic motions; materials research.

1. Introduction

This article for the memorial volume of Dr N S Satya Murthy is being written with a special sense of loss. I first came to know him when we joined the Training School of the then Atomic Energy Establishment, Trombay in July 1957. We graduated in August 1958 and joined the Establishment to work under P K Iyengar who was just starting neutron scattering activities at Trombay. Observation of the first diffraction pattern at Trombay, the first phonon, paramagnetic scattering – were all exciting events for us. There were many other shared scientific and personal experiences over more than twenty five years. I have chosen here to write about the growth over this period, of a part of neutron scattering, which is still alive and active. This is a dedication to a scientist whose judgement I valued and to a friend.

1.1 *Neutron scattering*

Neutron scattering is a multi-technique capable of giving a wide variety of information on different types of materials. This can be appreciated when one recalls that a neutron can get either scattered from the nucleus or magnetically from the electrons; it can be either coherently scattered or incoherently; the scattering could further be elastic, quasi-elastic or inelastic. The power of the method comes from the fact that it is possible by the present techniques to separate each one of these scatterings and thus gain information about different properties. In this article we will discuss the scope of incoherent quasi elastic neutron scattering.

Neutron scattering is 'elastic' if, in the scattering material, there is a time averaged static distribution of atoms over a time period large as compared to neutron interaction time. Typically with the best neutron spectrometers today times larger than 10^{-7} sec can essentially be considered too large and the scattering would be 'elastic' if the characteristic time of any dynamic process is larger than 10^{-7} sec. Motions faster than this can, in principle, be observed provided a suitable spectrometer is available.

1.2 Types of motions

Motion in general can be divided into two types: periodic and random. Periodic motions give rise to inelastic scattering, that is, a part of a neutron monochromatic beam would be scattered with a finite shift in energy related to the frequency of the periodic motion in a simple way. Random motions give rise to quasi-elastic scattering, that is, there is a continuous spectrum of energy shifts on both sides of the monochromatic energy, and centered about it, leading to a broadened line. Thus, quasi elastic scattering is basically a way to study random motions.

1.3 Types of materials

There is a large variety of materials in which random motions take place. Diffusion of molecules is one example which is most easily observed in liquids. Usually though, this is too slow in solids to be comfortably observed by neutrons. There are, however, special situations where it is not so. For example, hydrogen in certain metals diffuses fast enough to be easily observed by neutrons. Superionic conductors show fast movement of certain ionic species. Diffusion of metals near melting point has also been observed. Random motions need not always be translational. Rotational reorientation occurs in molecular liquids and a large variety of molecular solids; study of these reorientations leads to information on the rotational potential in which the molecules reside. Further, there are situations where both translational and rotational motions are present at the same time. One also comes across situations where more than one type of translational or rotational motion or a combination of them may be present.

The special advantage of using neutrons to study these materials lies in the fact that it is possible to study not only the rates of diffusion but also examine the path or geometry of the diffusion.

In this article we will briefly review some experiments of this type performed at Trombay and mention a few other examples.

2. Principles

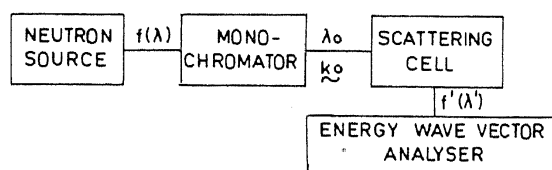
2.1 Basic interaction

Slow neutrons which typically have wavelengths in the range of 1 to 10Å are mainly scattered from the nucleus of atoms. The scattering, being essentially from a point scatterer ($\sim 10^{-13}$ cm), is isotropic. The extent of scattering, which is proportional to the square of the bound nuclear scattering amplitude, b , varies for any element from one isotope to another and also depends on the spin of the scattering nucleus. Hence, in a random assembly of atoms a configurational average, $\langle \quad \rangle$, has to be taken. This gives rise to a 'coherent' scattering cross-section, $\sigma_{\text{coh}} = 4\pi \langle b \rangle^2$ where the intensity is proportional to the square of the average scattering amplitude and an incoherent scattering which is the difference between the total, $4\pi \langle b^2 \rangle$, and the coherent scattering; $\sigma_{\text{inc}} = 4\pi [\langle b^2 \rangle - \langle b \rangle^2]$. Table 1 gives the coherent and incoherent scattering cross-sections and scattering amplitudes for some elements.

Some features of this table are worth noting: σ_{inc} for H practically dominates all scattering, σ_{coh} for H is comparable to that of other elements and the scattering amplitudes of H is negative whereas most other elements have a positive value of b .

Table 1. Coherent and incoherent scattering cross-sections and scattering amplitudes for some metals

	$\sigma_{\text{coh}} (10^{-24} \text{ cm}^2)$	$\sigma_{\text{inc}} (10^{-24} \text{ cm}^2)$	$b (10^{-12} \text{ cm})$
H	1.759	79.7	-0.3741
D	5.597	2.0	0.6674
C	5.554	0.0	0.6648
O	4.235	0.0	0.5805
Fe	11.437	0.21	0.954
Pb	11.106	0.0	0.9401

**Figure 1.** A typical arrangement for neutron scattering experiment.

These along with the other properties of neutrons, that is, their high penetrability in materials and wavelength-energy ($\lambda - E_0$) relationship similar in order-of-magnitude to that of material lattices are utilised in most neutron scattering experiments. In the problems which we will be discussing in this paper we confine ourselves to incoherent scattering from hydrogenous materials.

A typical block diagram for a neutron scattering experiment is shown in figure 1. A nuclear reactor provides a continuous source of polychromatic neutrons. A monochromator which may be just a large single crystal or a more complicated mechanical device, is used to get a mono-energetic beam of neutrons of energy E_0 having a wavelength λ_0 and wave-vector \mathbf{k}_0 ; $|k_0| = 2\pi/\lambda_0$. These neutrons are scattered from the material under study. In a diffraction experiment one measures the angular distribution of the scattered neutrons. In a more general inelastic or quasi-elastic scattering experiment both the angular (giving wavevector transfer, \mathbf{Q}) and energy distributions are measured. Thus, in the latter the scattering cross-section for wavevector transfer $\mathbf{Q} = \mathbf{k}_0 - \mathbf{k}'$ and energy transfer, $\varepsilon = \hbar\omega = E_0 - E'$, are determined. The instruments used at Trombay are a rotating crystal spectrometer which gives incident pulsed neutrons of 4.87 meV which after scattering are energy analysed by a time-of-flight technique and a triple axis spectrometer which gives neutrons of ~ 14 meV which after scattering, are analysed by means of crystal diffraction.

2.2 Cross sections

The partial differential cross section per unit energy interval per unit solid angle for incoherent scattering from an atom can be written as

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega dE'} = \frac{1}{\hbar} \frac{k'}{k_0} \frac{\sigma_{\text{inc}}}{4\pi} S_{\text{inc}}(Q, \omega)$$

where $S_{\text{inc}}(Q, \omega)$ is the incoherent scattering function corresponding to the atom from

which the incoherent scattering is being considered. If there are several different types of incoherent scatterer one has to take a sum over all the scatterers.

The scattering function has been shown by Van Hove to be the double Fourier transform over space and time of the self correlation function, $G_s(\mathbf{r}, t)$, of the incoherently scattering atom.

$$S_{\text{inc}}(\mathbf{Q}, \omega) = \frac{1}{2\pi} \int \exp[i(\hbar\omega - \mathbf{Q}\cdot\mathbf{r})] G_s(\mathbf{r}, t) d\mathbf{r} dt$$

For classical systems $G_s(\mathbf{r}, t)$ has a simple physical meaning and gives the probability of finding the atom in a unit volume at a position \mathbf{r} at time t given that it was at the origin at time $t = 0$. This interpretation allows one to calculate the scattering function in a convenient way for various physical situations.

3. Experimental results

3.1 Liquids

Some of the earliest observations of quasi-elastic scattering were those of Brockhouse (1958) on water. The group at Chalk River (Brockhouse *et al* 1963) later derived the self correlation function of hydrogen from their measurements on water. A very large number of measurements were made during these years on water. The importance of the molecular rotations was not fully appreciated in the early measurements; the observations were most often separated into quasi-elastic and inelastic parts and separately interpreted. The first measurement of quasi-elastic scattering at Trombay was on liquid CH_4 (Dasannacharya *et al* 1965) to be followed by liquid CD_4 (Venkatraman *et al* 1967) and liquid NH_3 (Dasannacharya *et al* 1972). Some of the early measurements on CH_4 in the condensed state had suggested that rotations in CH_4 may be nearly free as they are in liquid H_2 , the only other known example of a free rotor. The rotational transitions in free CH_4 are ~ 1.3 meV apart and it was possible to answer this question with the available resolution on the rotating crystal spectrometer if the experiments were performed on small enough wave-vector transfers. The first experiments showed that rotations were not free but somewhat hindered. It was possible to put this on a more quantitative basis and also relate it to infra-red and Raman scattering measurements (Venkatraman *et al* 1967) through the formulation given by Sears (1967). These measurements also showed that rotations in liquid methane were similar to that in solid methane just below the melting point where the solid is in plastic phase (See figure 2). Later, it was proposed by Agarwal and Yip (1967) that, to a good approximation, it is possible to relate the neutron spectra to the lowest order angular correlation function which can also be measured by infra-red measurements. The experiments on liquid ammonia at Trombay were analysed using this approach and the angular correlation function derived from this (see figure 3, Thaper and Dasannacharya 1974).

3.2 Solids

Though the rotational motion in solid CH_4 was observed experimentally (Dasannacharya *et al* 1965) its importance was not fully appreciated partly because the

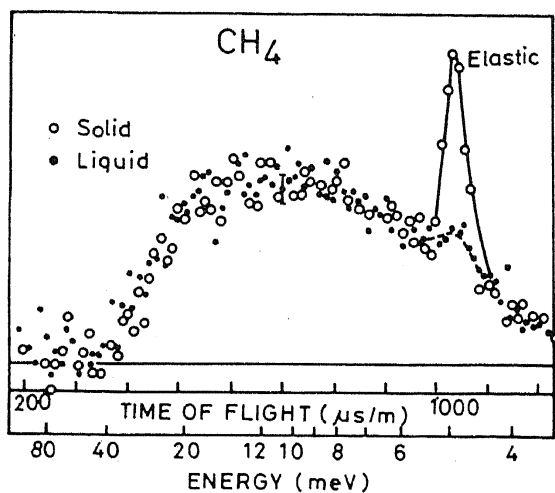


Figure 2. Spectrum of neutrons scattered from liquid (●) and solid (○) methane just above and below the melting point. The large hump indicates similarity of rotational motion in the solid and liquid states (Dasannacharya *et al* 1965).

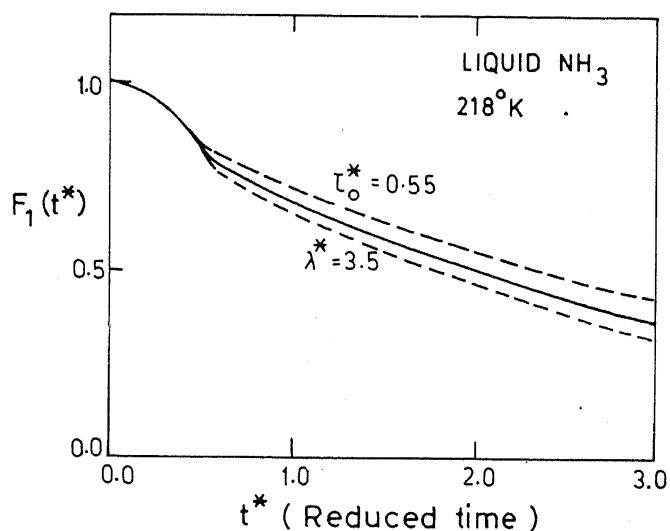


Figure 3. The first order angular correlation function for liquid NH_3 (Thaper and Dasannacharya 1974).

measurements were made with a good resolution which resulted in the elastic peak being completely separated from the quasi-elastic rotational background. Skold (1968) interpreted results of Brugger on solid CH_4 and showed that neutron spectra from molecular solids consists of a sum of an elastic and a quasi-elastic part. Almost simultaneously the results of Stockmayer and Stiller (1968) on adamantane were published. These were followed by measurements by Lechner *et al* (1969) from Argonne on neopentane and Kim *et al* (1970) from Trombay on ammonium sulphate. To understand these measurements let us look at some general features of quasi-elastic scattering from molecular solids.

The incoherent scattering from a hydrogen atom in reorienting a molecular group like NH_4 , CH_4 , CH_3 etc. is given by

$$\frac{d^2\sigma_{\text{inc}}}{d\Omega dE'} = \frac{\sigma_{\text{inc}}}{4\pi} \frac{k'}{k_0} \exp(-2w(Q)) \left[\pi A(Q) \delta(w) + \sum_p g_p(Q) \frac{f_p(Q)}{w^2 + f_p^2(Q)} \right]$$

with the normalising condition $\sum_p g_p(Q) + A(Q) = 1$. The relative intensity of the elastic and quasi-elastic part depend on the geometry of molecular reorientation. The width of the Lorentzian is related to the reorientation rate.

It is also possible to do a fixed elastic window (FEW) experiment (Brockhouse 1958) in which one detects only the elastically scattered neutrons within the resolution. Such an experiment examines the first term in the above equation along with a small contribution from the second term depending on the relative values of $f_p(Q)$ and the resolution of the instrument. This method being quick can be used for an initial survey of the problem giving information either as a function of wave-vector transfer, Q , or temperature, T , or both.

Figure 4 shows typical neutron scattering spectra when (a) characteristic reorientation time, τ , is much larger than $\hbar/\Delta E_r$, (b) when they are comparable and (c) when $\tau \ll \hbar/\Delta E_r$, where ΔE_r is the width of the energy resolution. These three conditions can be obtained in the same sample depending on temperature as shown on the right half of figure 4. A fixed elastic window experiment corresponding to the same temperature range would show a variation indicated at (e), starting from $\exp(-2W(T_1))$ at low temperature T_1 and reaching $A(Q)\exp(-2W(T_2))$ at the higher temperature T_2 , where $\exp(-2W)$ is the Debye-Waller factor.

From a detailed analysis of the spectra one measures $A(Q)$ and the characteristic times. A rough estimate of these can also be obtained from FEW experiments. We are now in a position to present some of the experimental results.

3.3 Ammonium sulphate

Ammonium sulphate is an improper ferroelectric ($T_c = 50^\circ\text{C}$); it is isomorphous with potassium sulphate which is not ferroelectric. It is, therefore, of interest to know the nature of hydrogen bonding and hydrogen dynamics across the phase transition in

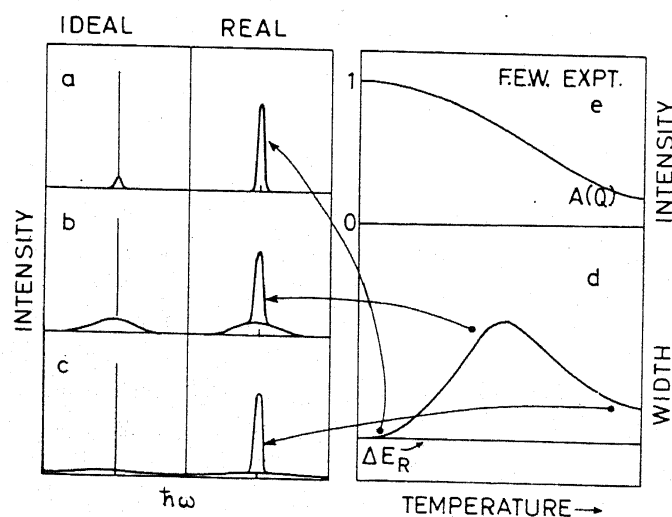


Figure 4. A typical quasi-elastic (q.e.) neutron scattering spectrum from a molecular solid consists of an elastic line and a broadened quasi-elastic line (a, b, c). If the q.e. component is comparable in width to the resolution one does not observe a broadening in a real experiment (a) so also if the q.e. component is very large compared to resolution no broadening is observed (c). The width variation is shown in (d), ΔE_R being the resolution. (e) shows the variation in intensity in a fixed-elastic window experiment as a function of temperature.

$(\text{NH}_4)_2\text{SO}_4$ and in mixed salts of $\text{K}_2\text{SO}_4 - (\text{NH}_4)_2\text{SO}_4$, which show ferroelectricity when the ammonium content is more than about 45%. There was a suggestion by O'Reilly and Tsang (1967) that the onset of ferroelectricity is connected with the cessation of flipping of ammonium ions by about 60° as ammonium sulphate goes into the ferroelectric phase whereas Schlemper and Hamilton (1966) had suggested a model based on the character of the hydrogen bond.

Quasi-elastic scattering from $(\text{NH}_4)_2\text{SO}_4$ as well as $(\text{NH}_4)_{0.16}\text{K}_{0.84})_2\text{SO}_4$ was studied at Trombay (Goyal and Dasannacharya 1978) as a function of temperature in the paraelectric phase of this material. By making measurements on these two samples it was possible to separately measure the reorientation rates and geometry of reorientation. Figure 5 shows the average form-factor for the two NH_4 and also for NH_4^+ (I) of the mixed system. The form-factors are consistent with a 120° reorientation about the NH bonds. At room temperature the reorientation times associated with NH_4 (I) and NH_4 (II) are $\sim 12 \mu\text{sec}$. The temperature dependence of the width of the quasi-elastic scattering is shown in figure 6, which is similar to the typical graph shown in figure 4(d). The activation energy for reorientation from the temperature dependence was derived to be 1.27 kcal/mole.

The resolution in these measurements was not good enough ($\Delta E \sim 200 \mu\text{eV}$) to study the ferroelectric phase. To understand this a fixed elastic-window experiment was done with a resolution of $1 \mu\text{eV}$ and the typical FEW curve (figure 7) consistent with NMR experiment was observed (Dasannacharya *et al* 1982). This resolution is actually too good to study the transition. It is, therefore, now planned to do these measurements through the transition temperature on a spectrometer with a $50 \mu\text{eV}$ resolution (Dasannacharya *et al* 1985).

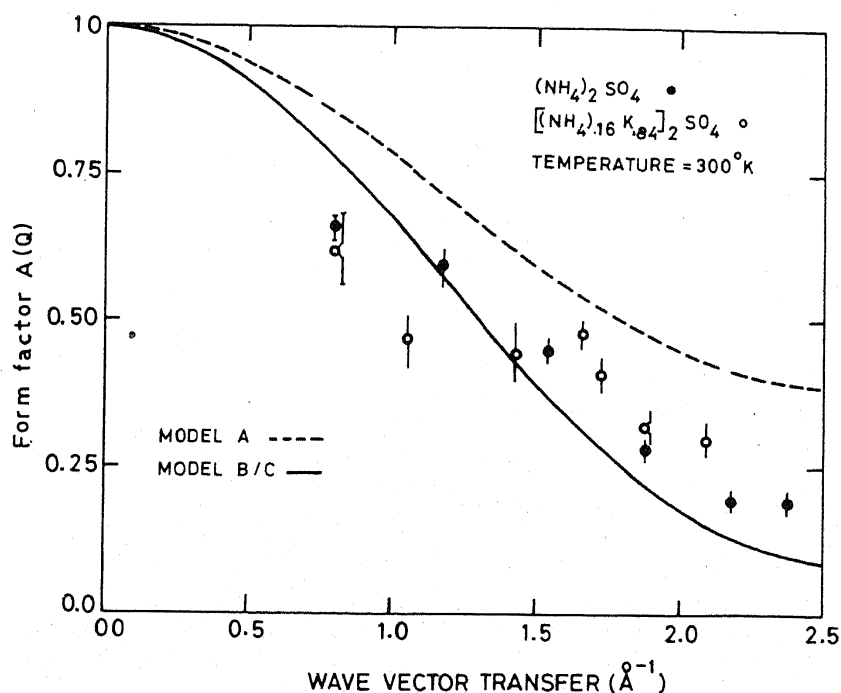


Figure 5. Average form factor of NH_4^+ ions in $(\text{NH}_4)_2\text{SO}_4$ and that of NH_4^+ (I) in $(\text{NH}_4)_{0.16}\text{K}_{0.84})_2\text{SO}_4$.

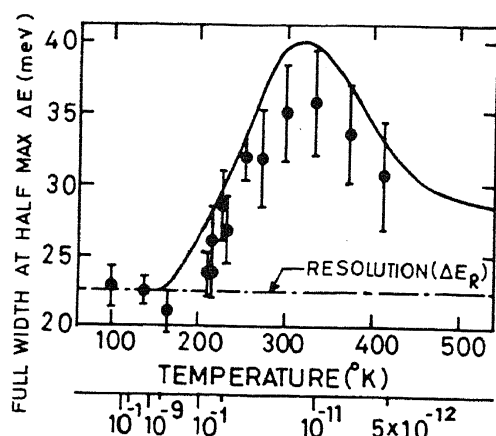


Figure 6. Temperature dependence of the width of the elastic plus quasi-elastic scattering in $(\text{NH}_4)_2\text{SO}_4$, similar to 4d.

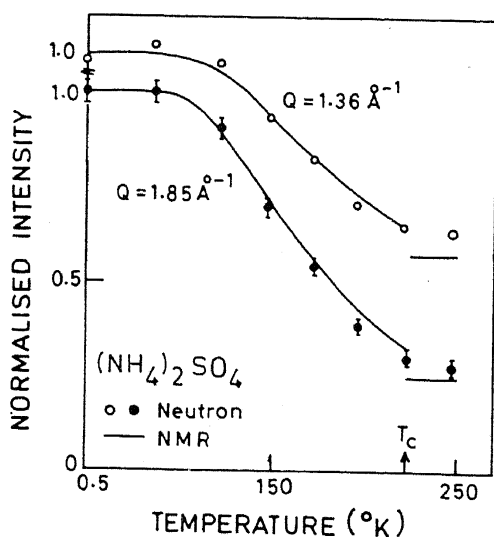


Figure 7. Temperature dependence of the fixed-elastic window experiment at $Q = 1.85 \text{ \AA}^{-1}$ and 1.36 \AA^{-1} in $(\text{NH}_4)_2\text{SO}_4$.

Similar measurements have also been carried out on $(\text{NH}_4) \text{BeF}_4$ and NH_4LiSO_4 (Goyal *et al* 1980).

2.4 Liquid crystal: $H_x\text{BPA}$

$H_x\text{BPA}$ undergoes several phase transitions. Figure 8 shows the neutron spectra measured on a rotating crystal spectrometer with a resolution of $200 \mu\text{eV}$ under different conditions, with alignment of the molecule achieved by applying a magnetic field. The spectra show broadening in the isotropic, nematic and smectic phases as well as in a supercooled smectic state at a temperature where ordinarily $H_x\text{BPA}$ is crystalline. Liquid crystals typically would have more than one rotational motion, corresponding to chain and body motions. However, body motions may be expected to be slower. If only the broadening due to chain motions are observed then the data can be analysed as a sum of an elastic part and a quasi-elastic part. The data shown in figure 8 were fitted under this assumption. The fits yield the variation of the elastic component versus wavevector transfer. If the body rotates along with the chain the variation of the elastic intensity would be much steeper than if only the chain motion were important. The variation observed suggested that the motion observed in the experiment was that of

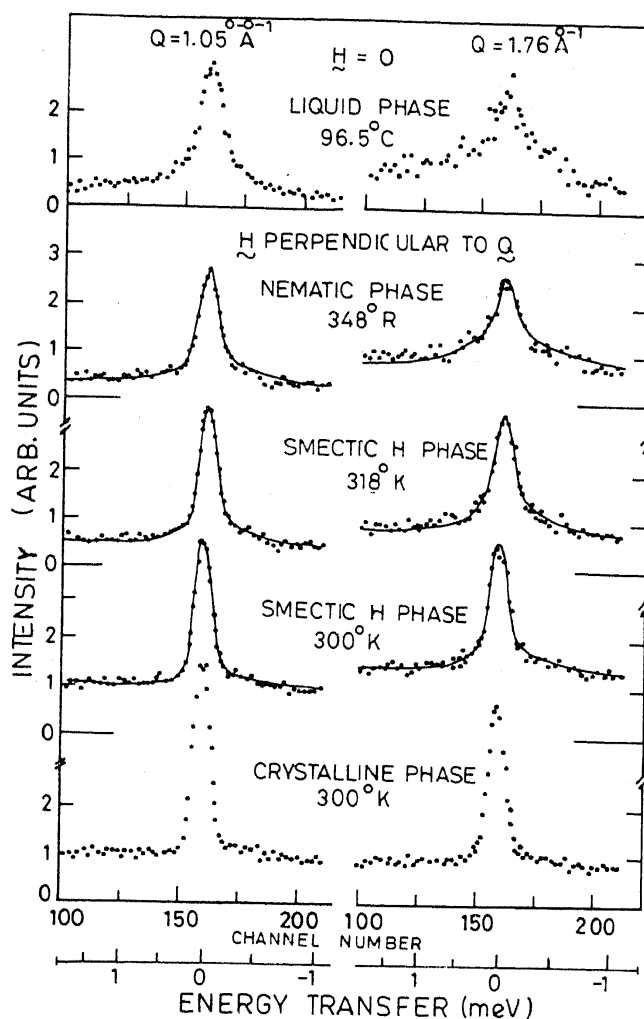


Figure 8. Spectra of quasielastically scattered neutrons from liquid crystal H_xBPA .

the chain and not of the body plus chain, justifying the analysis *a posteriori*. A residence time in the range 10^{-10} to 10^{-11} sec was derived for the chain motions from these experiments. This was one of the early measurements on various phases of a liquid crystal under a magnetic field (Sinha *et al* 1974). To observe the body motion much higher resolution, $< 10 \mu\text{eV}$, would be required. Such measurements have been performed extensively at the ILL, Grenoble.

2.5 Ammonium halides

Ammonium halides undergo a number of phase transitions. The highest temperature phase in NH_4X ($\text{X} = \text{I}, \text{Br}, \text{Cl}$) is a Na-Cl type phase which transforms to a disordered CsCl type phase. In both these phases the NH_4^+ ions reorient. A number of neutron experiments have been performed on these halides at Trombay (Goyal *et al* 1973). The highest temperature phase is of particular interest since in this phase the tetrahedral ammonium ion resides in first neighbour octahedral surroundings (Dasannacharya 1979). This allows a large number of possible orientations for the NH_4 ion, simulating a typical plastic crystal. Further, it is possible to quench the phase transitions to the CsCl

type and other lower temperature phases by incorporating NH_4 into KBr, KI or KCl lattices, thus allowing one to study this highly orientationally disordered phase right down to very low temperatures. To understand the nature of this reorientation it is necessary to make a composite study of the diffraction, inelastic scattering and quasi-elastic scattering.

A preview of NH_4Br , $(\text{NH}_4)_{0.16}\text{K}_{0.84}\text{Br}$, $(\text{NH}_4)_{0.16}\text{K}_{0.84}\text{I}$ and NH_4I was carried out (Goyal and Dasannacharya 1979) using fixed-elastic-window experiments using a triple axis spectrometer. This is shown in figure 9. It is noticed that the drop in intensity for NH_4Br , at 300°K in the CsCl phase, is gaussian and similar both for experiments with resolution of 1.3 meV and 12 meV showing that reorientation in this phase is much slower than what can be observed with this resolution. On the contrary the steep non-gaussian fall of all other specimens indicates a comparatively faster reorientation which is only slightly dependent on temperature (see KI). The fits to the data suggest a residence time of NH_4^+ ions ~ 5 psec.

These results were further quantified by detailed quasi-elastic scattering experiments. Some examples of which are shown in figure 10 (Goyal and Dasannacharya 1979). The results of these measurements were analysed on the basis of several models. Figure 11 shows the anticipated intensity of the 'elastic' part of the scattering as calculated on the basis of these models compared with the experiments. The data are consistent with rotational diffusion (h) and octahedral jump models (c, e and f). However, the analysis of the temperature dependence along with diffraction and inelastic scattering data show that none of them really describe the correct situation with regard to the geometry of reorientation. Inelastic scattering measurements at 110°K , in the mixed salts (figure 12, Dasannacharya 1979) clearly showed that librational modes exist (they were not observed in NH_4I at room temperature due to temperature broadening) ruling out pure

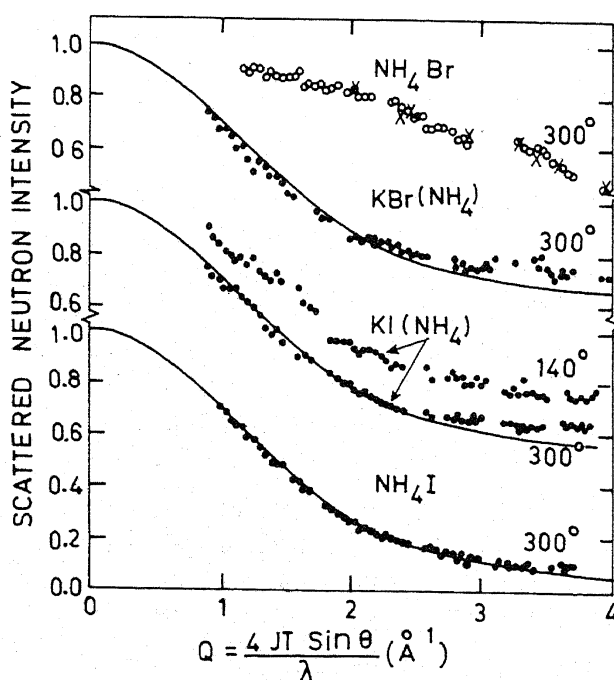


Figure 9. Fixed elastic window experiment on various ammonium halides. (●) with $\Delta E_R \sim 0.22$ meV, (o) with $\Delta E_R \sim 1.3$ meV and (x) with $\Delta E_R \sim 12$ meV.

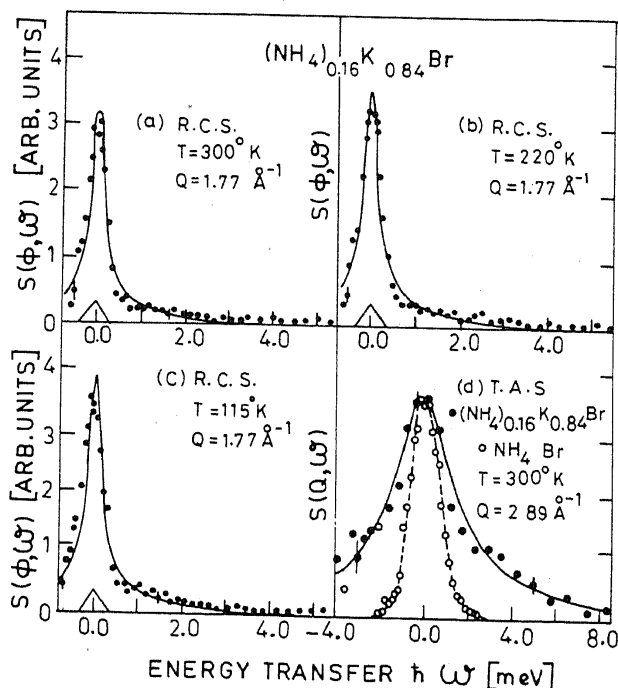


Figure 10. Typical spectra of neutron scattering from NH_4Br and $(\text{NH}_4)_{0.16}\text{K}_{0.84}\text{Br}$. Note that NH_4Br on triple axis spectrometer with $\Delta E_R \sim 1.3\text{ meV}$ (o) shows no broadening in the disordered CsCl phase whereas a broadening is seen in the mixed salt (●) which has a NaCl type structure.

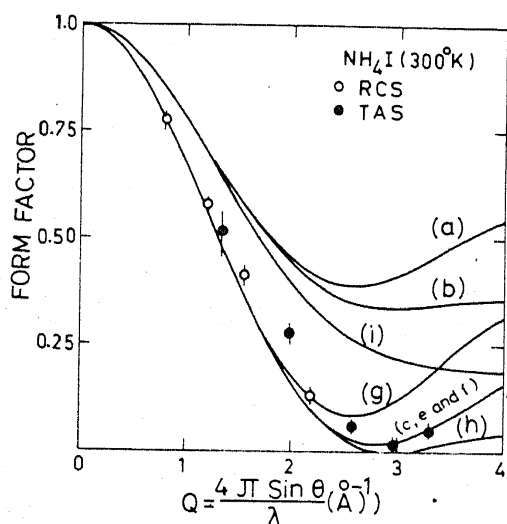


Figure 11. Form factor of NH_4^+ in $\text{NH}_4\text{I}(0)$ compared with several theoretical models.

rotational diffusion. At the same time the lack of strong temperature dependence between 110°K and 300°K strongly favours against 90° octahedral jumps which must require large energy. These contrary features along with the result of diffraction which shows peaking of hydrogen density along $[100]$ directions can be interpreted using the following model.

Assume that a hydrogen (H_1) points along one of the $[100]$ directions. During this period the other three hydrogens perform a rotational diffusion on a circle per-

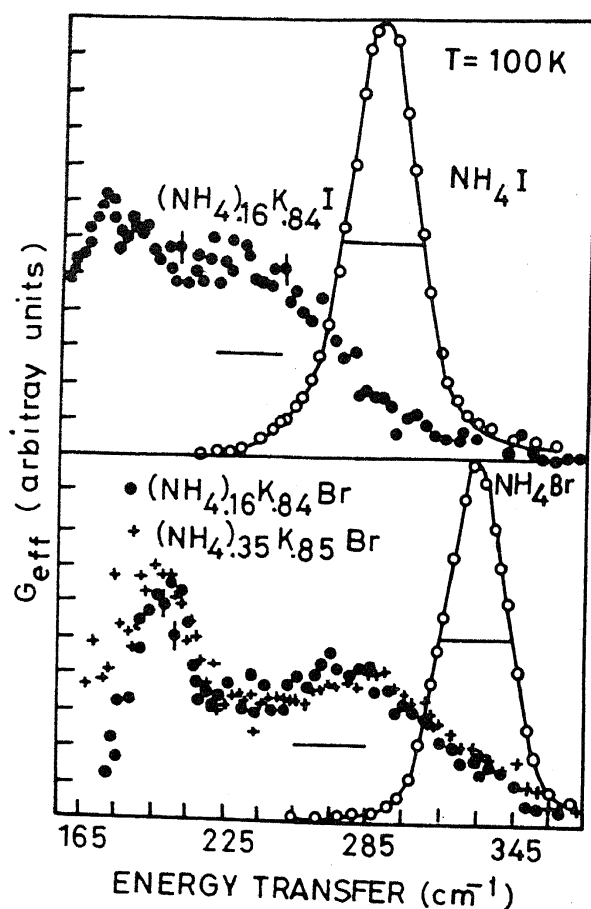


Figure 12. Inelastic scattering from pure and mixed salts as indicated. The mixed salt shows a librational peak at a larger energy transfer and a translation peak at a smaller one.

pendicular to this bond, while NH₄ librates about N-H₁. However, when one of the hydrogens (H₂, H₃ or H₄) is at the shortest angle away from one of the [100] directions it say (H₂) finds it quite easy to flip by just 19.46° and align itself along a [100]. Now NH₂ librates and H₁, H₃ and H₄ rotationally diffuse. Thus diffraction, inelastic and quasi-elastic scattering all find a plausible explanation in this model.

These examples show how reorientation rates as well as detailed geometrical information can be obtained from quasi-elastic neutron scattering experiments.

2.6 Miscellaneous remarks

Experiments at Trombay have been confined to hydrogenous substance so far due to reasons of available intensity and to comparatively faster reorientation which can be studied with a resolution of 0.2 meV. Both these restrictions can be removed. For example, quasi-elastic scattering measurements have been made on single crystals of Na (Goltz *et al* 1980) near melting point to study its self diffusion and on superionic (α) and normal (β) phases of silver iodide (Eckold *et al* 1976). The back-scattering and the spin-echo spectrometers have resolutions $< 1 \mu\text{eV}$ and the recently introduced ΔT -window spectrometer with a resolution $50 \mu\text{eV}$ is a bridge between the high and ordinary resolution spectrometers. With the availability of a better neutron

source in the form of the Dhruva reactor it would be possible to undertake a wider variety of experiments now at Trombay. Satya Murthy was leading the neutron scattering effort at Dhruva and it is unfortunate that he did not live to see its completion. This effort is being completed by his colleagues and, without naming all of them individually, this article is dedicated to his memory as much on their behalf as my own.

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