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NEW DIRECTIONS IN NEUTRON DIFFRACTION

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Abstract

The possibility of greatly improving or extending existing types of research and of opening quite new fields of research occur when new intense neutron sources are produced. Two examples will be given in the field of neutron diffraction from isotropic materials. These examples have been chosen both because of their scientific interest and because they illustrate the continuing need to stretch the capabilities of present accelerator and target designs.

Introduction

The advent of a new neutron source usually allows the users to open up new areas of study not accessible previously (e.g. the new developments at the Institut Laue Langevin), and the new generation of pulsed sources is expected to provide such opportunities. Some possible examples have been given in previous talks, but these have involved mainly a modest extrapolation of existing work. In this paper I will consider some more extreme possibilities.

Papers about neutrons have been published for some 50 years, and almost every conceivable speculation concerning their uses has been made. The versatility and utility of neutrons in the study of condensed matter is fully understood, but many of the experiments have fallen far short of the ideal neutron experiment envisaged in the speculations. Most experimentalists do what is possible as a first approximation to what is needed! Consequently from the ideal point of view we can categorise most results as coming from "bad experiments". The advent of a new source not only allows much needed improvements to be made, but also allows radical changes in experiments and some new directions to be opened. To discover which of these opportunities might become possible, we have to fit the properties of the new source to the speculations of the past. In the following discussion I shall present two examples of this fit.

High Energy Neutron Diffraction

Neutron diffraction experiments on liquids and glasses are usually carried out with wavelengths of about 1 A ($\sim$ 80 meV) largely because the intensity is high at this wavelength and results are not too bad. But it is well known that ideally the results would be improved if the energy were higher. Energies of $\sim$.3 eV have been used for some work and somewhat higher proposed: but what energy (E) is really needed? I will consider three arguments related to this question.

The first concerns the structure of liquids, non-crystalline biological materials and chemical solutions all of which may contain H or D. Because hydrogen has a mass almost the same as the neutron there is a significant neutron energy change (ΔE) on scattering. A correct diffraction experiment requires $\Delta E << E$, and since this is not achieved the observed data are corrected to make them correspond to this limit. This correction is accomplished by ad hoc models and usually amounts to many times the quantity being sought (namely the interference term in the cross section). The relative size of the correction at any given value of the momentum transfer ($\hbar q$) can be reduced by using a higher energy, because the given momentum transfer now corresponds to a lower angle of scatter. At high energies the principal effect is recoil and it is easy to show that a 1% correction to the cross section occurs at an angle (θ) of 8^o. One might say that reasonable results occur when the correction is the same magnitude as the signal. Since the interference term may be $\sim$1% of the self term at 10 A^{-1}, this means q $\sim$ 10 A^{-1} must occur for $\theta \sim 8^o$, or E $\sim$ 100 eV.

The second example concerns the measurement of the pair correlation functions, g(r), for materials in which the nearest neighbour peak is very sharp (and may be separated from the remainder of g(r) and also have some structure). This probably occurs for a number of metallic glasses and simple molecules. If we ask for a resolution in r space of $\sim$.05 A we need data to q $\sim$ 100 A^{-1}, and for a peak in g(r) at r = d $\sim$ 1 A the intensity (i.e. (Sin qd)/qd) in the diffraction pattern will have fallen to 1% at 100 A^{-1} compared to its value at $q \to 0$. Thus for this case it is useful to take data to 100 A^{-1}. It is still necessary to keep recoil corrections small but since the elements involved in many such samples are heavier than H, angles up to $\sim 25^o$ may be accepted and then E is $\sim$ 100 eV.

The final point concerns the general interpretation of the experimental condition $\Delta E << E$. Recoil of heavy elements at large angles will transfer several eV for E $\sim$ 100 eV. Since this should be enough to break chemical bonds it is sufficient, and suggests that $\sim$ 100 eV is a maximum requirement. More generally the energy transferred in neutron scattering can induce chemical reactions in the sample, for example we can knock a proton from one water molecule to another:

$$2H_2O \to H_3O^+ + OH^-$$

which requires about 3 eV. Since this is a light element sufficient recoil energy will be found at a low angle and an unusual shape will be seen in the cross section in the region where normal diffraction effects are seen also. Thus for samples in which chemical reactions might occur we need E >> 3 eV to avoid this type of distortion: that is E $\sim$ 50 to 100 eV. But if a "diffraction" pattern was recorded for E $\sim$ 3 eV, it would differ significantly from that observed for E $\sim$ 100 eV and the difference might be used to determine some properties of the chemical reaction. This opens up a new field of study ultimately leading to different experiments which determine $S(q,\omega)$ for the chemical reaction.

To perform neutron diffraction experiments of this kind requires an intense source for 50 < E < 150 eV, and pulsed spallation sources may meet this need. Because these neutrons are travelling at $\sim$ 7 μsec/m, short pulses $\sim$ 0.2 μsec are needed but the repetition rate may (if desired) be up to several kilohertz. In addition new instruments would be needed: Dr. K. Suzuki (Tohoku University) and I have designed one for a source similar to the S.N.S. and it is shown in figure 1. Since it is desirable to avoid contamination with lower energy neutrons we propose using a broad capture resonance in the detector. At present the most suitable appears to be cobalt at 132 eV, although the energy is perhaps twice as high as we

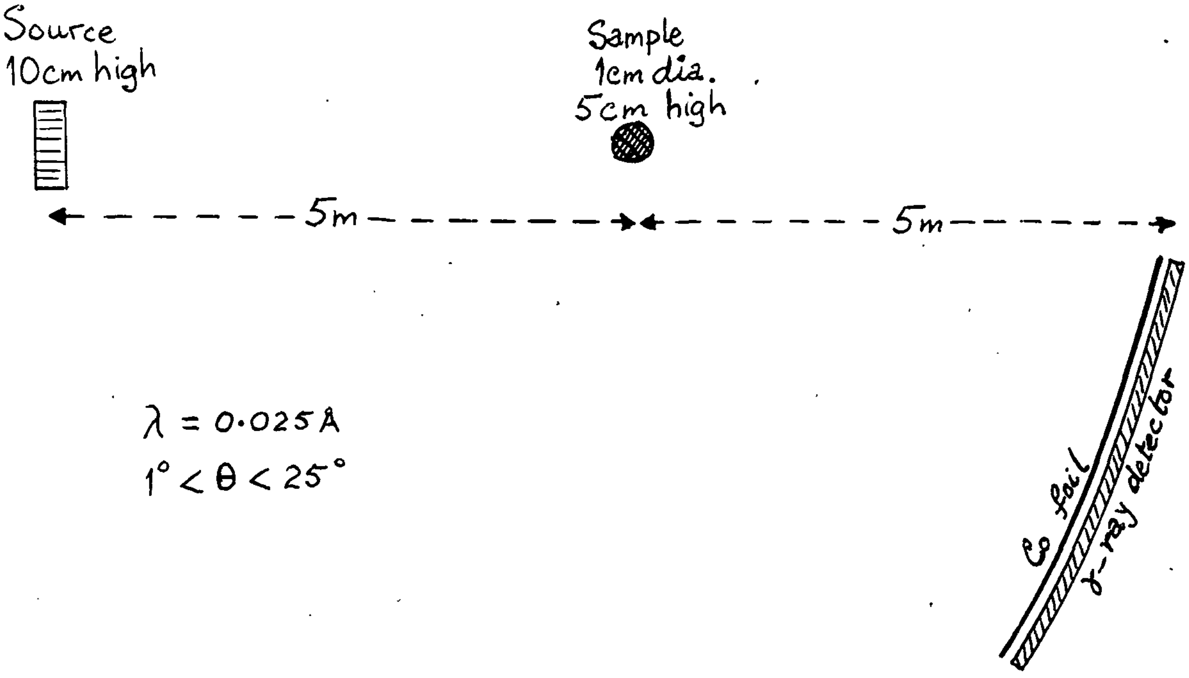

Figure 1. Single Energy Neutron Diffractometer for Amorphous Investigations (SENDAI): an instrument using T-o-F to select an energy in the neighbourhood of 100 eV, and which allows a high quality conventional fixed energy variable angle diffraction pattern to be measured out to $\sim$100 A^{-1}.

would like and there is a significant amount of scattering in this resonance. At the 100 barn level the width is 132±25 eV, and so a 1 mm thickness is appropriate (60 time channels of 0.2 μsec and a path of 10 m will cover 132±25 eV). Capture γ-rays would be detected by a position sensitive detector of large size (2 m long by 0.2 m high and 5 mm position resolution). Each time channel of 0.2 μsec would correspond to a well defined energy (such that $\Delta q \sim .5\ A^{-1}$ at 100 A^{-1}), and thus each time channel would give a structure factor of the convential kind with fixed energy and variable angle. For this reason it's called the Single Energy Neutron Diffractometer for Amorphous Investigations (SENDAI). After data reduction the 60 sets would be averaged, and added to data for $q < 4\ A^{-1}$ obtained at low angles in the present day type of diffractometer. Some other details are given in the figure. Assuming that the detector can be made 10% efficient, we estimate that $\sim 10^5$ counts for each of 400 angular positions would be obtained in 24 hours operation at the S.N.S. This study was done to demonstrate the feasibility of diffraction work using $E \sim 100$ eV, although pilot experiments with energies of 20-60 eV and conventional detectors giving lower rates may offer a better starting point. Also this would allow complementary issues, such as whether resonances can be exploited for anomalous dispersion experiments, to be explored.

Single Pulse Diffraction

The use of pulsed sources to study transient behaviour has been discussed frequently and some experiments have been undertaken in a number of laboratories. I will call these experiments "weak transient" studies since the sample is not damaged significantly by the transient and it may be exercised over a very large number of cycles. However, "strong transient" work is perhaps of greater interest, since in such experiments a single transient damages or destroys the sample which is taken to the experimental limit of pressure, temperature etc. Examples are provided by the impact of projectiles, intense shock waves, implosions etc. or by giant laser pulses, large electrical discharges etc. It is desired to study the state of matter as the transient progresses through the sample and up to the point at which the sample is severely damaged. In such experiments some data must be taken with a single neutron pulse, although one can imagine samples being recharged at the rate of several per minute down to $\sim$ 1/day. In any event the whole experiment (both time and position behaviour through the transient) might have to be completed with 10-100 samples. One might also argue that the proton target could be changed after 100 pulses.

Paper studies of accelerators giving intense pulses have been made at Triumf in connection with the Kaon factory proposal. TRINS 2B (Triumf Intense Neutron Source 2B) for example would deliver a 3 GeV proton beam with $\sim 10^{15}$ p.p.p. at 0.5 Hz. A better solution would be $\sim 10^{16}$ at 0.1 Hz, as calculations (using existing general purpose diffractometers as a base) suggest 10^{15} p.p.p. is the lower limit for such experiments. At this limit single pulse data (with modest statistical errors) could be taken on a relatively large sample (20 cm^2 beam area) of a simple crystalline material which would give a set of strong

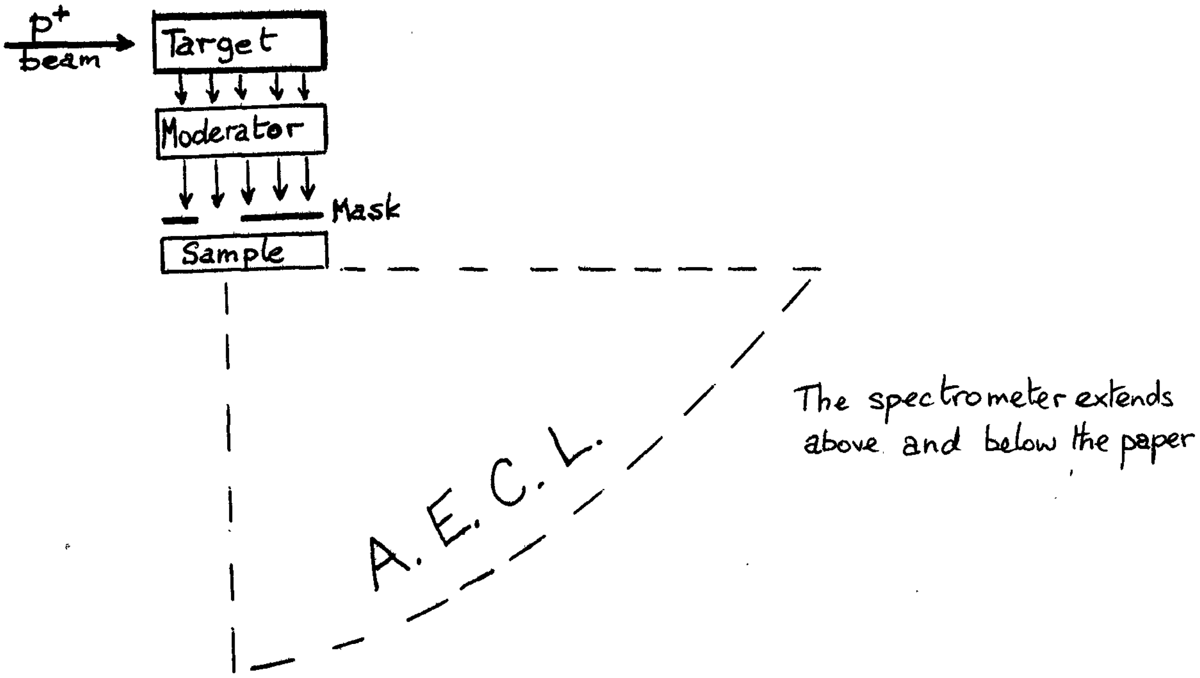

Figure 2. Angles and Energies Collected without Limit (AECL) Spectrometer used to measure the diffraction pattern of a point on a polycrystalline sample, which is being destroyed in a strong transient phased in time with the proton pulse.

Bragg reflections. The alteration in these reflections would be studied as the transient proceeds.

In the first method (figure 2) the sample is placed close to the moderator so that a pulsed white spectrum of neutrons passes through it at a given (time) phase relationship to the transient. With subsequent samples the phase could be altered. To obtain adequate statistics all wavelengths and angles of scatter should be collected and the data reduced to a single intensity vs. momentum transfer curve. Moreover fast electronics and detectors would be needed to handle the high peak counting rates (which are made manageable by the spread in time of the neutron pulse over the flight path). A new Angles and Energies Collected without Limit (AECL) spectrometer should be designed for this purpose. Initial calculations suggest this instrument is technically feasible, although the problem of making fast enough 'neutron only' detectors is difficult. If sufficient intensity is available the sample would be masked so that a pre-chosen region of it is irradiated and the masks moved for subsequent samples.

The above method has two disadvantages, the background will be high with the sample so close to the moderator, and many different samples are required to obtain phase shifted data. A simple way to overcome these disadvantages is to place the sample several metres from the source so that collimators may reduce the background and the spreading in time of the different wavelengths may be used to stobe a single sample. A number of diffraction patterns, one for each wavelength, is obtained to give the change in structure with time. Unfortunately this requires higher intensity, perhaps $\sim 10^{17}$ p.p.p, at which stage the performance of the proton target may be a limiting factor.

Conclusion

The advent of a new generation of spallation sources will allow new ground to be broken in the field of neutron diffraction. Two examples were considered in this paper, but others are to be expected. In the first example an intense average output was required and the pulse repetition frequency could be relatively high (kHz). In the second example intense single pulses were required at a very low repetition rate. From the point of view of the accelerator designer or target designer this look into the crystal ball has presented two opposite limits and both of them need to be explored.