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Polarisation Modulated Crosscorrelation Neutron Spectroscopy using ^{3}He Polarising Filters on a Pulsed Source

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Abstract

A polarisation modulated crosscorrelation spectrometer using polarised ^{3}He spin filters is outlined. Current developments in polarised ^{3}He technology indicate that this spectrometer will be viable on the third generation pulsed neutron sources now being planned, providing a novel spectroscopic tool for magnetic studies. This paper focusses on discussing the optimisation of the performance of the ^{3}He polarising filters.

1. Introduction

The concept of extending the crosscorrelation neutron time-of-flight (TOF) technique from reactor to pulsed sources was first explored experimentally by Kroo et al. [1], and theoretically by Matthes [2]. The method relies on the use of a pseudorandom modulation device placed before the scatterer which allows the scattered neutron flight time to be measured; this together with the total TOF measurement enables the incident and scattered neutron wavevectors (and energies) to be determined. The first modulating devices were disc choppers on which a pseudorandom sequence was etched in the neutron absorbing material.

Cywinski and Williams [3] proposed an alternative cross-correlation method, polarisation (or spin) modulated crosscorrelation spectroscopy, based on a pulsed source total scattering polarisation analysis instrument with ^{149}Sm filters as the polariser and spin analyser. A limiting feature of ^{149}Sm polarising filters is that they are only effective over a modest neutron energy band, ~ 1 - 100 meV. Instead of modulating intensity (as with choppers) the polarisation state of the incident neutrons is modulated pseudorandomly using a pulsed spin flipper placed in front of the scatterer. Subsequent crosscorrelation of the measured intensity with the modulating sequence provides a TOF spectrum which is the difference between the TOF spectra of neutrons with the two spin polarisations. The technically demanding components of the instrument are the broad-band polarising filters.

It is now opportune, following the spectacular advances achieved in developing ^{3}He polarising filters [4], to review the potential of spin correlation spectroscopy, particularly in relation to instruments for third generation (Power > 1Mw) spallation neutron sources. This paper focusses on examining the ^{3}He polariser properties required for a spin crosscorrelation spectrometer.

2. Spin Crosscorrelation Spectroscopy - Basic Principles

A schematic diagram of the basic components in a pulsed source spin crosscorrelation spectrometer is shown in Figure 1. A background chopper is recommended to minimise fast neutron moderation and scattering in the bulky environment surrounding the scatterer. It is important to maximise the solid angle covered by the spin analysers, and the feasibility of building banana-shaped ^{3}He cells which analyse over ± 90° in the equatorial plane has already been demonstrated [5]. Spin flipper efficiencies approaching 100% over broad neutron energy bands are now commonplace, see ref. [6], and Badurek [7] has built very

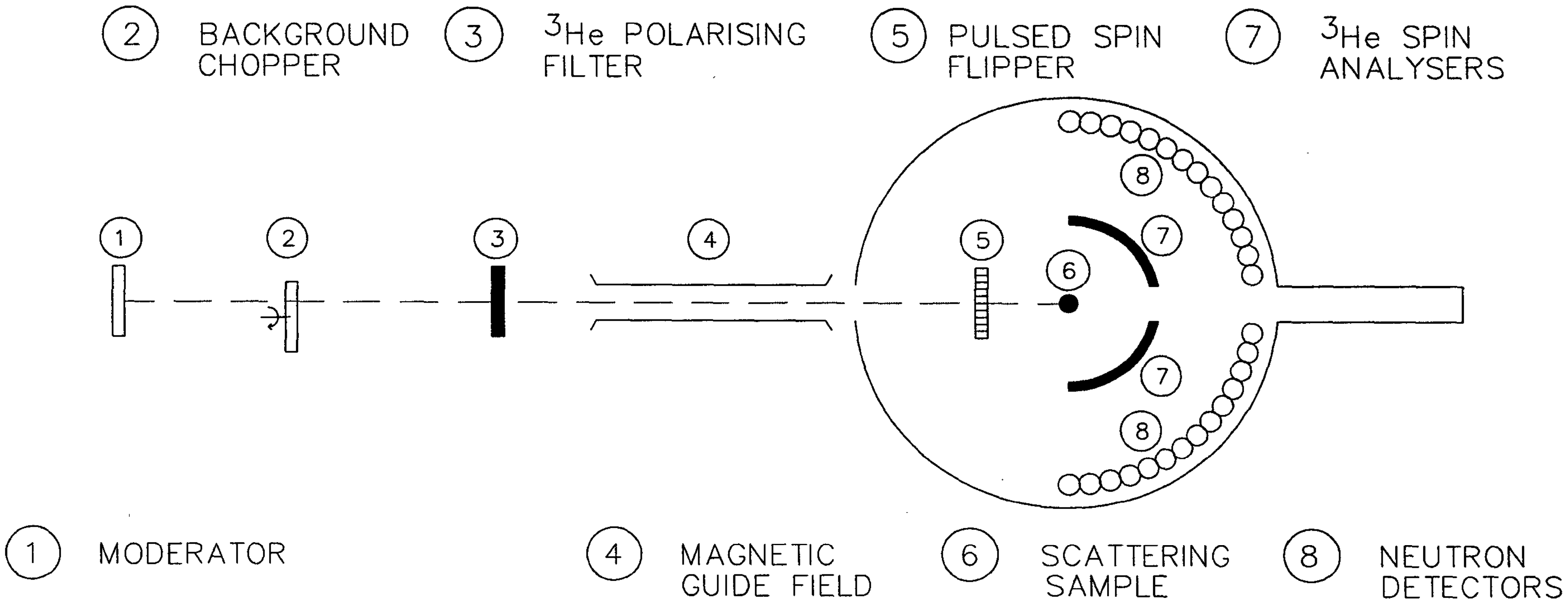


Figure 1: Schematic layout of a pulsed source spin crosscorrelation spectrometer.

compact pulsed spin flippers with switching times ~ 1μs which would be ideal for this application. Modulating sequences can be readily changed electronically and it is straightforward to provide the flipper with a pseudorandomly varied phase with respect to the source pulse.

Expressions for the momentum transfer ($\hbar Q$) and energy transfer ($\hbar\omega$) ranges, as well as their resolutions for this type of spectrometer were derived by Cywinski and Williams [3]. The larger neutron energy range and analysed solid angle range of ^{3}He filters compared with ^{149}Sm filters means that the accessible dynamic range ($\mathbf{Q}$, ω) of this spectrometer is much larger than that indicated in the original publication. Major contributions to the $\hbar\omega$ and Q resolutions derive from terms which are proportional to $1/L_2^2$, where L_2 is the scatterer/detector distance. There are similar terms containing $1/L_1^2$, where L_1 is the primary flightpath, and we suggest that L_1 and L_2 should be made as equal as possible. L_1 ~ 10m, L_2 ~ 4m is a more optimum geometry than L_1 = 12m, L_2 = 2m as given in reference [3].

Figure 2 shows the distance-time diagram for the spectrometer and indicates the method of time-labelling; the detected events illustrated represent (1) energy loss, non-spinflip, (2) elastic, spin flip and (3) energy gain, non-spinflip scattering (for the same type of polariser and analyser). A neutron arriving at the detector at time τ can be characterised by two time labels (τ, t_2). τ is the total TOF from moderator to detector , and t_2 is provided by the bit of the spinflipper pseudorandom modulating sequence which is current at the time of arrival of the neutron at the detector. t_2 is an intermediate time label through which the sample to detector TOF τ_2 is recovered by the crosscorrelation procedure; the incident neutron TOF is therefore $\tau_1 = \tau - \tau_2$.

Cywinski and Williams [3] showed that, in essence, the spin crosscorrelation method is a switching between two transfer functions:

the spinflipper ‘off’ transfer function, representing mainly non-spinflip scattering,

$$N_{\tau,\tau_2} \propto \tfrac{1}{2} T_1(\tau_1) T_2(\tau_2) \left\{ \left(\frac{d\sigma}{d\Omega}\right)^{TOT}_{\tau,\tau_2} + P_1(\tau_1) P_2(\tau_2) \left[\left(\frac{d\sigma}{d\Omega}\right)^{NSF}_{\tau,\tau_2} - \left(\frac{d\sigma}{d\Omega}\right)^{SF}_{\tau,\tau_2} \right] \right\} \qquad (2.1)$$

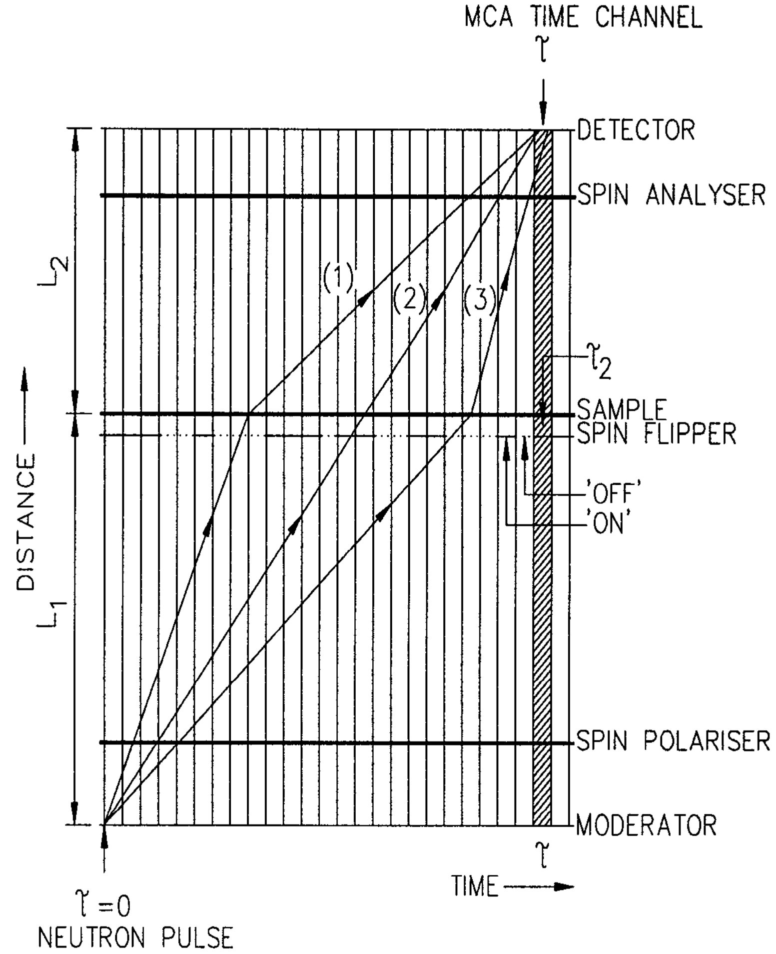


Figure 2: Distance-time diagram for a pulsed source polarisation modulated crosscorrelation spectrometer.

where NSF, SF and TOT refer to the non-spinflip, spinflip and total differential cross-sections, P and T are the polarising efficiencies and unpolarised beam transmittances of the filters (see section 3.2) with subscripts 1 and 2 denoting the values of these parameters in the incident and scattering beams respectively, and

the spinflipper 'on' transfer function, representing mainly spinflip scattering,

$$S_{\tau,\tau_2} \propto \tfrac{1}{2} T_1(\tau_1) T_2(\tau_2) \left\{ \left(\frac{d\sigma}{d\Omega}\right)^{TOT}_{\tau,\tau_2} + P_1(\tau_1) P_2(\tau_2) \left[1 - 2f(\tau_1)\right] \left[\left(\frac{d\sigma}{d\Omega}\right)^{NSF}_{\tau,\tau_2} - \left(\frac{d\sigma}{d\Omega}\right)^{SF}_{\tau,\tau_2} \right] \right\} \qquad (2.2)$$

where $f(\tau_1)$ is the spin flipper efficiency.

The difference function, which is measured in the crosscorrelation experiment, is

$$D_{\tau,\tau_2} = S_{\tau,\tau_2} - N_{\tau,\tau_2} \propto f_1(\tau_1)P_1(\tau_1)P_2(\tau_2)T_1(\tau_1)T_2(\tau_2)\left[\left(\frac{d\sigma}{d\Omega}\right)^{SF}_{\tau,\tau_2} - \left(\frac{d\sigma}{d\Omega}\right)^{NSF}_{\tau,\tau_2}\right] \tag{2.3}$$

$$\propto G(\tau_1,\tau_2)\left[\left(\frac{d\sigma}{d\Omega}\right)^{SF}_{\tau,\tau_2} - \left(\frac{d\sigma}{d\Omega}\right)^{NSF}_{\tau,\tau_2}\right] \tag{2.4}$$

where $G(\tau_1, \tau_2)$ encapsulates the spectrometer dependent terms. One method for optimising the polarising filter parameters in spin crosscorrelation spectrometers is to maximise this G term. This approach is developed later in section 3.2.

An alternative approach is to maximise the ratio R_{τ,τ_2} of the difference counts in a particular channel to the "background of ignorance" due to the sample scattering. This ratio is given by

$$R_{\tau,\tau_2} = (1-c)D_{\tau,\tau_2} \Big/ \left(c\sum_{\tau_2} S_{\tau,\tau_2} + (1-c)\sum_{\tau_2} N_{\tau,\tau_2}\right) \tag{2.5}$$

where c is the duty cycle of the pseudorandom sequence. For a duty cycle c = 0.5 and a perfectly efficient spinflipper (f=1) this ratio becomes

$$R_{\tau,\tau_2} = \frac{P_1(\tau_1)P_2(\tau_2)T_1(\tau_1)T_2(\tau_2)\left[\left(\frac{d\sigma}{d\Omega}\right)^{SF}_{\tau,\tau_2} - \left(\frac{d\sigma}{d\Omega}\right)^{NSF}_{\tau,\tau_2}\right]}{\sum_{\tau_2} T_1(\tau_1)T_2(\tau_2)\left(\frac{d\sigma}{d\Omega}\right)^{TOT}_{\tau,\tau_2}} \tag{2.6}$$

The maximisation of R_{τ,τ_2} requires a detailed knowledge of all the spinflip and non-spinflip cross-sections and will vary from experiment to experiment. We may conclude that this method for optimising the polarising filter parameters P and T is impractical.

The need to maximise the difference count is such a dominating feature of crosscorrelation spectroscopy that in the polarisation modulated version using filter polarisers we propose that the appropriate filter parameter to be maximised should be the product of the polarising efficiency and transmittance P(E)T(E); see section 3.2. We shall see however that, since the neutron energy dependent filter transmittances are important in determining the background of ignorance, variations in the transmission products $T_1(\tau_1)\,T_2(\tau_2)$ due to ^{3}He nuclear spin relaxation need to be considered carefully in the proposed spectrometer.

The SF and NSF cross-sections are related to the nuclear spin incoherent cross-section, the nuclear coherent and isotopic incoherent cross-section, and the geometry-dependent magnetic cross-section. A unique feature of neutron polarisation analysis is that an unambiguous separation of magnetic and nuclear cross-sections can be effected by manipulation of the polarisation direction with respect to the scattering vector [8].

3. ^{3}He Neutron Polarising Filters

3.1 Practical Aspects

Following a major development programme over the past 5 years, the currently favoured method for polarising gaseous ^{3}He is by direct optically pumping of metastable ^{3}He atoms in a ^{3}He plasma at a pressure ~ 1mbar [4]. Recent advances have followed the introduction of powerful pumping lasers at wavelengths ~ 1083 nm. The polarisation build-up time is fast (1μs) and the main drawback of the technique is that it only functions a low pressures. Fortunately, compression of the gas by a factor ~ 10,000 (which produces atomic densities suitable for the polarising filter application) results in only a modest loss in ^{3}He nuclear polarisation, and polarising filter cells with gas pressures ~ 10 bar can be produced. The cells are readily emptied and refilled with polarised gas after the ^{3}He nuclear polarisation has relaxed to an unacceptably low level.

The current status of the development of ^{3}He polarising filters may be summarised as follows [5]: i) initial ^{3}He polarisation values ~ 60-65% are achievable, ii) compression enables neutron spin filter cells of ~ 300 cm^2 volume with 4 bar pressure and 50 - 55% ^{3}He polarisation to be prepared in about 4 hours, and iii) ^{3}He spin relaxation times in excess of 100 hours can be achieved in caesium-coated cells. In a typical operating cycle the filters are exchanged daily. Improvements in all these performance parameters can be expected over the next 5 years, and the goal of 70% ^{3}He polarisation in the spin filter cells remains a realistic perspective [9]. It is also likely that longer relaxation times will be achieved as further knowledge of the depolarisation mechanisms is gained [10].

3.2 ^{3}He Filter Performance Quality Factors and Optimisation

The principle underlying the operation of ^{3}He filters as neutron polarisation filters is the large spin dependence of capture into the broad (Γ = 270 keV) unbound resonance (J = I - ½=0) of the intermediate ^{4}He* state. The capture cross-section σ_c is large (5327 b atom^{-1} at a neutron energy E =25.3 meV, decreasing inversely with neutron velocity), and the ratio of the cross-section for antiparallel neutron spin capture to the total cross-section has been experimentally determined to be 1-010 ± 0.032 [11]. The neutron spin dependent cross-sections are related to the ^{3}He nuclear polarisation $P_{^3He}$ by

$$\sigma_{\pm} = \sigma_c \,(1 \mp P_{^3He}) \tag{3.1}$$

and the transmittances $T_{\pm}$ of (±) spin neutrons through the filter are given by exp-($\sigma_{\pm}$Nt), where Nt is the ^{3}He atomic thickness (at cm^{-2}). These transmittances have a substantial neutron energy dependence through the energy dependence ofσ_c ($\propto E^{-1/2}$).

The performance of a ^{3}He polarising filter may be defined by two parameters: its polarising efficiency P, which is the transmitted beam polarisation for an unpolarised incident beam,

$$P = (T_+ - T_-)\,/\,(T_+ + T_-) = \tanh\,(\sigma_c P_{^3He} Nt) = \tanh\,\alpha P_{^3He} \tag{3.2}$$

where $\alpha = \sigma_c Nt$ is the opacity of the filter, and its unpolarised beam transmittance,

$$T = (T_+ + T_-)\,/\,2 = \exp(-\alpha)\,\cosh\,(\alpha P_{^3He}). \tag{3.3}$$

For given values of $P_{^3He}$ and σ_c (fixed neutron energy) the filter designer is confronted with optimising the product of the gas pressure and filter thickness Nt. Nt can be increased to produce a higher neutron polarisation P, but only at the expense of lowering the transmittance T.

Tasset and Ressouche [12] have published a detailed analysis of the optimisation of α for the classical polarised neutron experiment which measures the ratio of scattered intensities for ‘up’ and ‘down’ spin neutrons - this ratio relates straightforwardly to the asymmetry.

These authors deduced that the optimum opacity α_{OPT} depends on the scattering experiment, and needs to be increased where reflections have a greater magnetic component. However, in the regime of low magnetic amplitudes where the classical diffraction experiments are most powerful it was concluded that α_{OPT} corresponds to a maximisation of $P\sqrt{T}$, as proposed earlier by Williams [13]. For asymmetry (or ratio) measurements we therefore define a filter quality factor

$$Q_1 = P\sqrt{T}. \tag{3.4}$$

However in many applications of polarised neutron beams, for example in inelastic scattering experiments, it is sufficient to measure the countrate difference for the two neutron spin states; see Williams [14]. For these the statistical accuracy of the measurements is proportional to product of the polarisation and beam intensity [14]. This also pertains in spin crosscorrelation spectroscopy, see eqn (2.3), where the difference cross-section depends on the PT products for the polariser and analyser individually. We have therefore used the filter quality factor

$$Q_2 = PT \tag{3.5}$$

to optimise the filter opacities α in the spin crosscorrelation spectrometer. Qualitatively, difference experiments place more emphasis on the filter transmittance, hence require lower opacities than experiments which measure asymmetries.

The optimisation of Q_1 has been derived by Surkau et al. [4] who give the optimum opacity $\alpha_{1(OPT)}$ for asymmetry experiments to be:

$$\alpha_{1(OPT)} = P_{^3He}^{-1} \ \mathrm{arctanh}\left\{\left[2 + (2P_{^3He})^{-2}\right]^{1/2} - 0.5\, P_{^3He}^{-1}\right\} \tag{3.6}$$

The equivalent optimisation of Q_2 gives the particularly simple expressions:

$$\alpha_{2(OPT)} = P_{^3He}^{-1} \ \mathrm{arctanh}\ \left\{P_{^3He}\right\} \tag{3.7}$$

$$= P^{-1} \ \mathrm{arctanh}\ \{P\} \tag{3.8}$$

where the nuclear polarisation $P_{^3\mathrm{He}}$ and transmitted neutron polarisation P are equal.

The calculated values of $\alpha_{1(\mathrm{OPT})}$ and $\alpha_{2(\mathrm{OPT})}$ as a function of $P_{^3\mathrm{He}}$ are shown in Figure 3. The variations of both these quantities in the ^{3}He polarisation range of practical importance ($P_{^3\mathrm{He}} < 0.8$) are small; opacity changes have much less influence on the filter parameters than changes in the neutron energy dependent capture cross-section. As expected from our qualitative argument $\alpha_{1(\mathrm{OPT})} > \alpha_{2(\mathrm{OPT})}$ for all $P_{^3\mathrm{He}}$ values, though the importance of the present results is that they show quantitatively that the optimum opacities of the ^{3}He filter can be reduced by a factor ~ x 0.65 in applications such as spin crosscorrelation spectroscopy which measure a difference signal.

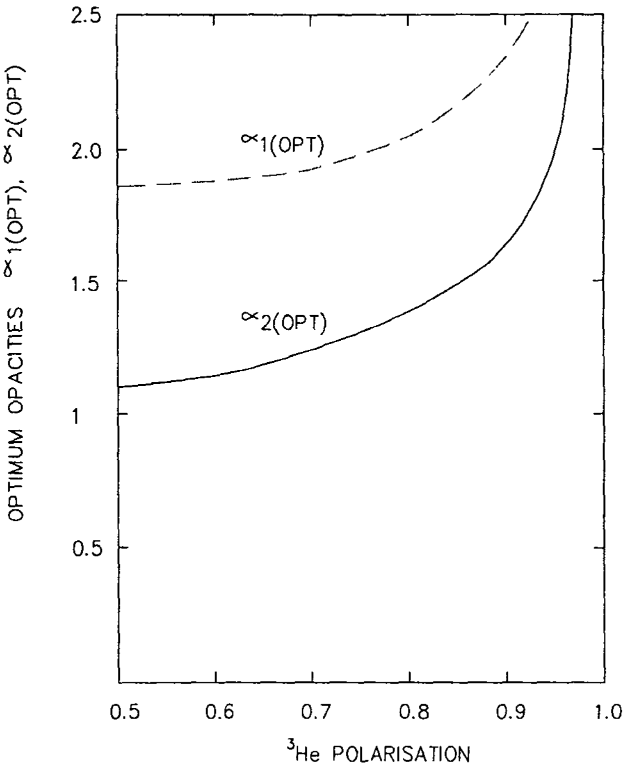
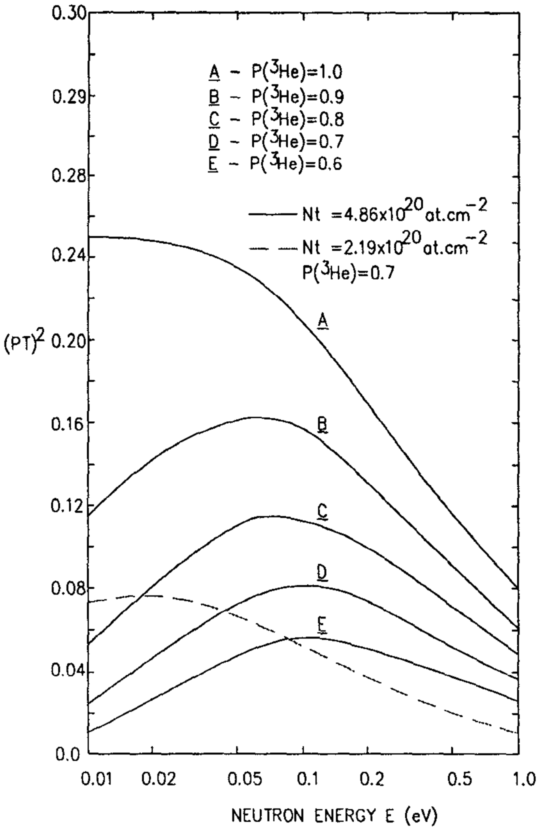


Figure 3: Optimum opacities $\alpha_{1(\mathrm{OPT})}$ and $\alpha_{2(\mathrm{OPT})}$ for ^{3}He polarising filters obtained by maximising Q_1 and Q_2 for ^{3}He polarisation values > 0.5.

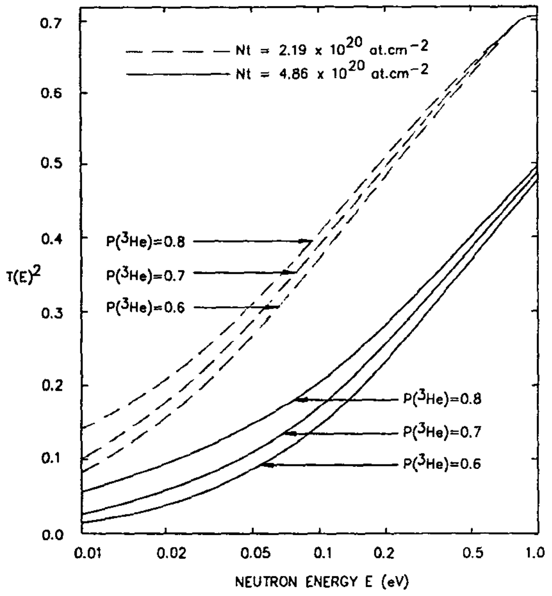
We now examine the neutron energy dependence of Q_2^2 (or P^2T^2) as a measure of the quality of performance of a two-filter spin crosscorrelation spectrometer when measuring elastic scattering - this is the filter-dependent term in the G term of eqn. (2.4). P^2T^2 is plotted as a function of neutron energy first for a filter with atomic thickness Nt = 4.86 x 10^{20} at cm^{-2} and a range of $P_{^3\mathrm{He}}$ values in Figure 4. This atomic thickness corresponds to $\alpha_{2\,(\mathrm{OPT})} = 1.23$ when $P_{^3\mathrm{He}} = 0.70$ for neutrons with energy E = 0.1 eV, and, in practical terms, to a 5 cm long filter cell with 4 bar ^{3}He pressure at 300K. The large $(PT)^2$ variation with ^{3}He polarisation is evident and illustrates i) the significant gains which result from increasing $P_{^3He}$, and ii) the potentially problematic effect of ^{3}He spin relaxation to the instrument performance - this point will be addressed later. The effect of changing the atomic thickness Nt on all polarising filter parameters (PT, P^2T^2, T^2 etc) at a fixed ^{3}He polarisation value is simply to linearly translate these curves in the direction of the E axis. This is a consequence of the neutron energy dependence of σ_c ($\alpha E^{-1/2}$), and is illustrated for a filter with Nt = 2.19 x 10^{20} at cm^{-2} with $P_{^3He}$ = 0.70 by the dashed curve of Figure 4. The selection of the appropriate atomic thicknesses of the filters (once the limiting ^{3}He polarisations are known) will depend on the neutron energy requirements of the experiment.



NEUTRON ENERGY E (eV)

Figure 4: Quality factors in elastic spin cross-correlation measurements with He-3 polarising filters of optimum opacity $\alpha = 1.24$ (He-3 polarisation = 0.70) as a function of neutron energy.

The neutron energy dependences of $T(E)^2$ for filters with atomic thicknesses 2.19 x 10^{20} at cm^{-2} and 4.86 x 10^{20} at cm^{-2} and ^{3}He nuclear polarisation values 0.6, 0.7 and 0.8 are shown in Figure 5. We recall that the lower atomic thickness filter was optimised for operation in the thermal neutron energy range with $(PT)^2$ maximum at 20 meV (see Fig. 4). Although this filter gives higher $T(E)^2$ values (and consequently a larger background of ignorance in the spin crosscorrelation experiment), we note that the fractional changes in this parameter due to changes in $P_{^3He}$ are much smaller than for the denser filter. For example, the fractional reduction $\Delta(T(E))^2/(T(E))^2$ for the Nt = 2.19 x 10^{20} at cm^{-2} filter is ~ 6% over the neutron energy interval 20 meV - 100 meV for a $P_{^3He}$ reduction of 10%. The corresponding reduction is $\Delta(T(E))^2/(T(E))^2$ for the Nt = 4.86 x 10^{20} at cm^{-2} filter is ~ 40%, although this filter produces a background of ignorance of approximately one-third that for the less dense filter.



NEUTRON ENERGY E (eV)

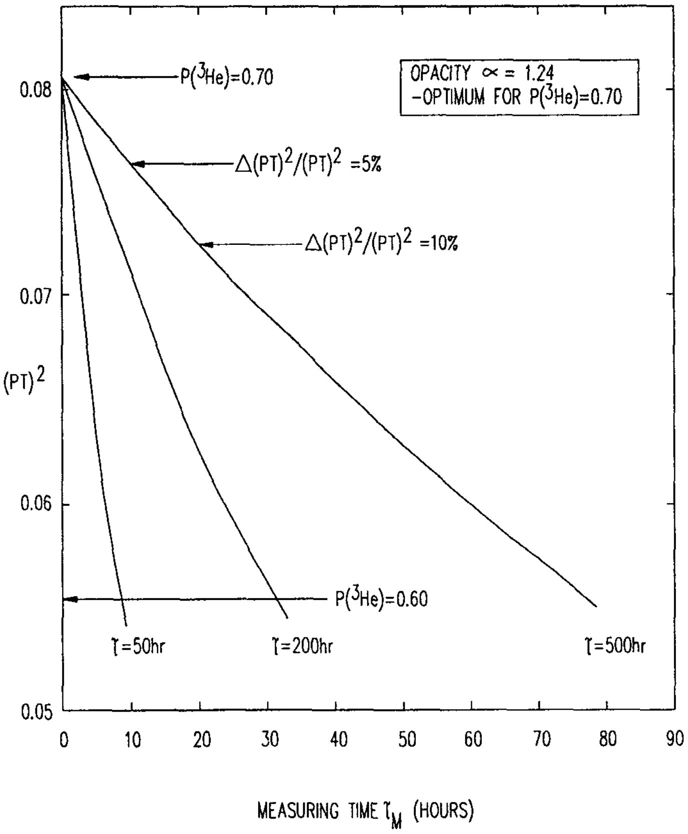
Figure 5: Neutron energy dependences of $T(E)^2$ for two ^{3}He polarising filters with nuclear polarisation values 0.6, 0.7 and 0.8.

Correlation techniques demand that the background of ignorance (which is often substantial) be as constant as possible and, in view of this requirement, we recommend that the lower atomic thickness filter would be preferable for measurements in the thermal neutron energy range. It is worth noting that $P_{^3He}$ reductions of 10% due to spin lattice relaxation would take place in approx. 12 hours in the best "long-life" filters built so far. This loss in ^{3}He polarisation results in a **reduced** background of ignorance in the spin crosscorrelation method, and the data must be corrected to account for this.

3.3 Effect of ^{3}He nuclear spin relaxation.

One disadvantage of the currently favoured method for polarising ^{3}He is that it cannot provide a stable filter performance due to ^{3}He spin relaxation. Currently, spin relaxation times in excess of 100 hours have been achieved, but even this is potentially problematical in long experiments with filter polarisers and analysers; we anticipate that even on third generation pulsed sources measuring times up to several tens of hours will be required periodically.

Fig 6 shows the reduction in the quality parameter $(PT)^2$ for a ^3He polarising filter with initial polarisation $P_{^3He}$ = 70% and optimum opacity $\alpha_{2(OPT)} = 1.24$, where the spin lattice relaxation times τ are 50 hr, 200 hr, and 500 hr. Again we restrict our calculations to the case relevant to elastic scattering processes. It is difficult to prejudge what fractional change in $(PT)^2$ can be tolerated during a single measurement since, in principle, this variation can be allowed for (with some uncertainty) at the data analysis stage. In the optimistic case that $\tau \sim 500$ hr will be achieved over the next 10 years (this will require more R & D effort on wall depolarisation) and that $\Delta(PT)^2/(PT)^2 = 10\%$ is acceptable, then this would allow measuring times up to ~ 20 hrs in the spin crosscorrelation experiment. Clearly any progress in lengthening the ^3He spin relaxation time will impact strongly on the feasibility of this experimental technique.



MEASURING TIME t_M (Hours)

Figure 6: Effect of He-3 nuclear spin relaxation on the spin cross-correlation signal for elastic scattering with optimum filter opacity for 70% ^3He polarisation.

The effect of ^3He spin relaxation in the polariser and analyser is not expected to have a significant effect on the signal to background ratio $R_{\tau_1\tau_2}$ in the crosscorrelation experiment. Note that a reduction in $P_{^3He}$ causes a **reduction** in the filter transmittance T, hence reducing the denominator in eqn (2.6). Since similar transmittance products appear in the numerator, the principal cause of the reduction in $R_{\tau_1\tau_2}$ due to ^{3}He spin relaxation can be attributed to smaller polarisation products P_1P_2 rather than to lower filter transmittances.

4. Summary

A preliminary evaluation of the feasibility of a polarisation modulated crosscorrelation neutron spectrometer, suitable for the third generation pulsed sources, is described. The spectrometer uses the recently developed optically pumped ^{3}He filters as the polarising elements, and the study has focussed on optimising the performance parameters of these filters. Detailed calculations on filters with two atomic thicknesses, Nt = 2.19 x 10^{20} at cm^{-2} and Nt = 4.86 x 10^{20} at cm^{-2}, showed that the former was more suitable for spin crosscorrelation experiments in the thermal neutron energy range. The effect of ^{3}He nuclear relaxation has been examined with the conclusion that R & D effort towards producing longer ^{3}He spin relaxation times would be beneficial to this technique. The method is only marginably viable given the current status of the development. ^{3}He polarisations in excess of 80% are needed compared with $P_{^3He}$ ~ 60% now achievable, but there is every prospect that such improvements will take place over the next 10 years.

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