

## Polarised Neutrons

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### 1. Introduction

The large majority of polarised beam neutron diffraction work on steady state sources has been carried out using the magnetised ferromagnetic single crystals  $\text{Co}_{0.92}\text{Fe}_{0.08}$  and the Heusler Alloy  $\text{Cu}_2\text{MnAl}$  as polarising monochromators. These polarisers have only a very limited applicability on pulsed sources, where we are concerned with white beams, sometimes up to eV energies. This paper is chiefly devoted to describing polarising filter methods which can be used either for white or epithermal beams. Efficient optical polarisers are readily available for cold neutrons<sup>(1,2)</sup>; their properties are well-understood and will not be discussed in this paper.

Polarised neutron measurements on pulsed sources will ultimately require full polarisation analysis<sup>(3)</sup>, ie spin selection in both the incident and scattered beams. In the case of elastic measurements the best instrumental arrangement is to use two wide-band polarising filters (Figure 1a). Inelastic experiments will probably best be performed with an inverse geometry instrument, since this only requires changing the analyser arrangement in Figure 1a. Possible designs include using polarising crystals<sup>(4)</sup> to combine spin/energy analysis (Figure 1b), or a focussing analyser system where a conventional 'curved' crystal analyser bank is used with a polarising filter (Figure 1c), as suggested by Scherm<sup>(5)</sup>. A detailed investigation of the optimum method to carry out inelastic polarisation analysis experiments will commence shortly at the Rutherford Laboratory.

### 2. Polarising Filters

Polarising filters operate by having substantially different linear attenuation coefficients for the two neutron spin states. The spin selection process can be either by preferential scattering or by preferential absorption. The main advantages of polarising filters are

that they can accept wide beam divergences and that they are often efficient over a very broad energy range.

The important parameters which determine the effectiveness of a neutron polarising filter are:

- (i) the polarising efficiency,  $p$ , ie the polarisation attained by an unpolarised neutron beam on being transmitted through the filter,

$$p = (T_+ - T_-) / (T_+ + T_-), \quad (1)$$

- (ii) the total transmittance,  $T$ , ie the (Total number of transmitted neutrons)/(Number of neutrons incident on filter) ratio,

$$T = (T_+ + T_-) / 2 \quad (2)$$

where  $T_+$  and  $T_-$  are the transmittances for the "up" and "down" neutron spins. The maximisation of a quality factor  $p\sqrt{T}$  is often used to optimise the filter thickness, though in general the best thickness should be evaluated by taking into account several parameters. In polarisation analysis experiments where there is a small amount of spin-flip scattering, it is more beneficial to increase  $p$  at the expense of  $T$ , ie to use a thicker filter than that given by maximising  $p\sqrt{T}$ <sup>(6)</sup>. The different types of polarising filters that may be used on pulsed neutron sources will now be described.

### 3. Polarised Proton Filters

Protons have a sufficiently large neutron spin dependent scattering cross-section that a target of polarised protons can act as a useful polarising filter. High proton polarisations ( $P_p \gtrsim 0.7$ ) are required, and there now seems to be two technically feasible methods of achieving this. Both use methods for transferring the easily-attainable electron polarisation in a paramagnetic dope into the proton spin system.

The first, and historically the most-tried technique, is to use dynamic polarisation (see Figure 2). In this method some of the protons are in dipolar coupling with the saturated electron spins of the dope and by irradiating at well-defined frequencies above and below that of the main ESR transition it is possible to create both positive and negative proton polarisations. The method is technically rather difficult, though it is

now being applied to polarise neutrons by Japanese workers<sup>(7)</sup>. There is a substantial amount of data available on polarising both resonance and thermal energy neutrons by transmission through a  $\text{La}_2\text{Mg}_3(\text{NO}_3)_{12}\cdot 24\text{H}_2\text{O}$  (LMN) polarised proton target<sup>(8)</sup>. Under typical experimental conditions the target is saturated with 4 mm microwaves at a temperature  $\sim 0.8\text{K}$  with a 1.87T magnetic field applied along the c-axis<sup>(9)</sup>. The possibility now exists of also using hydrogen-rich organic compounds doped with  $\text{Cr}^{5+}$  as filter materials; their main advantage over LMN for the neutron polariser application is that they are not required in single crystal form. There is however one difficulty - that of restricting the quantity of  $^3\text{He}$  (used as refrigerant) in the path of the neutron beam.

The proton polarisation method being developed at the Argonne Laboratory uses the spin refrigerator principle first introduced by Langley and Jeffries<sup>(10)</sup>. The best results have been obtained with single crystals of yttrium ethyl sulphate (YES) in which 0.01% of the  $\text{Y}^{3+}$  atoms are replaced by the paramagnetic dope  $\text{Yb}^{3+}$ <sup>(11)</sup>.  $\text{Yb}^{3+}$  acts as an effective  $s = \frac{1}{2}$  paramagnetic ion with a highly anisotropic g-factor ( $g_c \sim 3.3$ ,  $g_a \sim 0$ ) and when a single crystal is rotated about the b axis the spin dynamics is such that the high  $\text{Yb}^{3+}$  polarisation can be transferred to the proton spin system (see Figure 3). Typical experimental conditions are: magnetic field (poor homogeneity)  $\sim 1.5\text{T}$ , temperature  $T \sim 1\text{K}$ , spinning frequency  $\omega \sim 100\text{ Hz}$ . The main attraction of the technique is its simplicity compared with dynamic polarisation. Neutron tests are scheduled for Los Alamos for early 1981<sup>(11)</sup>.

The principal advantage of polarised proton filters is that it is the only method available for polarising true white beams even up to keV energies. The expected performances of an LMN polarising filter over a range of neutron energies, a range of filter thicknesses  $t$ , and for various values of the proton polarisation  $p_p$  have been calculated using the experimentally determined cross-section data of Lushchikov et al<sup>(8)</sup>, and an optimum filter thickness  $t_{\text{opt}} = 1.5\text{ cm}$  was selected after maximising for  $p\sqrt{T}$  over the thermal and epithermal neutron range. The calculated polarising efficiencies and transmittances of this filter for several values of the proton polarisation, and over the neutron energy range 0-0.6 eV are shown in Figures 4 and 5 respectively. The total and polarisation dependent cross-sections approach constant values at a neutron energy  $\sim 0.8\text{ eV}$ , thus the  $p$  and  $T$  values at energies  $> 0.8\text{ eV}$  are essentially the same as those shown for the higher energy limits of the curves. Polarised proton targets can now be built to give proton polarisations  $p_p > 0.80$ , and the curves show that this gives an efficient performance at all thermal and epithermal energies.

#### 4 Resonance Absorption Filters

If an unpolarised neutron beam whose energy corresponds to a resonance is transmitted through a target in which the nuclear spins are polarised, then one of the two neutron spin states is preferentially absorbed, and the transmitted beam is polarised. The absorption cross-sections ( $\sigma_{A\pm}$ ) for the two neutron spins is given by:

$$\sigma_{A\pm} = \sigma_A (1 \pm p_N \rho) \quad (3)$$

where  $\sigma_A$  is the absorption cross-section for zero nuclear polarisation  $p_N$ ,  $\rho = I/(I+1)$  if the compound nucleus formed has spin  $(I+\frac{1}{2})$ , and  $\rho = -1$  if it has spin  $(I-\frac{1}{2})$ . This spin-dependence of the capture has been used extensively in nuclear physics to measure the spins of resonant states. A polarising filter operating by this principle has a total transmittance

$$T = A \exp(-\sigma_A Nt) \cosh(\rho p_N \sigma_A Nt) \quad (4)$$

and polarising efficiency

$$p = -\tanh(\rho p_N \sigma_A Nt) \quad (5)$$

where the direction of  $p_N$  is defined as for the neutron polarisation,  $N$  and  $t$  are the nuclear density of the absorbing nucleus and filter thickness respectively, and  $A$  is an attenuation factor due to the non-absorbing nuclei in the filter. Equations (4) and (5) give the requirements for a good polarising filter functioning by selective capture, ie (1) it must be possible to obtain a high nuclear polarisation ( $p_N > 0.8$ ); this can often be obtained statically by cooling materials which have a large hyperfine field at the nucleus to dilution refrigerator temperatures, (2)  $|\rho|$  should approach unity, which means that the resonance should be of the  $(I-\frac{1}{2})$  type, or if of the  $(I+\frac{1}{2})$  type  $I$  should be large so that  $I/(I+1) \rightarrow 1$ , and (3)  $\sigma_A$  should be much greater than the total cross-sections for the non-absorbing nuclei in the filter.

A filter containing polarised  $^{149}\text{Sm}$  nuclei has been successfully developed at the Rutherford Laboratory<sup>(12)</sup> and new absorption filters are being investigated in order to extend the applicability of this method into the epithermal neutron region. Absorption filters offer particular advantages over scattering filters as spin analysers since the latter will almost certainly give higher detector backgrounds.

5. Polarised samarium filter

A material containing polarised  $^{149}\text{Sm}$  ( $I = 7/2$ ) nuclei acts as a good polarising filter for thermal neutrons since  $^{149}\text{Sm}$  has a strong resonance capture peak at  $E_0 = 0.97$  eV ( $\lambda \sim 0.92$  Å) which has been well-characterised by neutron measurements to be predominantly of the  $(I+\frac{1}{2})$  type<sup>(13)</sup>. The polarising filter material used at the Rutherford Laboratory was a deuterated cerous magnesium nitrate single crystal  $\text{Ce}_2\text{Mg}_3(\text{NO}_3)_{12}24\text{D}_2\text{O}$ , in which  $\sim 6\%$  of the Ce atoms were replaced by  $^{149}\text{Sm}$  atoms (CSMN). The deuterated salt was used rather than the hydrated one so as to reduce the neutron spin-independent attenuation factor A. The  $^{149}\text{Sm}$  nuclear polarisation was produced by applying a magnetic field  $H \sim 0.1$  Tesla along the crystal c-axis and by cooling the crystal to temperatures  $T < 0.02\text{K}$  in the mixing chamber of a  $^3\text{He}/^4\text{He}$  dilution refrigerator. The  $^{149}\text{Sm}$  nuclear polarisation was therefore set up by a static process, the Rose-Gorter method<sup>(14, 15)</sup>; the mechanism involves the electron-nuclear spin interaction, the electrons being essentially fully polarised at the temperatures and magnetic fields used. The hyperfine interaction along the c-axis of CSMN is  $\sim 0.041\text{K}$  in temperature units, and this corresponds to a hyperfine field at the nucleus  $\sim 300$  Tesla.

The polarising efficiency and transmittance values for the filter are strongly neutron energy dependent, due to the resonant nature of  $\sigma_A$ . This will not in general be disadvantageous in a polarised beam measurement of flipping ratios. Polarising efficiency and transmittance measurements at two wavelengths ( $\lambda = 0.96$  Å and  $\lambda = 1.18$  Å) gave values which were within a few per cent of the theoretically predicted ones. Figure 6 shows the calculated spin-dependent transmittances and polarising efficiencies of the filter at the refrigerator base temperature (16 mK) as a function of neutron energy, calculated on the basis of the measurements at the two wavelengths. The obvious disadvantage of the Sm filter for pulsed source applications is that it does not perform at neutron energies  $E \gtrsim 0.14$  eV. This is not a severe limitation in low Q elastic measurements since in order to operate without frame overlap (dictated by the machine p.r.f. and instrument length) it turns out that for our first experiments we are not able to operate at  $E \gtrsim 0.2$  eV. There are now plans in the UK for constructing a two samarium filter polarisation analysis spectrometer at the Harwell Linac pulsed neutron facility. A large Q-scan ( $0.3 < Q(\text{Å}^{-1}) < 1.7$ ) in total scattering measurements of magnetic diffuse scattering will be possible

at one scattering angle setting of the filter analyser by using a polychromatic beam ( $12 \text{ meV} < E < 130 \text{ meV}$ ). Metallic polarising filter materials containing  $^{149}\text{Sm}$ , eg  $\text{SmCo}_5$  with better thermal conductivities than CSMN are being developed for this instrument<sup>(16)</sup>.

6. Polarised europium filter

$^{151}\text{Eu}$  has two broad resonances which extend up to a neutron energy  $E \sim 0.6$  eV. Calculations show that it should be possible to polarise Eu-containing materials at temperatures  $\sim 10\text{--}15$  mK to produce a polarising filter with good transmittance and polarising efficiencies up to this energy<sup>(17)</sup>. The spin selection is not quite as efficient as that for  $^{149}\text{Sm}$  thus one would probably use a  $^{149}\text{Sm}/^{151}\text{Eu}$  two-foil combination to perform well over the entire range  $0 < E \text{ (eV)} < 0.6$ . The predicted performance of such a combination is shown in Figure 7; the calculations assumed that the filters are cooled to 15 mK, that the electron magnetisation is fully saturated, and that the hyperfine field at the  $^{151}\text{Eu}$  nucleus is the free ion value  $H_{\text{eff}} \sim -34$  Tesla.

This filter combination could be used to extend the Q range possible in elastic experiments but, more importantly, it could be used as the polariser in an inverse geometry inelastic instrument to study magnetic excitations up to  $\hbar\omega \sim 0.25$  eV.

7. Polarised erbium filter

$^{167}\text{Er}$  has two narrow resonances at energies  $E = 0.584$  eV ( $J = I - \frac{1}{2} = 3$ ) and  $E = 0.460$  eV ( $J = I + \frac{1}{2} = 4$ ). Further, ferromagnetic intermetallic materials such as  $\text{Fe}_2\text{Er}$  are known with a sufficiently high hyperfine field at Er ( $H_{\text{eff}} \sim 850$  Tesla) to enable high nuclear polarisations to be achieved at temperatures  $T \leq 30$  mK. Calculations of the performance of a polarised  $^{167}\text{Er}$  have been carried out<sup>(18)</sup> with typical results (for  $Nt \sim 1.37 \times 10^{21}$  at  $\text{cm}^{-2}$ ):

Neutron Energy eV	Polarising Efficiency p	(+) Spin Transmittance $T_+$	(-) Spin Transmittance $T_-$
0.460	0.99	0.454	0.0023
0.584	-0.985	0.0072	0.976

The performance at  $E = 0.584$  eV is close to the theoretical maximum for any polariser; this is a result of the  $I-\frac{1}{2}$  nature of the resonance (see equation (3)).

This filter could be used in the incident beam of a chopper spectrometer to provide 'monochromatic' highly polarised beams at the resonance energies. The main scientific applications would be in studying magnetic excitations in aligned samples where  $\hbar\omega \gtrsim 0.25$  eV by partial polarisation analysis.

#### 8. Polarised holmium filter

$^{165}\text{Ho}$  has a sharp (FWHM  $\sim 0.15$  eV) resonance peak at  $E = 3.92$  eV with a compound nucleus spin  $J = I + \frac{1}{2} = 4$ <sup>(19)</sup>. Ho is also known to be one of the easier nuclei to polarise<sup>(20)</sup>, for example a polarisation  $P_N \sim 0.95$  can be generated in a magnetically saturated sample at a temperature  $T \sim 0.1\text{K}$ . We here speculate that such a filter might be used in an inverse geometry instrument to combine the functions of spin analysis and detection by using the resonance detector principle. An extension of this idea to combine it with a spin precession coil to Fourier chop the scattered beam so as to improve the inherent resolution of the resonance detector spectrometer has been proposed recently at the Argonne Laboratory<sup>(21)</sup>. The 3.92 eV resonance could be used to study magnetic and electronic excitations at low  $Q$  up to  $\hbar\omega \sim 1.0$  eV.

#### 9. Conclusions

Polarised beam work on pulsed neutron sources will rely heavily on developing effective polarising filters. The Rutherford Laboratory programme will concentrate on extending the range of applicability of the resonance absorption method and some of the possibilities have been described.

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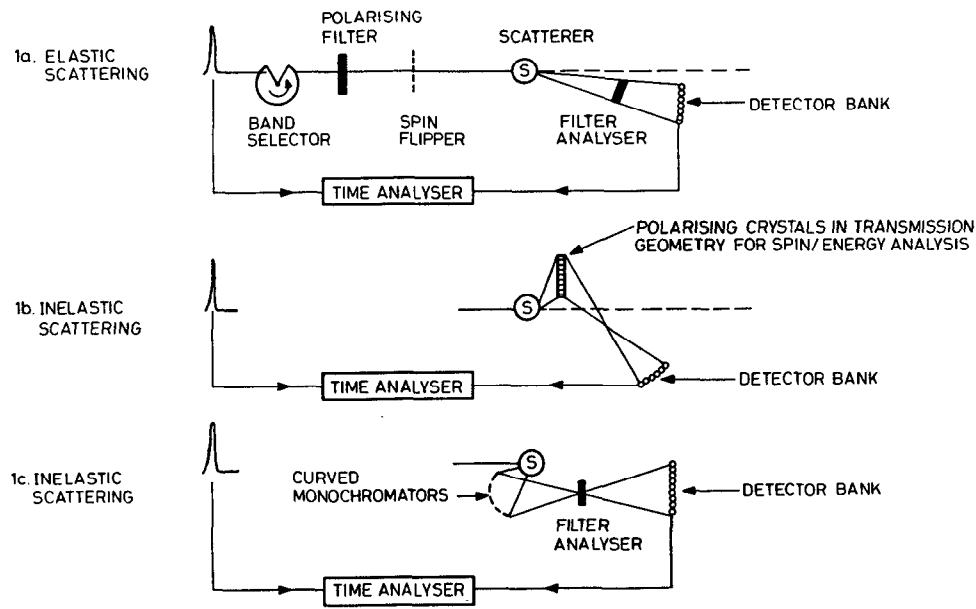
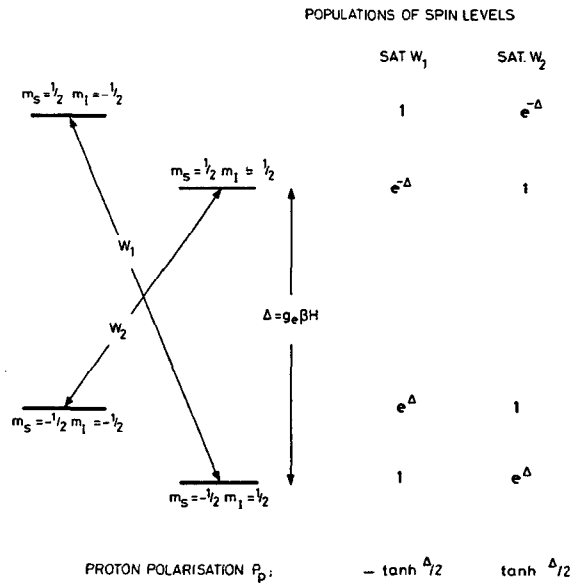
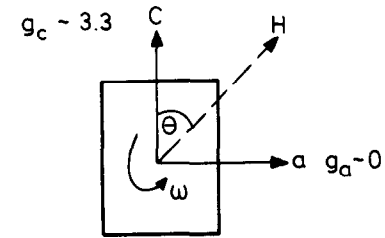


FIGURE 1. POLARISATION ANALYSIS ON PULSED SOURCES

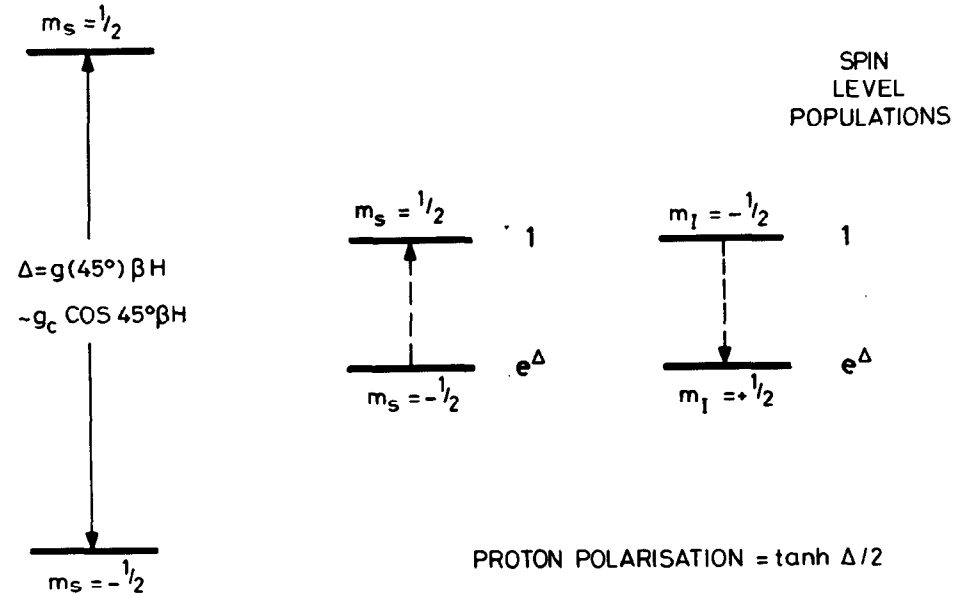


PROTON SPIN I AND ELECTRON SPINS IN DIPOLE COUPLING  
eg  $\text{Na}^{3+}$  IN LMN,  $\text{Cr}^{5+}$  IN ETHYLENE GLYCOL.

FIGURE 2. DYNAMIC PROTON POLARISATION



YES ( $Yb$ ) ROTATED ABOUT b AXIS



PROTON POLARISATION =  $\tanh \Delta/2$

$\theta = 45^\circ; T_{ie}^{-1}$  - FAST     $\theta = 90^\circ; T_{ie}^{-1}, T_{ip}^{-1}$  - SLOW

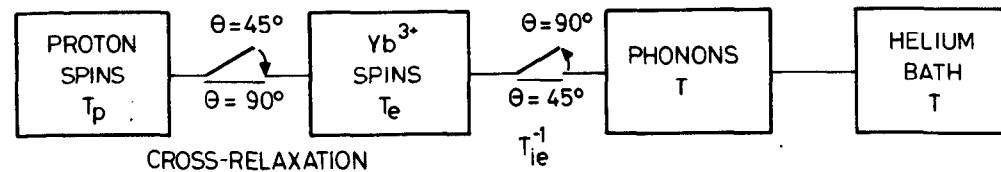


FIGURE 3. PROTON POLARISATION IN SPIN REFRIGERATOR

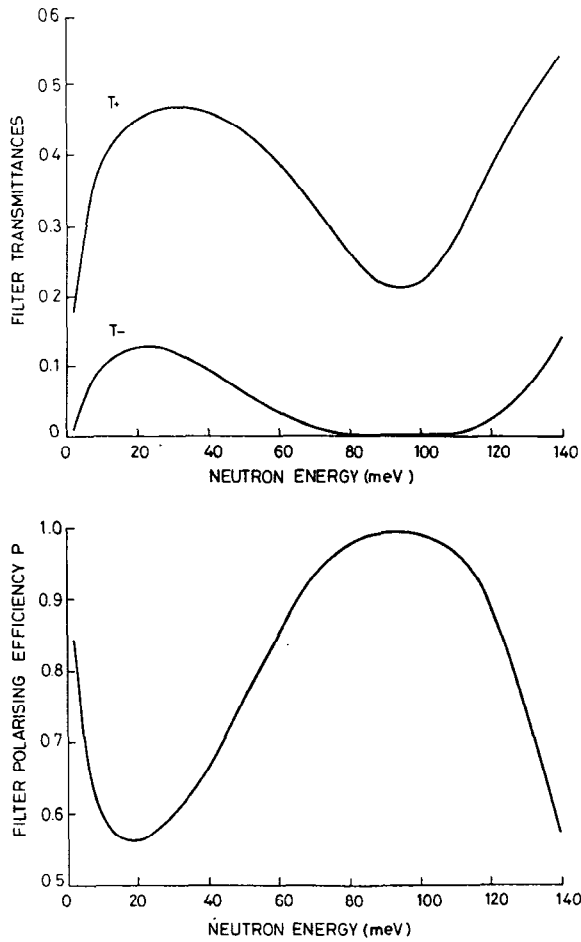


FIGURE 6. SPIN-DEPENDENT TRANSMITTANCES AND POLARISING EFFICIENCY OF Sm FILTER vs NEUTRON ENERGY

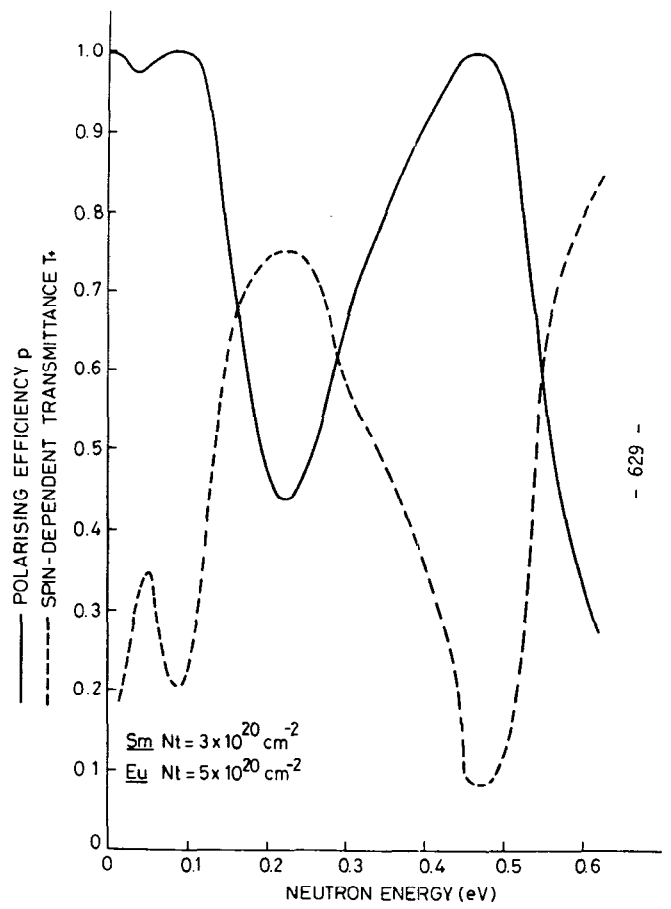


FIGURE 7. SPIN-DEPENDENT TRANSMITTANCE AND POLARISING EFFICIENCY OF Sm/Eu FILTERS IN SERIES vs NEUTRON ENERGY

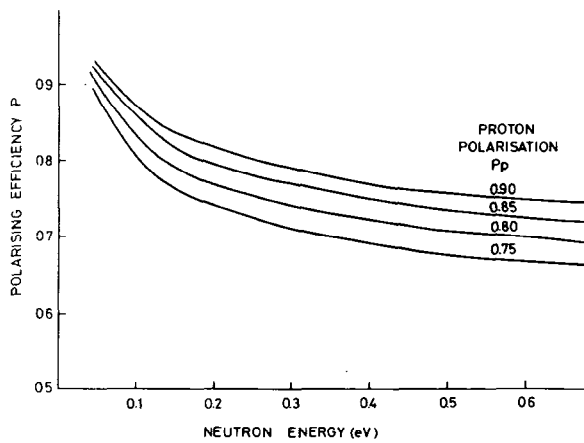


FIGURE 4. NEUTRON ENERGY DEPENDENCE OF THE POLARISING EFFICIENCY OF A 1.5cm THICK LMN POLARISING FILTER OVER THE PROTON POLARISATION RANGE  $0.75 < P_p < 0.90$

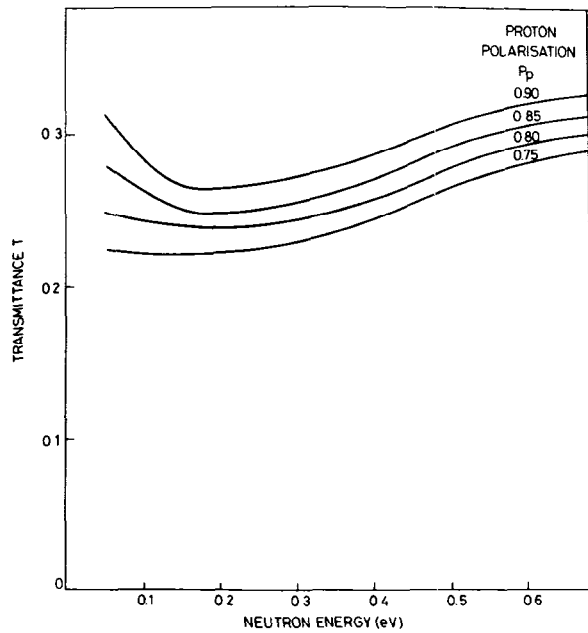


FIGURE 5. NEUTRON ENERGY DEPENDENCE OF THE TRANSMITTANCE OF A 1.5cm THICK LMN POLARISING FILTER OVER THE PROTON POLARISATION RANGE  $0.75 < P_p < 0.90$