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POLARISING FILTER DEVELOPMENTS AT ISIS

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As part of the development program for the polarised neutron instrument POLARIS on the ISIS spallation neutron source, tests have been performed on prototype neutron polarising filters for pulsed neutrons. The principle of operation is to align samarium nuclei at millikelvin temperatures. There is then preferential absorption of one of the two neutron spin states near the 96 meV nuclear resonance absorption in samarium. The best results obtained so far have been with a filter of sintered $SmCo_5$ cooled to 0.014K in a dilution refrigerator. The measured polarising efficiency of this filter is between 98% and 67% with corresponding total transmittances of 10% and 33% within the energy range 10 meV to 150 meV. The average value of P^2T over this range is greater than 70% of the theoretical optimum obtainable by this method.

1. INTRODUCTION

The POLARIS instrument on the ISIS spallation neutron source is at present devoted to developing polarised neutron techniques on pulsed neutron sources. The measurements described here, performed on the POLARIS beamline, are part of the program for developing efficient neutron polarisers for such sources. We wish to develop devices which will polarise both over a broad energy band in the cold and thermal neutron ranges and at epithermal neutron energies. The first requirement is necessary so that time of flight techniques can be used on a 'white' beam. The second because at present the only practical devices for polarising neutrons at high energies utilise Bragg reflections from magnetic single crystals such as the Heusler alloy Cu_2MnAl or iron-cobalt. The resulting monochromation of the neutron beam is very wasteful on a pulsed neutron source where time of flight techniques can be used to define the neutron energy. It is possible to produce low energy 'white' polarised neutron beams by using supermirrors. In principle, supermirror guides could also be constructed to operate at epithermal energies but so far their useful range of operation is limited to energies below 30 meV. Polarising filters have advantages over supermirror guides even if these could be made to operate at epithermal energies. Probably the most important advantage is that a filter will accept all beam divergences whereas the divergence of the beam transmitted by a guide is limited. However the polarising filters are more difficult to operate.

The theory of operation of resonance absorption neutron filters has been fully described previously (1,2,3), so we give only a brief description here. The filters operate by preferentially absorbing one of the two neutron spin states at energies close to neutron resonance absorption energies in aligned rare earth nuclei (4). The method depends upon producing a high atomic moment alignment in the filter either by using a ferromagnetic material or by applying a magnetic field (> 0.1 Tesla) to a paramagnetic material. At sufficiently low temperatures the nuclear moments will then align, parallel to the atomic moments, in the hyperfine field produced by the orbital motion of the 4f electrons - the Rose-Gorter effect (5).

The Sm^{149} isotope has a nuclear polarisation of ~ 90% at 15 mK and there is a Breit-Wigner resonance centred at ~ 96 meV and with a width of ~ 67 meV (6,7). The wings of the resonance extend sufficiently far to make this isotope a useful neutron polariser at all energies between 0 and 200 meV. There is also a higher energy resonance at ~ 870 meV, which could be used to polarise neutrons at energies between 750 and 950 meV. The capture cross-sections in Sm^{149} for the two neutron spin states are

$$\sigma_{\pm} = \sigma(1 \pm \rho P_N) \tag{1}$$

where $\rho = \frac{I}{I+1}$ is a nuclear spin weighting factor. Sm^{149} has a nuclear spin of 7/2 so that the two spin state capture cross-sections are in the ratio 8:1 at $P_N = 1$. In this paper we describe measurements on prototype filter materials containing polarised samarium nuclei.

We have concentrated on developing metallic filter materials as these have better thermal conductivity. At the high neutron intensities on ISIS it is anticipated that the extraction of the heat produced within the filter by the neutron capture process is likely to be problematical. A metallic material will allow heat to be easily conducted from within the filter to its surface. Probably more importantly a good metal to metal contact between the filter and the dilution refrigerator, used to attain the necessary millikelvin temperatures, will eliminate problems associated with the Kaptitza thermal resistance at the filter surface.

In principle a paramagnetic material will give a better performance than a ferromagnetic material unless a filter consisting of a single ferromagnetic domain can be produced. This is because of the presence of depolarisation effects in ferromagnets, which limit both the ultimate polarisation of the beam and reduce the transmitted intensity. However, so far we have been unable to find a metallic material containing samarium which behaves paramagnetically at ~ 15 mK, the temperature necessary to generate a high nuclear polarisation. At such low temperatures even small interactions will tend to cause magnetic ordering.

In the following section we describe measurements made on a 5% solid solution of samarium in palladium, which we hoped would behave paramagnetically, but which showed evidence of magnetic interactions between the samarium atoms. We also describe the results of measurements on a filter consisting of the ferromagnet $SmCo_5$. This material has produced by far the best polarising performance of any polarising filter so far developed.

2. MEASUREMENTS ON $Pd_{0.95}Sm_{0.05}$

The samarium and palladium used in the preparation of the filter were supplied by Rare Earth Products and Johnson Matthey Chemicals Ltd. The purity of the samarium was 99.99% and the palladium was 'specpure'. The alloy was prepared in the Department of Materials Science and Metallurgy at the University of Birmingham. Appropriate amounts of the two metals were argon arc-melted several times on a water cooled copper hearth and the resulting button was homogenised for two weeks at 1200°C in an argon atmosphere. The homogeneous alloy was cold rolled to a thickness of 0.84 mm and then annealed for a further six hours at 800°C to relieve internal strains. Energy dispersive X-ray analysis gave a composition of 5 at % Sm. The filter was soldered to a copper plate which was attached by screws to the bottom of the dilution unit of a dilution refrigerator supplied by Oxford Instruments Ltd.

In the first measurement, the transmittance of the filter was measured at a temperature of 1.2K. The transmittance of the filter is essentially

$$T_h = \exp(-\,Nt\,\sigma) \tag{2}$$

where Nt is the number of samarium atoms/cm^2 and σ is the absorption cross-section of samarium. All other absorption and scattering processes within the filter are negligible compared to the samarium absorption. The samarium absorption cross-section as a function of energy E can be written as

$$\sigma(E) = \left(\frac{A_1}{E}\right)^{\frac{1}{2}} \frac{A_3}{1 + \left[\frac{2(E-A_1)}{A_2}\right]^2} + \frac{A_4}{E^{\frac{1}{2}}} + \left(\frac{A_5}{E}\right)^{\frac{1}{2}} \frac{A_7}{1 + \left[\frac{2(E-A_5)}{A_6}\right]^2} \tag{3}$$

The first and third terms are Breit-Wigner forms for the nuclear resonances nominally at 96 meV and 872 meV (6,7). The second term is a negative energy resonance which becomes significant only at low energies and gives a capture cross-section which is inversely proportional to the neutron velocity. The neutron energy is related to its time of flight, t, by $E = 5.2276 \times 10^6\ L^2/t^2$ where L, the moderator to detector distance is in metres, t is in µsec and E in meV. [The moderator-detector distance, 10.55 m was calculated by fitting the transmitted time-of-flight spectra from uranium and iridium for which the resonance energies are known accurately.]

Figure 1 shows the measured PdSm transmittance and a fit to the data using the functional form given by equations 2 and 3.

We obtained the values

A_1 = 96.3 (± 0.1) meV
A_2 = 62.7 (± 0.3) meV
A_3 = 5.17 (± 0.03)
A_4 = 0.34 (± 0.01)
A_5 = 878.0 (± 1.0) meV
A_6 = 68.1 (± 0.1) meV
A_7 = 0.774 (± 0.001)

The quoted errors are estimates obtained by observing the variation in the final fitted parameters as the input values to the fitting routine were varied. The energy values are in good agreement with resonance parameters given by Marshak et al (6) and Asami (7). The number density Nt was calculated from A_3 and A_4 using a value for σ (98 meV) of 18200 barns and for σ (870) of 2760 barns; this gave the value Nt = 2.84×10^{20} Sm atoms/cm^2. This number density is slightly greater than the optimum value for a samarium filter (8).

In order to characterise the filter performance it is also necessary to find the ratio of the neutron transmittance at operating temperature (T_c) to that at 1.2K (T_H). At 1.2K the nuclear polarisation, P_N, is insignificant. It can easily be shown (1,2) by using equation (1) to calculate the transmittances of the two neutron spin states, that

$$\frac{T_c}{T_H} = \cosh\,(\rho P_N\, Nt\sigma) \tag{4}$$

The measured transmittance ratio for $Pd_{0.95}\,Sm_{0.05}$ is shown in Figure 2 together with the fit to equation (4). Using the value of $Nt\sigma$ calculated from the transmittance at 1.2K, the nuclear polarisation, P_N, was found to be 0.336. The measurements were performed at a temperature of 25 mK (measured by Co^{60} nuclear orientation thermometry) and in a field of 2.5 tesla. Under these conditions the expected nuclear polarisation for Sm is ~ 0.78.

The most probable explanation for the low nuclear polarisation is that there are magnetic interactions between the Sm atoms. This hypothesis is supported by the observation that when the magnetic field was reduced to zero, some nuclear polarisation was retained. Figure 3 shows the measured and fitted transmittance ratio at zero field. The fit gave a residual polarisation $P_N = 0.18$, confirming that the filter is not behaving as an ideal paramagnet.

The transmittance, T, and the polarising efficiency, P, of the filter are given by

$$T = \exp(-\,Nt\sigma)\,\cosh(\rho P_N\, Nt\sigma) \tag{5}$$

$$P = \tanh\,(\rho P_N\, Nt\sigma) \tag{6}$$

P and T, calculated using the fitted values of $Nt\sigma$ and P_N, are shown in figure 4 as a function of wavelength. Both P and T are significantly lower than the values achievable in a fully aligned Sm filter.

3. MEASUREMENTS ON SINTERED $SmCo_5$

$SmCo_5$ is widely used as a commercial ferromagnet with high coercivity and magnetic inductance. The filter used for the neutron measurements was also prepared and characterised at the Department of Physical Metallurgy, University of Birmingham. Slices of dimensions 4 cm x 1 cm x 0.1 cm were cut from a parent block of sintered $SmCo_5$ using a low speed 'isomet' diamond cutting wheel. These slices had their magnetic C-axis in the plane of the slice and along the 1 cm edge. Each slice was subsequently spark planed to a thickness of 0.024 cm to give the required Sm atomic thickness.

A small sample of the material was analysed on a Sucksmith type magnetic balance to find the degree of magnetic alignment. The remanence both parallel ($Br_{\parallel}$) and perpendicular ($Br_{\perp}$) to the alignment direction was measured. The mean value $\langle \cos\theta \rangle$ where θ is the angle between the crystallographic c axis of the individual particle and the alignment direction of the filter was determined using an empirical model and found to be 0.91.

A filter of dimensions 3 x 4 cm was constructed by laying the slices side by side. This was contained inside a hollow aluminium filter holder and the measurements were performed with the filter immersed in the He^3-He^4 mixture of the dilution refrigerator. Good thermal contact between the material and the mixture was expected because of the large surface area/volume ratio of the sintered material, where the measured particle diameter was ~ 1 micron. The procedure used to characterise the filter performance was identical to that described in section 2. The neutron beam was collimated to a 1 x 2 cm size to ensure that all detected neutrons had passed through the filter. The values of the ^{149}Sm nuclear resonance parameters obtained from the fit to the filter transmittance at 4.2K were

A_1 = 96.1 (± 0.1) meV
A_2 = 67.3 (± 0.1) meV
A_3 = 4.15 (± 0.04)
A_4 = 0.235 (± 0.01)
A_5 = 870.2 (± 0.1) meV
A_6 = 75.2 (± 0.1) meV
A_7 = 0.61 (± 0.01)

These are also in good agreement with previously published values of these resonance widths and positions. In figure 5 we show the measured transmittance ratio for $SmCo_5$ together with a fit to the data. The temperature of the dilution unit measured by Co^{60} nuclear orientation thermometry was 14 mK. The solid line is a fit to the function (1,2)

$$\frac{T_c}{T_H} = \exp(-Dt)\ [\cosh(Kt) + \nu\sinh(Kt)]\ \exp(C_4/\sqrt{E}) \qquad (5)$$

$$\text{where}\quad Kt = [(Nt\,\sigma\rho C_1)^2 + D^2]^{\frac{1}{2}} \qquad (5a)$$

$$Dt = C_2(1-C_3{}^2)/E \qquad (5b)$$

$$\nu = Dt/Kt$$

This expression is more complicated than that of equation 4 because of the inclusion of depolarisation effects within the ferromagnetic material. In the model used to calculate depolarisation (1,2) the fitting parameter C_2 is related to the mean particle size and C_3 is the particle alignment factor, $\langle\cos\theta\rangle$. The parameter C_4 must be included to account for the different concentration of He^3 in the mixture surrounding the filter at 1.2K and 14 mK (9). Finally, parameter C_1 is equal to the product of the particle alignment $\langle\cos\theta\rangle$ and the local nuclear polarisation with respect to the particle c axis. This product is the apparent nuclear polarisation experienced by neutrons passing through the filter. We obtain the values

C_1 = 0.81 (± 0.01)
C_2 = 5.0 (± 3.0) meV
C_3 = 0.9 (± 0.2)
C_4 = 1.05 (± 0.05) $(\text{meV})^{\frac{1}{2}}$

It can be seen from the quoted errors that the fit is very sensitive to the value of C_1 (= $P_N \langle\cos\theta\rangle$) but relatively insensitive to C_2 and C_3. This is not unexpected when there is good alignment. In the limit of perfect alignment, equations 5 reduce to equation 4 and there is no dependence on C_2 and C_3. A more accurate value for C_3 can be obtained by using the value P_N = 0.91 calculated for Sm at the measured temperature of 14 mK. The alignment factor is then

$$\langle\cos\theta\rangle = \frac{C_1}{P_N} = 0.89$$

This is in excellent agreement with the value determined from the measurements of remanent magnetisation. The value of the mean particle diameter derived from C_2 is again in excellent agreement with that found during the filter characterisation.

The parameter C_4 can also be checked independently. The attenuation of the He^3 in the mixture at 1.2K was measured by taking the ratio of the transmittance at 1.2K with the mixture condensed around the filter to that at 4.2K with the mixture extracted. This transmittance was fitted to the function

$$T_{He} = \exp(-C/E^{\frac{1}{2}}) \tag{6}$$

and gave C = 2.31 $meV^{\frac{1}{2}}$. At 14 mK the mixture contains 6% He^3 (10) so the proportion of He^3 in the mixture at 1.2K can be calculated from C and C_4.

Proportion of He^3 in mixture at 1.2K = $6\% \times \frac{2.31}{1.05} = 13.2\%$

This is in very satisfacatory agreement with the nominal value of 15%.

It can be seen that excellent fits to the data were obtained and that there is good agreement with other measurements where comparison is possible. The parameters can therefore be used with some confidence to determine the performance of the filter at different neutron energies. The polarising efficiency is (1,2).

$$P = \frac{\tau \tanh\ (Kt)}{1 + \nu \tanh\ (Kt)} \tag{7}$$

$$\text{where } \tau = \frac{Nt\,\sigma\rho C_1}{Kt} \tag{7a}$$

and the transmittance is

$$T = \exp(-Dt-Nt\,\sigma)\ [\cosh(Kt) + \nu\sinh(Kt] \tag{8}$$

The values of P and T obtained from the fitted parameters for the $SmCo_5$

filter are shown in Figure 6. In Figure 7 P and T for an 'ideal' filter with the same value of σNt but with $\langle\cos\theta\rangle = 1$ are shown for comparison. It can be seen that the performance of the filter is close to the best obtainable. It can be shown that the effective intensity of the polarised beam produced by a filter is proportional to the product P^2T for a wide range of sample properties (8). Figure 8 shows P^2T as a function of neutron wavelength for the measured and ideal filters. The prototype filter gives between 70 and 80% of the effective neutron intensity of the ideal filter.

DISCUSSION

The performance of the $SmCo_5$ filter described in this paper is as to our knowledge the best measured so far of any neutron polarising filter. It could be used as a component of a polarised neutron instrument on a pulsed source. We now plan to measure the filter performance at higher neutron intensities so as to examine whether there is any onset of beam heating effects. The measurements described here were performed at 2% of full ISIS intensity on a 1 x 2 cm filter area. This corresponds to $\sim 10^6$ neutron/sec absorbed by the filter. At full intensity and with an incident beam size of 3 x 4 cm the heat load will be increased by a factor of 300. We also intend to measure the filter polarising efficiency directly by scattering the transmitted beam from a Heusler alloy polarising crystal (3).

Although the polarising efficiency of the $SmCo_5$ filter is worse than that expected from a perfect paramagnet (3), the material has a compensating advantage. This is that there is no need to apply a magnetic field to align the atomic moments. This makes the technique for obtaining a polarised beam simpler. More importantly it provides the possibility of constructing a large area filter for analysing scattered neutrons. The feasibility of building a large area $SmCo_5$ filter will be explored in the future.

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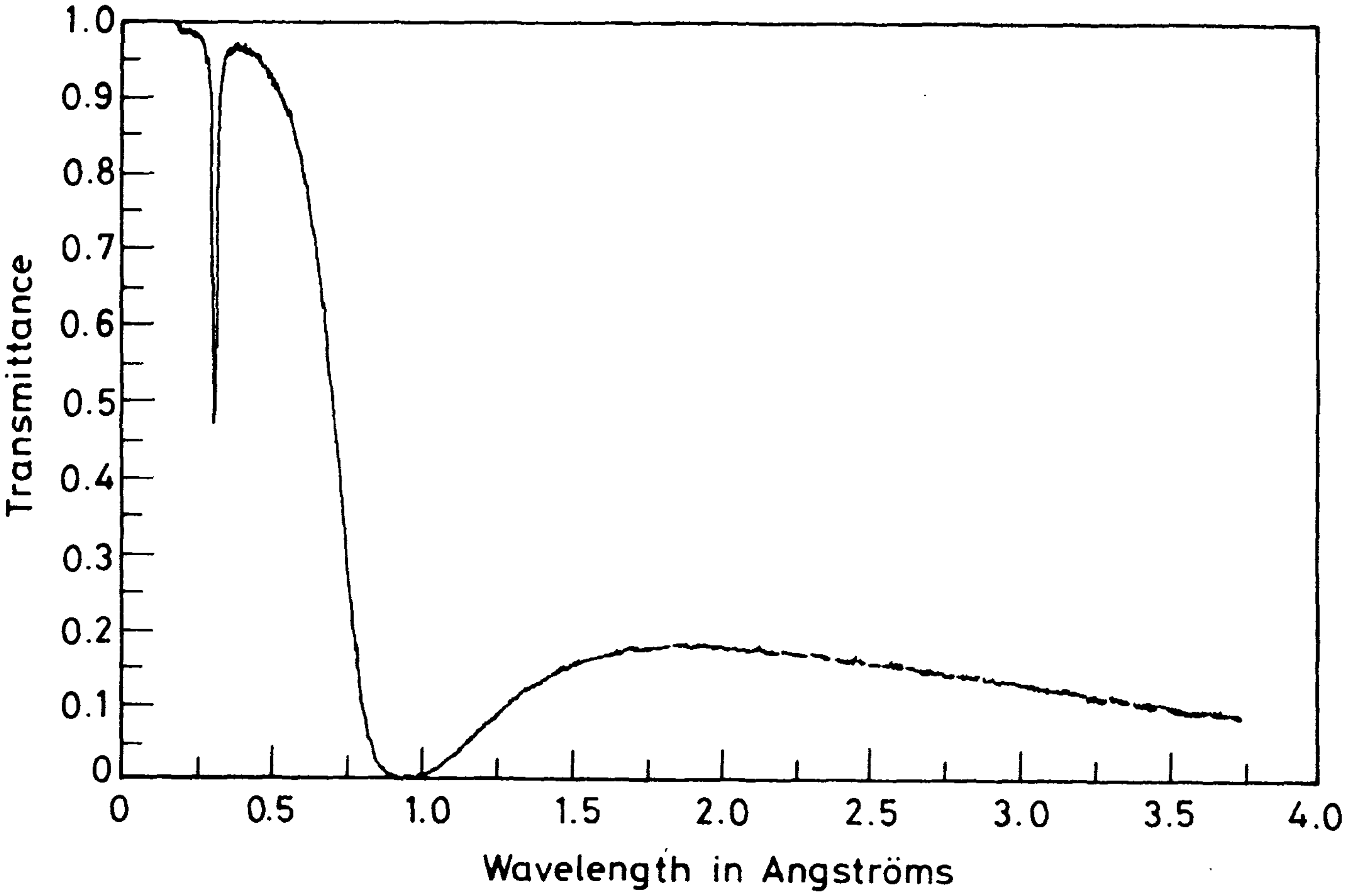

Figure 1. Transmittance of $Pd_{0.95}Sm_{0.05}$ at 1.2K

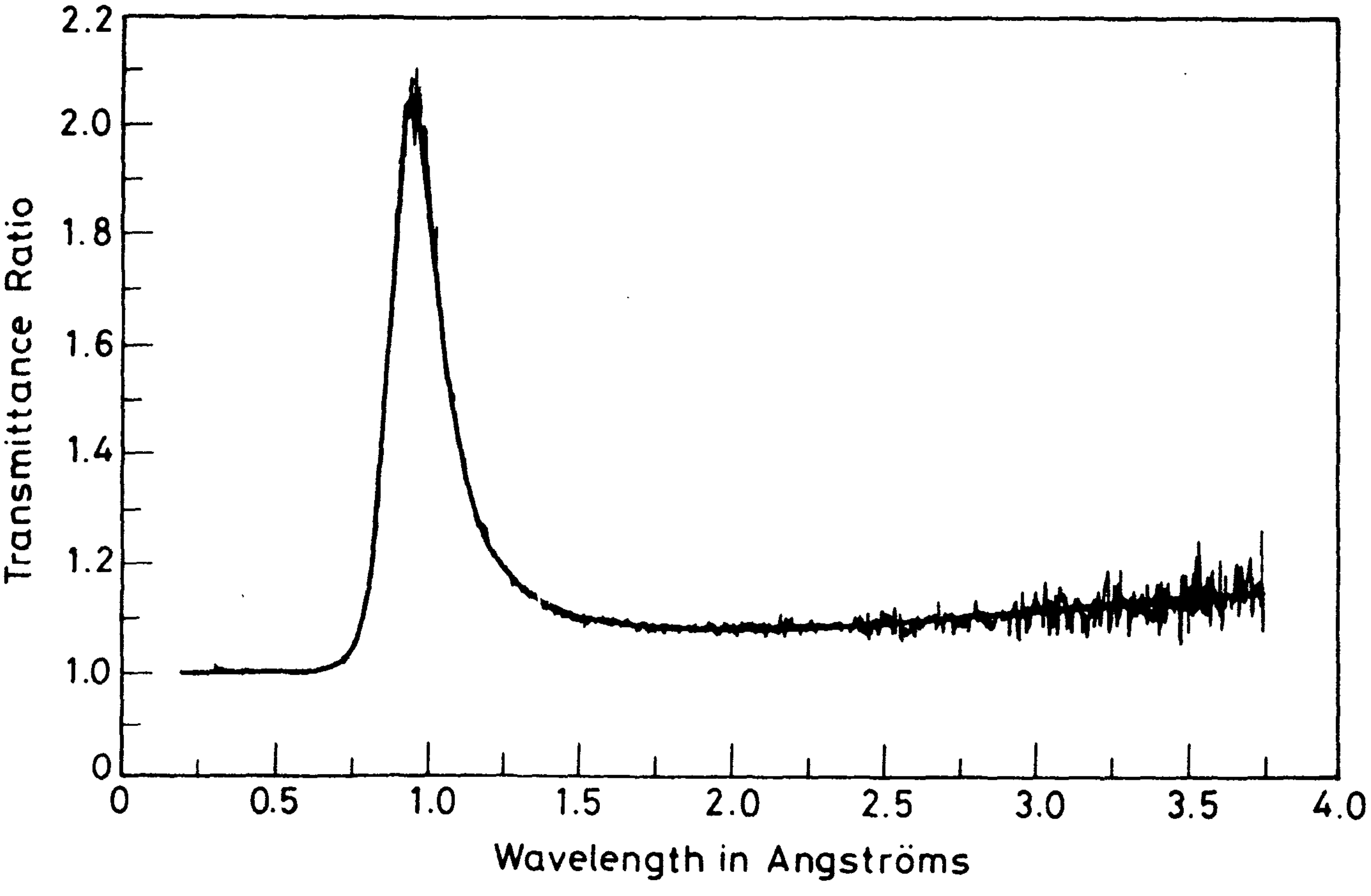

Figure 2. Transmission Ratio for $Pd_{0.95}Sm_{0.05}$, 25mK : 1.2K. Applied field = 2.5 Tesla

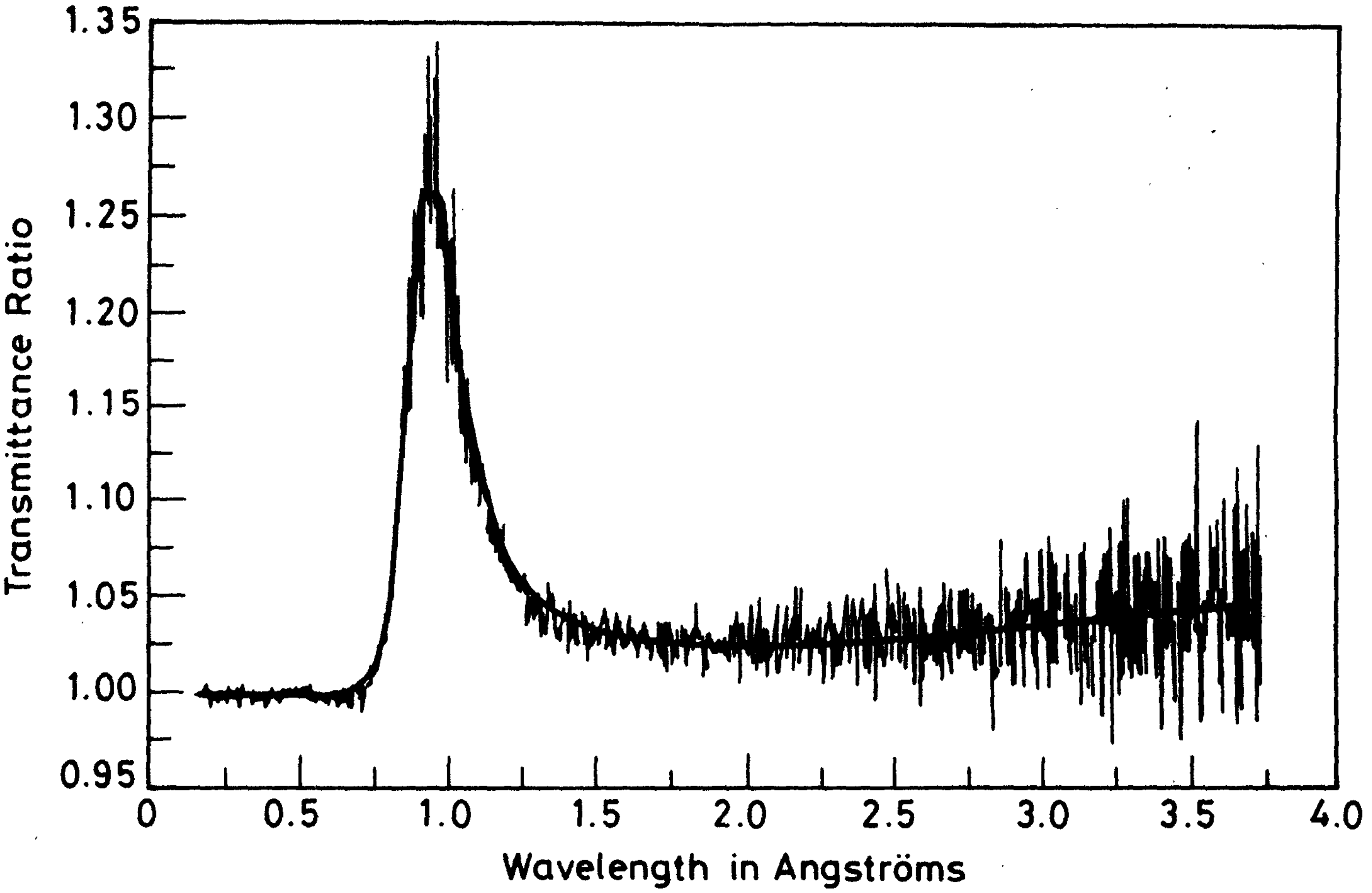

Figure 3. Transmission Ratio for $Pd_{0.95}Sm_{0.05}$, 25mK: 1.2K. Applied field = 0

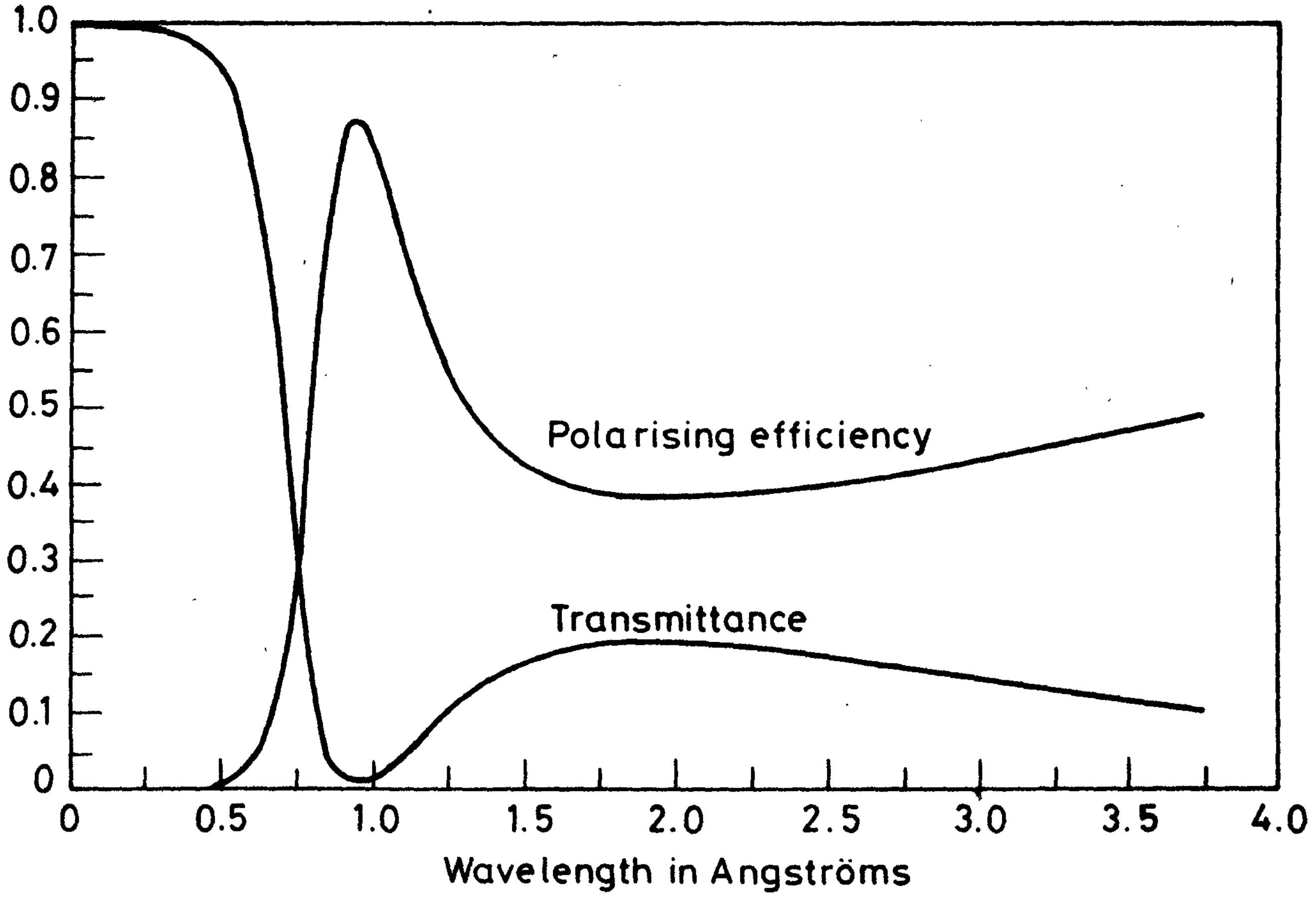

Figure 4. Polarising efficiency and Transmittance of $Pd_{0.95}Sm_{0.05}$. Temperature = 25mK. Applied field = 2.5Telsa

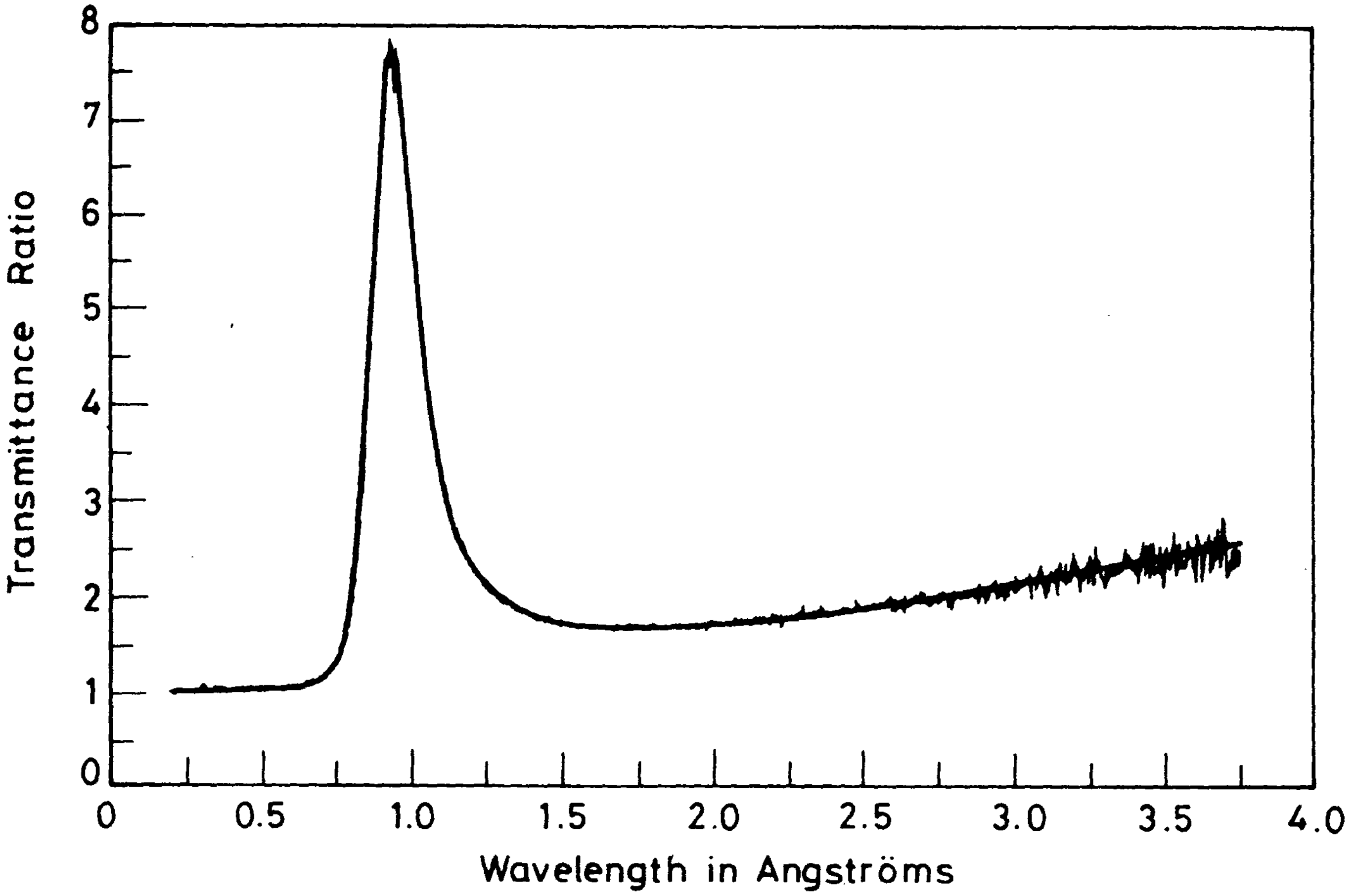

Figure 5. Measured and Fitted Transmittance Ratio for $SmCo_5$ Filter. 14mK: 1.2K

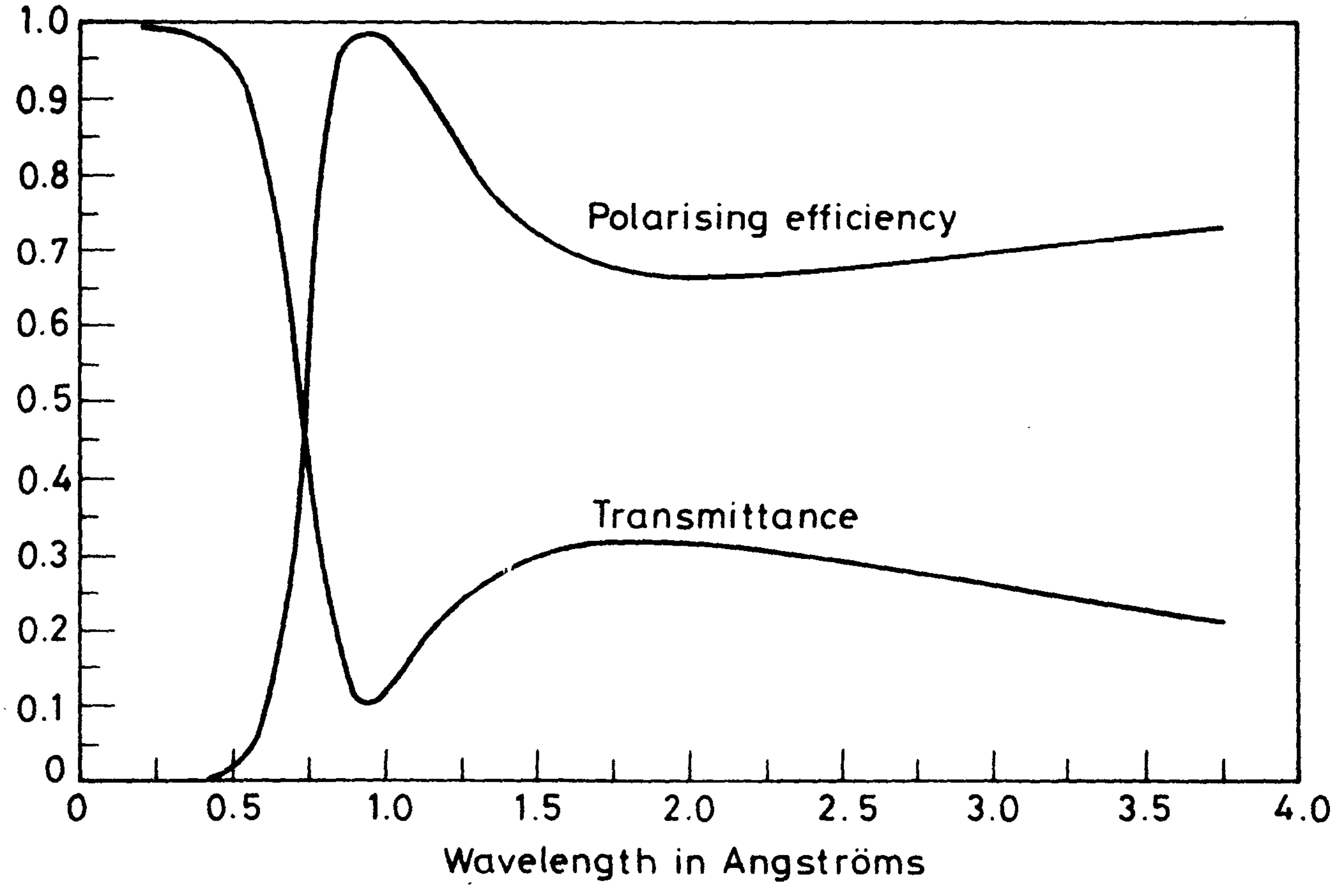

Figure 6. Polarising efficiency and Transmittance of $SmCo_5$ Filter. Calculated from Fitted Parameters

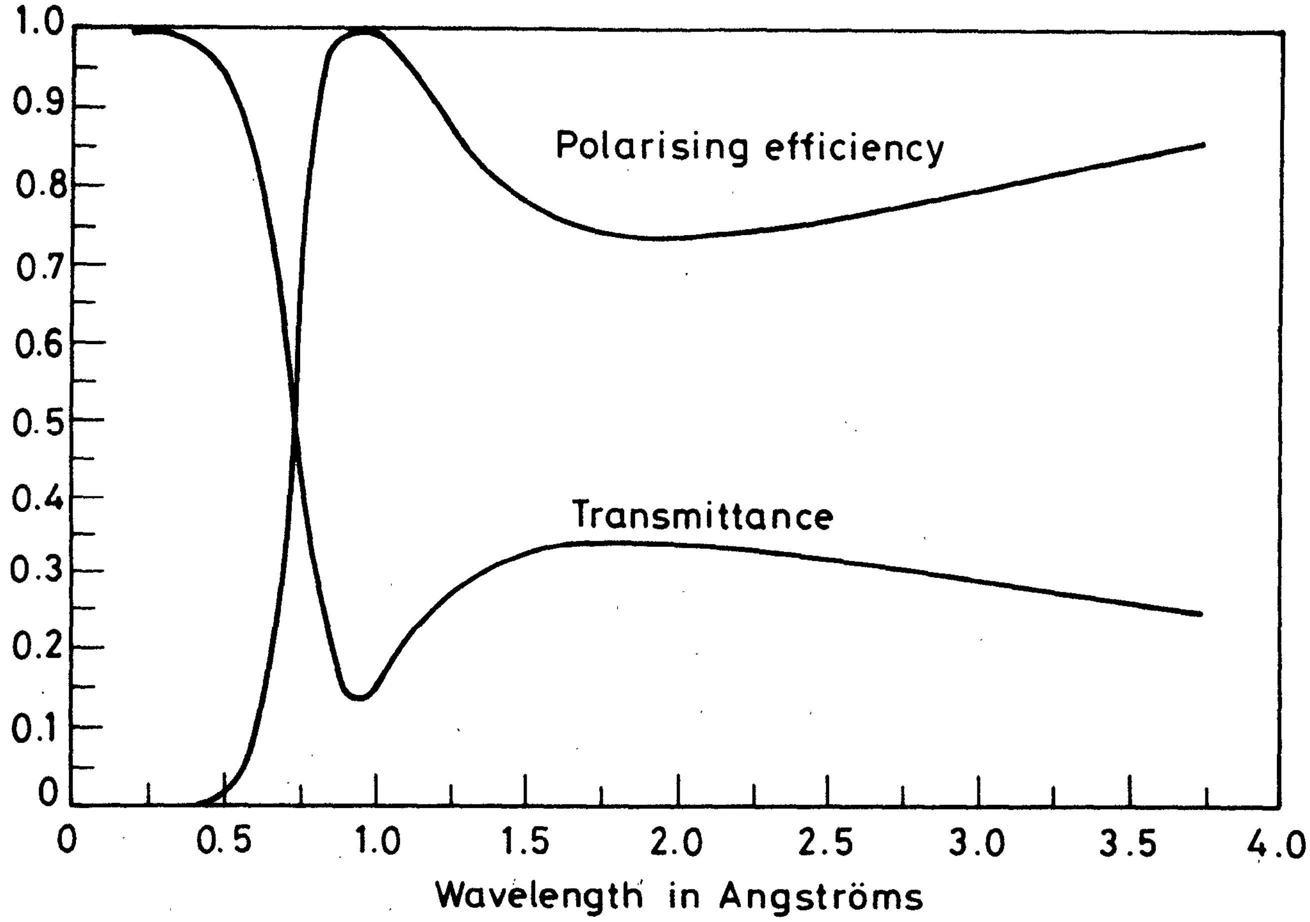

Figure 7. Polarising efficiency and Transmittance of 'Ideal' Sm Co$_5$ Filter

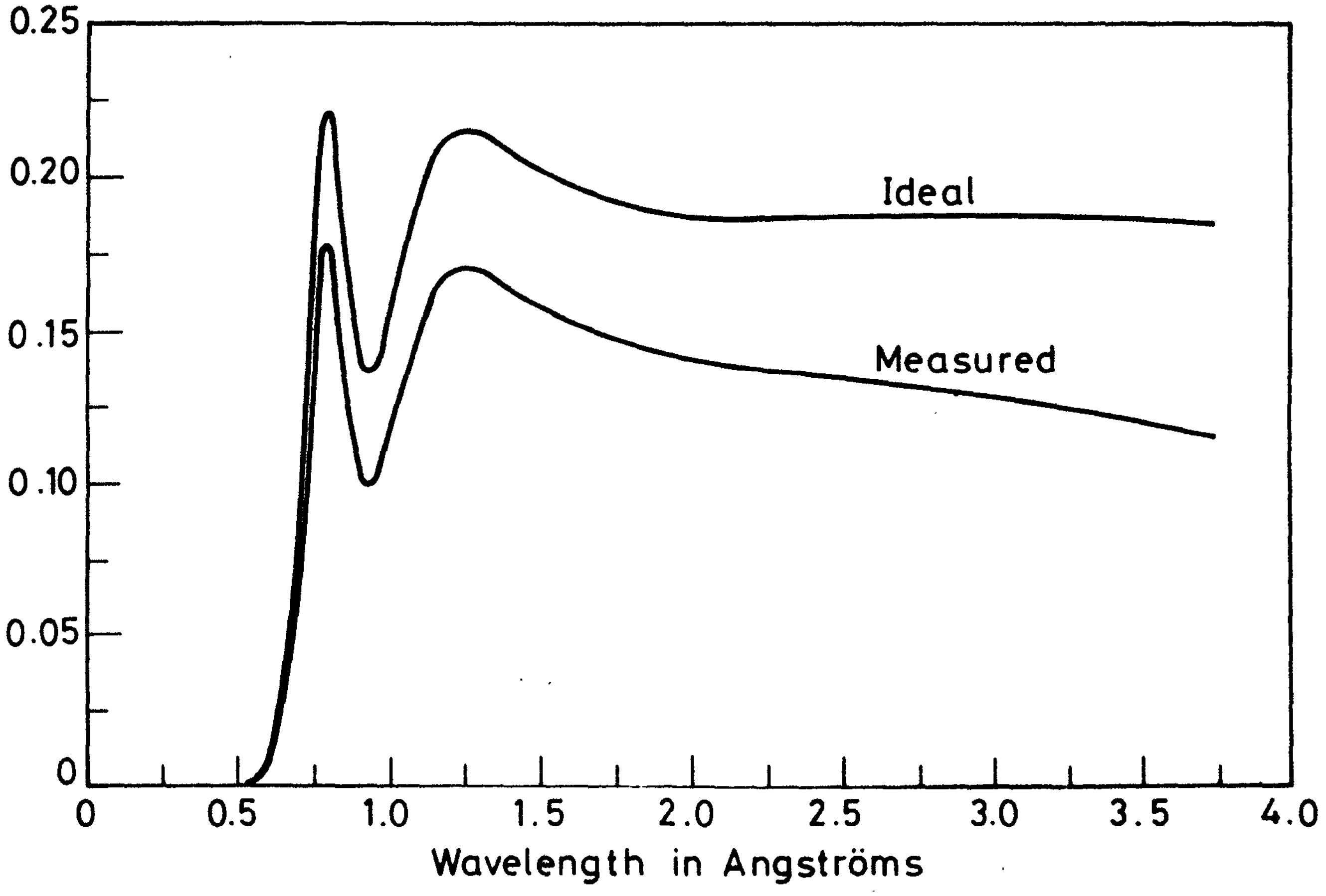

Figure 8. Measured and 'Ideal' values of the Product P^2T