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Anisotropy and Energy Dependence of the Differential Neutron Scattering Cross-Section in Vanadium

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ANISOTROPY AND ENERGY DEPENDENCE OF THE DIFFERENTIAL NEUTRON SCATTERING CROSS-SECTION IN VANADIUM

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ABSTRACT

Time of flight spectra produced by scattering from vanadium have been collected at angles of 53° and 153°.The ratio of these spectra exhibits significant anisotropy and energy dependence of the differential cross-section. This behaviour is attributed to inelastic scattering.A theoretical calculation using the harmonic approximation gives good agreement with the data but appears to slightly underestimate these effects.Some implications for the use of vanadium as a calibration sample for neutron spectrometers are discussed.

ANISOTROPY AND ENERGY DEPENDENCE OF THE NEUTRON SCATTERING CROSS-SECTION IN VANADIUM

1.Introduction

Vanadium is widely used as a calibration sample in neutron scattering experiments [1].It is essentially an incoherent scatterer of neutrons (coherent scattering cross-section, σ_c=0.02 barns,incoherent cross-section,σ_i=4.98 barns[2]) and is generally assumed to scatter neutrons isotropically with a cross-section which is independent of energy.The results of a vanadium calibration run can be used to determine the absolute cross-section of processes in other samples and also to calibrate incident neutron intensities and detector efficiencies.

In 1984 Mayers[3] calculated the differential cross-section of vanadium using the Gaussian approximation for multi-phonon processes and concluded that the cross-section was significantly anisotropic and energy dependent, due to inelastic processes. In this paper, an exact calculation of the vanadium cross-section in the harmonic approximation is compared with measurements performed at the UK spallation neutron source ISIS.The predicted anisotropy and energy dependence of the differential cross-section is observed although the theory appears to underestimate these effects.The implications for the calibration of neutron instruments are briefly discussed.

2.Theory

One would expect that vanadium,which has a body centred cubic structure ,should be well described by the isotropic harmonic approximation. For an isotropic harmonic system,the incoherent

partial differential cross-section (pdcs) can be calculated exactly if the density of states is known. The pdcs is given by[4],

$$d^2\sigma/d\Omega dE_2=(\sigma_i/4\pi)(k_2/k_1)S(q,\omega) \quad (1)$$

where σ_i is the incoherent scattering cross-section, k_1 is the incident neutron wave-vector, k_2 is the scattered neutron wavevector, $q=k_1-k_2$ is the momentum transfer, (with units $\hbar=1$), and ω is the energy transfer. The neutron scattering function $S(q,\omega)$ is given by[4]

$$S(q,\omega)=1/(2\pi)\int\exp[q^2/(2M)\{\gamma(t)-\gamma(0)\}]\exp(-i\omega t)dt \quad (2)$$

where M is the atomic mass and

$$\gamma(t)=\int Z(\omega)n(\omega)\exp(-i\omega t)/\omega d\omega \quad (3)$$

$Z(\omega)$ is the normalised phonon density of states and $n(\omega)$ is the Bose-Einstein occupation factor,

$$n(\omega)=[\exp(\omega/k_BT)-1]^{-1} \quad (4)$$

where T is temperature and k_B is Boltzmann's constant.

A Fortran program has been written to calculate $S(q,\omega)$ numerically for an isotropic harmonic system and hence via (1) the pdcs. The Fourier transforms in equation (2) and (3) are calculated using a fast Fourier transform routine. The differential cross-section for a given incident neutron energy E_1 and scattering angle θ is then calculated by integrating the pdcs over all scattered neutron energies E_2.

$$d\sigma(\theta,E_1)/d\Omega=\int(d^2\sigma/d\Omega dE_2)dE_2 \quad (5)$$

The results of a calculation of $4\pi(d\sigma/d\Omega)/\sigma_i$ for vanadium as a

function of energy at a number of different angles and at a temperature of 300K is shown in figure 1.The density of states was that derived from experiment by Glaser et al[5].The results in figure 1 agree well with the previous results of Mayers[3] except at neutron energies less than 10 meV, where the present calculation gives greater anisotropy.

The rise in the incoherent cross-section at low neutron energies is caused by the presence of k_1 in the denominator of equation (1).This causes inelastic scattering to give a relatively large contribution to the cross-section. At low energies, inelastic processes are primarily due to phonon annihilation and the rise in the cross-section correspondingly becomes small at low temperatures[3].The energy at which $d\sigma/d\Omega$ becomes isotropic then moves towards zero. At room temperature,figure 1 shows that $d\sigma/d\Omega$ is roughly isotropic at 7 meV.The apparent exact isotropy of $d\sigma/d\Omega$ at 7 meV is due to the coarseness of the plot.The curves actually cross over the range 6.4 to 7.4meV. At neutron energies < 7 meV,the cross-section is greatest at large angles while for energies > 7meV the cross-section is greatest at small angles. The cross-section is virtually independent of energy for neutron energies greater than 30 meV and as shown previously[3], at these energies the anisotropy is well described by the Placzek expansion[8]. At low energies the Plazcek expansion is inaccurate and in contrast to the theory and experimental results presented here, predicts that the differential cross-section is greatest at small angles.

3.Experiment

The experimental measurements were performed on the POLARIS beamline at ISIS.This provides a pulsed beam of neutrons with a range of energies from 0 to the MeV region although the minimum energy which can be measured is limited by frame overlap to 2

meV. A schematic diagram of the apparatus is shown in figure 2.Two He3 gas detectors, housed in identical B_4C shielding and collimation to reduce background and placed at angles of 53° and 153° were used.The sample was vanadium powder in a cylindrical thin walled vanadium can. The relevant information on instrument and sample is given in table 1 below.

TABLE 1

Incident flight path (L_1)=12.0 metres
Scattered flight path (L_2)=0.5 metres.
Sample radius=0.479 cm
Sample height=5.5 cm
No of vanadium atoms/unit vol=$3.5\times10^{22}/cm^3$.

Two measurements were performed:the first with the apparatus as in figure 2 and the second with detectors 1 and 2 interchanged. Careful measurements were taken to ensure that the alignment of the detector housings and the distance from the sample was identical in the two positions, so that the solid angle subtended by the detector was the same in each case. The scattering angles were determined from the positions of Bragg peaks in time of flight spectra collected from a nickel powder sample.

A correction of the measurements for attenuation and multiple scattering in the sample was performed, using the method of Blech and Averbach[6].The differential scattering cross-section is derived from the measured data D_m via[7],

$$d\sigma_s(\theta,E)/d\Omega=C(E)D_m(\theta,E)/A(\theta,E) \qquad (6)$$

C(E) is a factor independent of angle,which accounts for the incident spectrum,detector efficiency,number of scattering atoms in the sample etc[7].$A(\theta,E)$ is a correction factor for multiple scattering and attenuation which must be calculated for the

particular sample size,scattering angle and neutron energy.The calculated values of A at the two scattering angles are shown as a function of neutron time of flight in figure 3.It can be seen that although there is a significant dependence of A on E (due to the variation in absorption cross-section with energy) the dependence on scattering angle is weak.

4.Results

In figure 4 a typical unprocessed time of flight spectrum from a detector at an angle of 153° is shown.Figure 5 shows the ratio of spectra at 153° to spectra at 53° for detectors 1 and 2,as a function of time of flight.The advantage of taking a ratio is that the term C(E) does not need to be evaluated as it cancels. C(E) is usually determined from a vanadium calibration with the assumption that the vanadium cross-section is isotropic and independent of energy.Since it is these assumptions which are being tested,such a procedure is evidently inappropriate in this context.The only other alternative for determining C(E) is by calculation which is difficult and uncertain. A further advantage of taking a ratio is that due to the weak angular dependence of $A(\theta,E)$, any small errors in the calculation of A introduced by the assumptions implicit in the treatment of Blech and Averbach, should have little effect. It can be seen that the agreement between the ratios from the two different detectors is very good.

In figure 6 the mean of the two ratios shown in figure 5 is plotted as a function of energy. It was assumed in the data analysis that the energy of the incident neutrons was determined by their time of flight as if they had been elastically scattered. The error introduced by this approximation should be negligible ,as $L_1 \gg L_2$ (see table 1) so that the time of flight is determined primarily by the incident energy.[3] The sharp peaks and troughs in the ratio, which can also be observed in figure 5, are caused by Bragg peaks due to the weak coherent scattering

cross-section.The peaks appear at lower energies at 153° than at 53° so that in the ratio 153°/53° they appear as peaks at low energies and dips at higher energies. The calculation slightly underestimates the anisotropy and variation of the differential cross-section with energy.However the data clearly confirms the predicted qualitative behaviour of $d\sigma/d\Omega$ and is in reasonably good quantitative agreement.

The ratio of the spectra at the two angles will be slightly affected by detector efficiency effects. Neutrons which lose energy when scattered are more likely to be detected than elastically scattered neutrons, as detector efficiencies increase with decreasing neutron energy. High energy incident neutrons scattered at 153° will lose more energy than neutrons scattered at 53° and thus the measured ratio of scattering intensities (153°/53°) will be higher than the true ratio. Conversely at low incident energies, inelastic scattering is dominated by events in which the neutron gains energy from the sample. The neutron gains more energy at 153° than 53° and is therefore less likely to be detected. Thus the measured ratio at low energies is less than the true ratio of scattered intensities. It is clear that a correction for this effect will worsen the agreement with calculation at both high and low energies. The size of the correction at high energies was estimated as one or two percent ie the ratio would change from 0.91 to $\simeq$0.90. At low energies the correction is small as the detectors are almost 100% efficient.

The source of the disagreement between theory and experiment is unclear. There may be inaccuracies in the density of states which was used in the calculation, although it seems unlikely that this would significantly change the results. The differential cross-section is calculated from an integration over ω and should be insensitive to the detailed behaviour of the density of states. The calculation also gives very good agreement with the Placzek expansion for $d\sigma/d\Omega$ at high energies. It therefore seems

more likely that there is a systematic error in the measurement than in the theory. Not withstanding these comments, agreement between theory and experiment is clearly rather good.

5.Discussion

The anisotropy and variation with energy of the vanadium differential cross-section has implications for many neutron experiments where vanadium is used as a calibration sample.Some of these have been discussed previously[3].We mention two specific examples which are relevant to the analysis of powder diffration data.

The POLARIS instrument,which is a medium resolution high count rate powder diffractometer, is a good example of an instrument where the energy dependence of the vanadium cross-section is significant. Measured powder diffraction spectra must be normalised to allow for the variation of detector efficiencies and incident spectrum with energy, before the true relative magnitude of different Bragg peaks in a time of flight spectrum can be obtained. If it is assumed that the vanadium scattering cross-section is independent of energy,the spectrum intensity will be overestimated at low energies. The scattering angle on time of flight powder diffraction instruments such as POLARIS, is generally high as this reduces the angular contribution to the resolution and it is at high angles that the variation with energy of $d\sigma/d\Omega$ is greatest.As can be seen from figure 1 this can lead to an underestimate in the magnitude of Bragg peaks by up to 20%.

A neglect of the anisotropy of the vanadium cross-section will cause errors on neutron instruments where the incident beam is monochromatic and the scattering intensity is measured as a function of angle.For example on a powder diffraction instrument a calibration using vanadium will give errors in the magnitudes

of Bragg peaks measured, unless the anisotropy in the vanadium cross-section is allowed for.

In conclusion we note that the predicted anisotropy and variation with energy of the differential cross-section for scattering of neutrons from vanadium, has been observed and shown to be in reasonable agreement with calculation. This is itself of some physical interest, but it also has direct relevance to the calibration of neutron spectrometers.Experimentalists should be aware of these effects so that they can evaluate their significance and correct their data if necessary.

Acknowledgements

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FIGURE CAPTIONS

Figure 1 Plot of relative differential cross-section, $4\pi(d\sigma/d\Omega)/\sigma_i$, for vanadium at 300K as a function of energy, at angles 10°,50°,90°,130° and 170°.

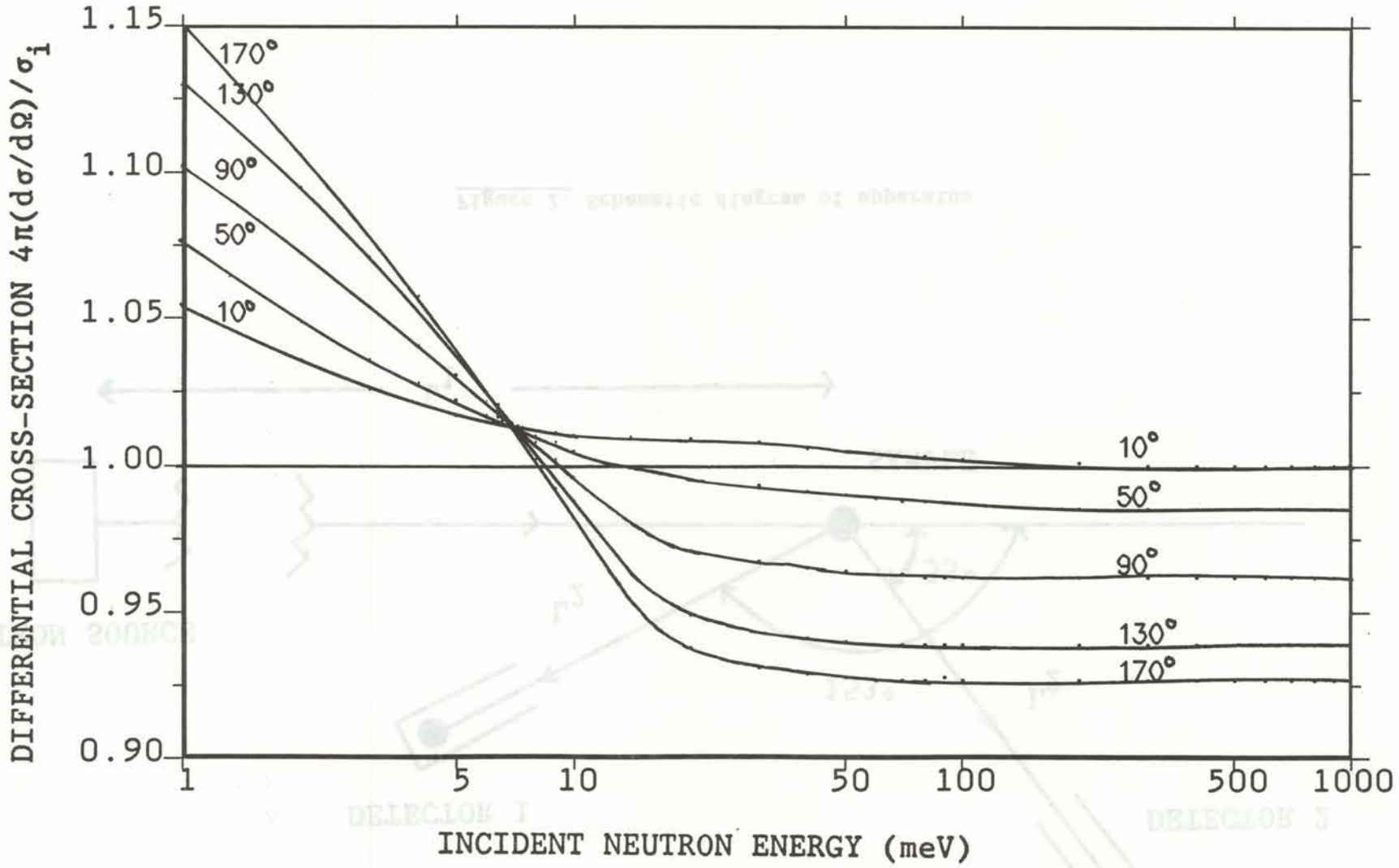
Figure 2. Schematic diagram of apparatus

Figure 3. Correction factor A as a function of time of flight; + scattering angle=53°,---- scattering angle=153°.

Figure 4. Time of flight spectrum taken with single He3 detector at a scattering angle of 153°.

Figure 5. Ratio of scattering 153°/53°. Points with error bars are from detector 1,line is detector 2.

Figure 6. Line is mean of measured ratios in figure 4.Points are calculated ratios of differential cross-sections; 153°/53°.



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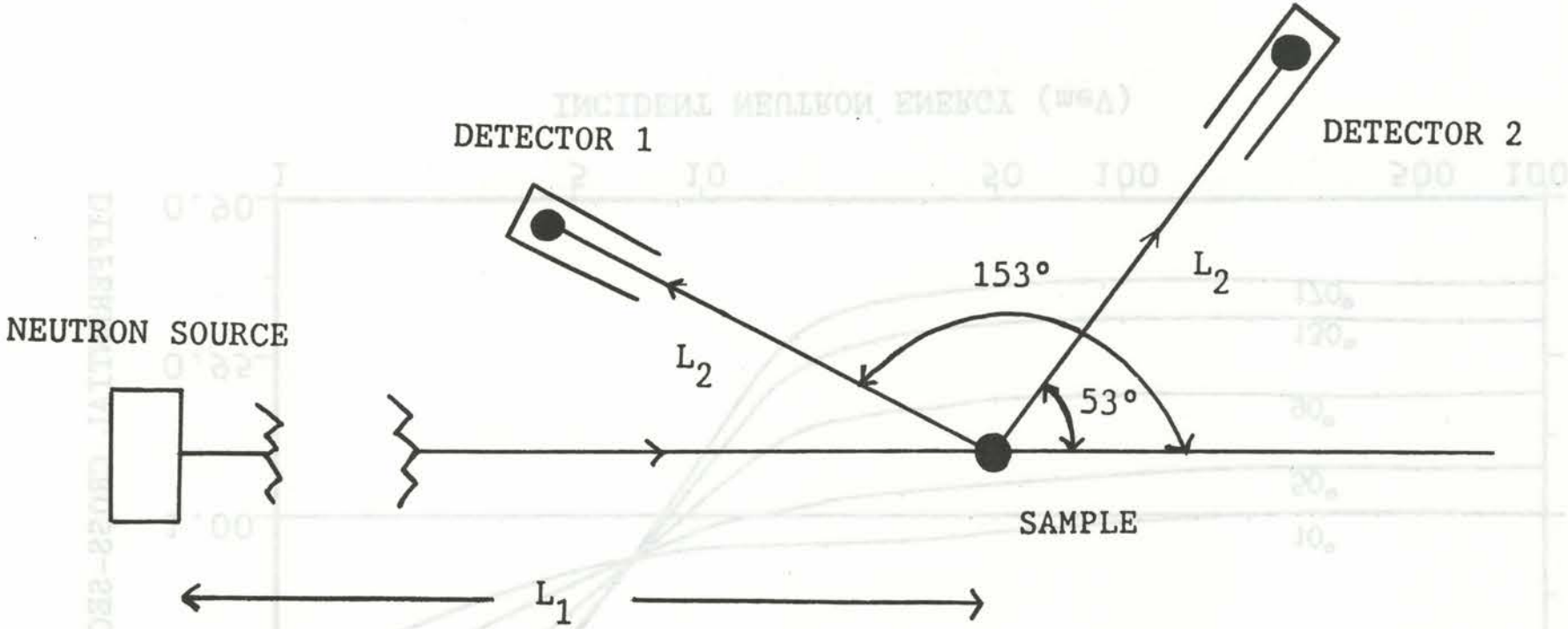
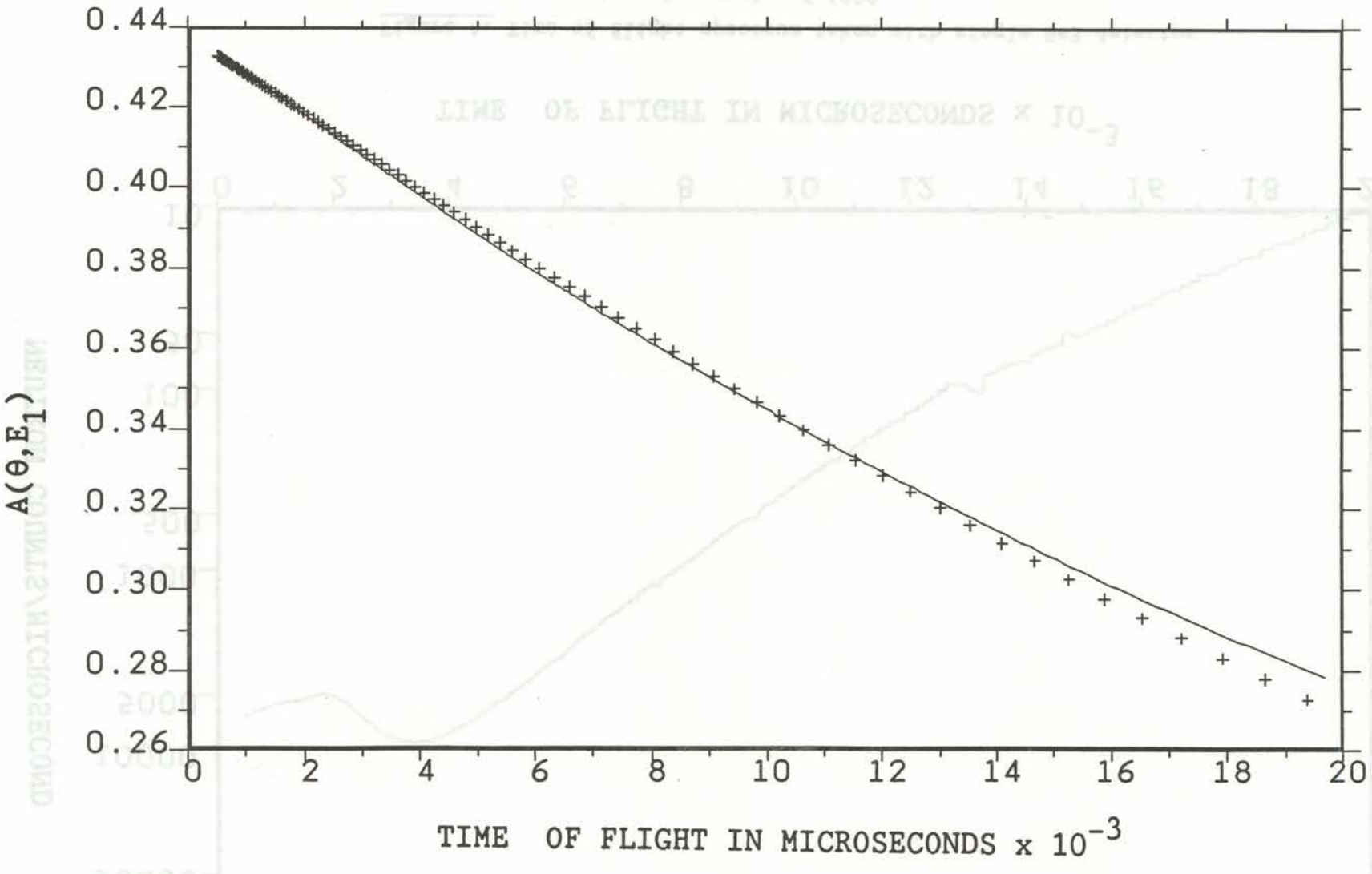


Figure 2. Schematic diagram of apparatus



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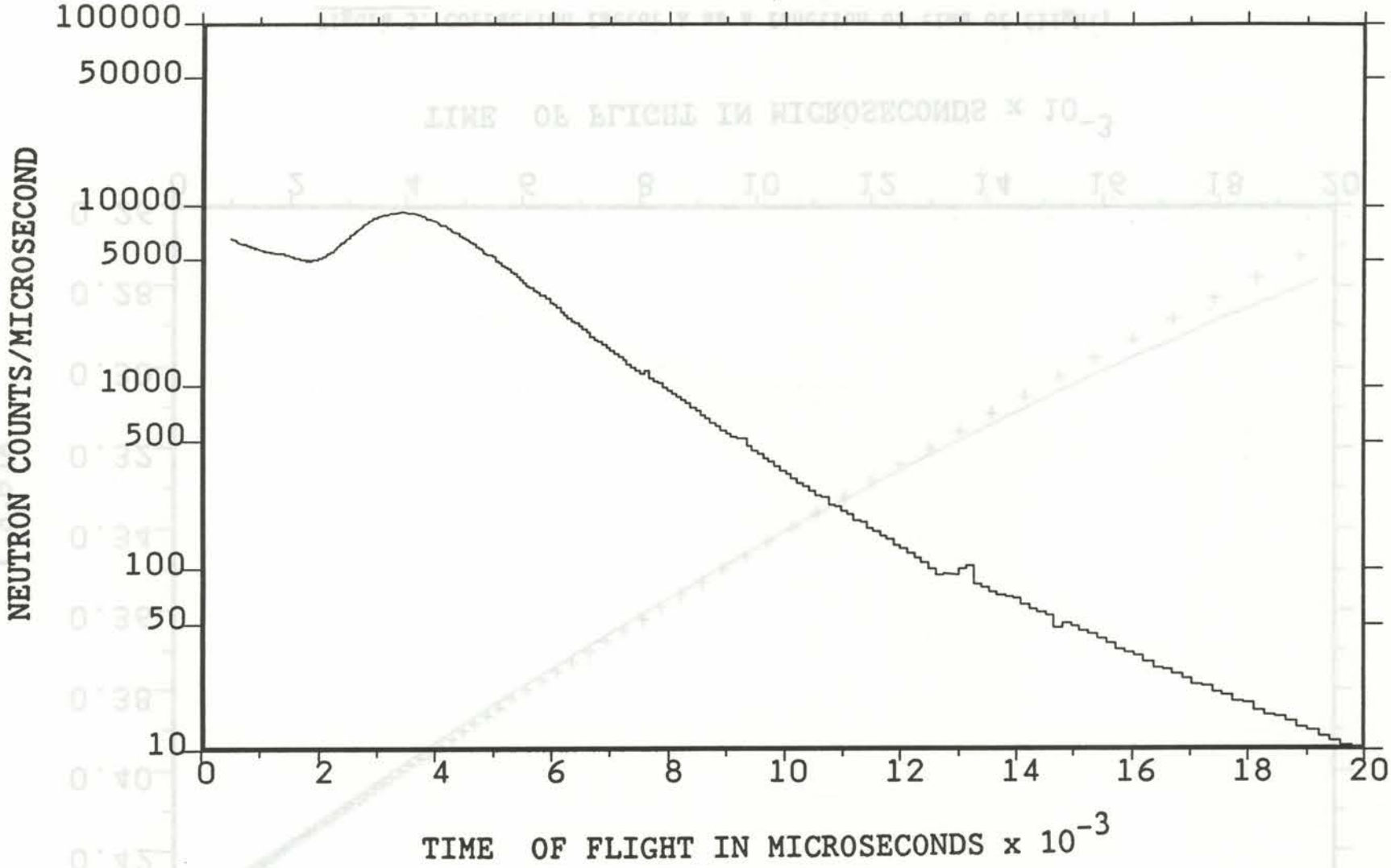

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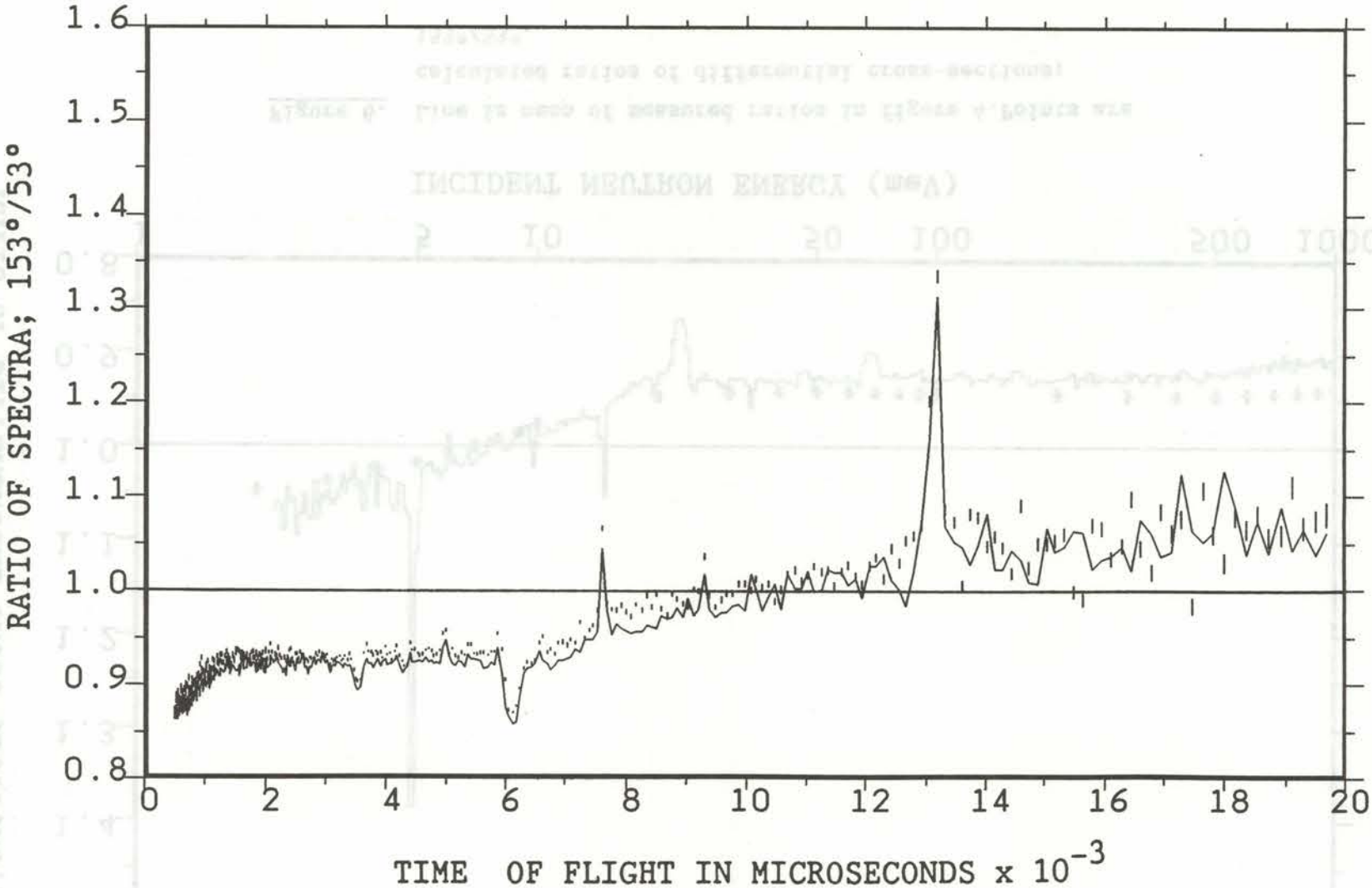

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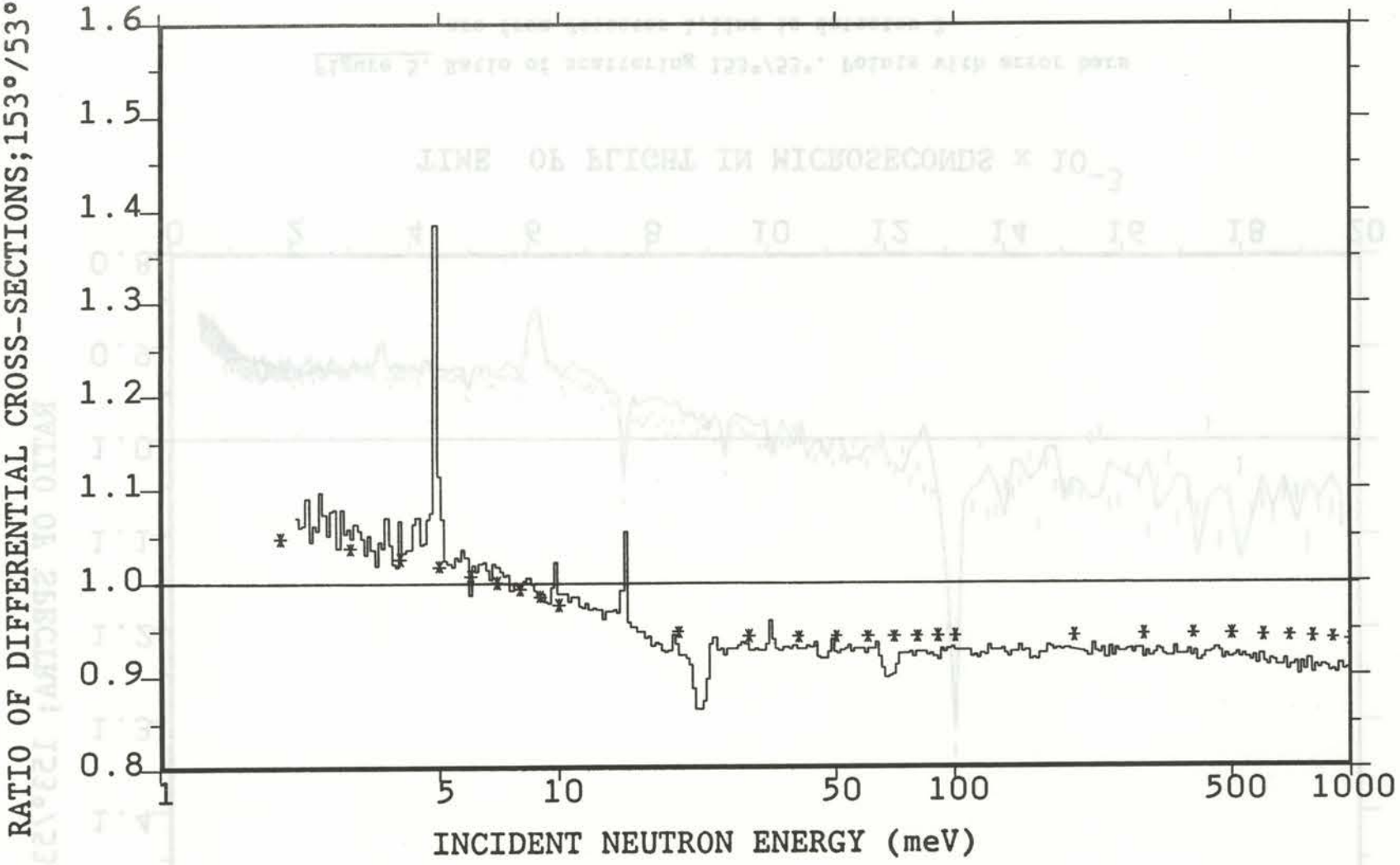

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