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Solid state effects on the reaction cross-section below a few eV

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Introduction

The use of the ideal gas model for Doppler ensures that a 1/V nuclear cross-section remains 1/V when Doppler broadened. However the ideal gas model does not take into account solid and chemical binding effects.

The reaction energy of neutron of mass n incident on a free nucleus of mass M is

$$E_R = E_n \left(\frac{M}{(M + n)} \right)$$

where E_R is the energy at which the reaction takes place and E_n the incident kinetic energy of the neutron. The above is only valid for a mono-atomic ideal gas and is not valid for nuclei bond in a lattice or compound. Energies of up to a 100 meV are required to knock a nucleus out of the crystal lattice or break the chemical bonds in the compound. If this energy is not available, the result is that all or nearly all of the kinetic energy of the incident neutron goes into the nuclear reaction. For a 1/V nuclear cross-section this will result a non 1/V 'Doppler' broadened cross-section in the incident neutron energy range up to a few eV.

REFIT calculations

Two Doppler broadening subroutines have been added to the neutron cross-section analysis program REFIT. Previously only the ideal gas model was available in the program. The two new subroutines enable the Doppler broadening calculation to be carried out using approximations to the phonon spectrum associated with the solid sample of the nucleus under examination. One was written by Meister based on the Einstein model and the other a simplified version with neutrons gaining from or losing energy to the lattice in discrete amounts.

The subroutine used to calculate the Einstein model for Doppler broadening was given to me for inclusion in REFIT about 1995 and should now be replaced with Meister latest version. The other subroutine was written in late 1993 but not used seriously in any fits until early 1995 when some transmission measurements on a sample ^{237}Np became available. These measurements were carried out at Geel by Brusegan and a group from Saclay. The residual from the fits using the ideal gas model across the resonances below ~5 eV showed some oscillations centred about the peak energies of each resonance (see figure 1). In the fits carried out with phonon models the oscillation were much reduced (see figure 2).

As a recent test of the Doppler broadening in the program a pure 1/V reaction cross-section was broadened using both the ideal gas model and the phonon models.

Using a nuclear 1/V reaction cross-section of 100 barns at 1 eV and a target mass of 10 AMU, the ideal gas model gave a broadened cross-section of 104.92 barns divided by the square root of the energy over the calculated range from 1 meV to 100 eV to with in the accuracy of the code ($\sim$1 in 10^5). The square root of ten plus the mass of the neutron (1.008665) divided ten is 1.04922. The phonon models for Doppler broadening at a sample temperature of 25.3 meV, a Debye temperature of 25.3 meV and a single phonon of energy of 12 meV, both gave none 1/V cross-section below a few eV. The product of the cross-section and the square root of the energy is shown in figure 3 and decreases from a value of 102.45 at 1 meV to 102.30 at between 30 and 40 meV, then increases to a value of 104.92 at $\sim$4 eV and remaining constant at this value to the maximum of the calculated range. The exact shape depending on the sample temperature and the solid state parameters.

Measurements!

The changes to the shape of the reaction cross-sections will be best seen in measurements on solid samples of light nuclei in the energy region below a few eV. A quick look through the EXFOR data files of isotopes expected to have 1/V reaction cross-sections showed that there were few suitable measurements on solid samples.

The capture cross-section of hydrogen is assumed to be 1/V and much larger effect should be seen in solid compounds of hydrogen because of the small nuclear mass. In the EXFOR data file there is only one measurement listed of the capture cross-section and that is between 20 and 64 keV.

There are several measurements of the (n,p) reaction cross-section of ^{3}He which show that the cross-section does have the 1/V expected form up to neutron energies of several keV.

The ^{10}B and ^{6}Li (n,α) cross-sections for both nuclei are used as standards for measurements on other materials and have been studied extensively. The measurements show that the (n,α) cross-section has the 1/V form from a few eV to several hundreds of eV. Below a few eV there are few measurements on solid samples, measurements on solutions and gaseous samples indicate 1/V form to the cross-section.

The measurement by Czirr and Carlson (Knoxville Conf. 1979) of the ratio of counts from a B F_3 ionisation chamber to those from a thin natural lithium glass scintillator show an increase from a value of 0.9880$\pm$0.0044 at 610 eV to unity at 12 eV then to a value of 1.015$\pm$0.0016 at 1.12 eV. This may be an indication of either a difference in form of the nuclear cross-section for ^{6}Li or ^{10}B from the expected 1/V or some solid state effects in the lithium glass. The observed change from 12 eV down to 1.12 eV is about three times the calculated value using the phonon model for a metallic sample of lithium and assuming the B F_3 is and ideal gas. Also the calculated value was constant above $\sim$4 eV.

A measurement on samples of ^{10}B carried by Brisland, Croft, Bond and myself reported at Julich conference in 1991 was not recorded in the EXFOR files. The measurement was used to determine the ^{10}B content of six metallic samples and covered the energy range from

~0.15 to ~ 200 eV. The measured transmission of the thinnest sample is shown in figure 4 indicating a small increase in the observed transmission below ~0.5 eV, above the 1/V extrapolation from fits to the region above 0.5 eV. The thicker samples did not have a measurable transmission below ~0.5 eV. This increase at the time was thought to be due to background problems associated with the measurement of transmission as small as or smaller than 0.01. The measured signal to background with a thin rhodium and cadmium sample at 1.25 eV was better than 1000 to 1 and counts were in agreement with those observed below the cadmium cut off. The observed increase in the transmission if correct, indicates a decrease in the total cross-section of about 1% over the neutron energy range from 0.5 to 0.2 eV. This decrease in not inconsistent with the value of ~1.5% given by the REFIT phonon model calculation for metallic boron.

As the fractional change in 1/V cross-section is approximately proportional to 1/2M, to see any effect in the cross-sections of nuclei heavier than ~20 would require very accurate measurements. For the most isotopes there are accurate measurements at 25.3 meV and with a Maxwellian spectrum but measurements covering the range from thermal to several eV are not accurate enough check the theory or calculation.

Conclusions

If the low energy neutron cross-sections calculated in REFIT are true, then the effect is best seen in the reaction cross-sections of the light nuclei. The effect may be masked as many integral and differential measurements of the reaction cross-sections have been carried out relative to the ^{6}Li and ^{10}B (n,α) cross-section. However only the ones using thin solid standard samples in the energy region below a few hundred meV will be affected, as the reaction cross-section is proportional to the 'known' cross-section of the standard. In measurements using a thick standard sample, the reaction yield does not depend so much on the cross-section as all the incident neutrons are absorbed. Measurements using solution or gaseous standard samples may be unaffected as solid state effects will be minimal, but for compounds there may be some deviations due to chemical binding.

This also raises some question about nuclear reactor calculation where the 1/V reaction cross-section is assumed to be nearly independent of the temperature.

What changes will be caused by a small decrease in the 1/V cross-section in the region below a ~100 meV, whose magnitude is dependent on the temperature?

What is the energy dependence of the hydrogen capture cross-section in the energy region below 1 eV?

Checks need to be carried out on the theory and the program.

One of the easier experiments would be to measure the reaction cross-section of thin sample of ^{10}B metal relative to the gas ^{3}He at best or B F_3. A more careful look to see if any experiments have been carried out on any of the light nuclei to find the energy dependence of a reaction cross-section from a few meV to several eV that are not listed in EXFOR or CINDA.

Appendix

In the phonon approximation the cross-section is sum of two components
(i) the sum of the interactions of a neutron with a nucleus were it can gain from or lose to the lattice energy in discrete intervals.
(ii) a multi phonon component that is approximated by the ideal gas model

The equation given below are simplification and modified forms of equation given in chapter 4 of the book by Marshall and Lovesey Theory of Neutron Scattering, Clarendon Press 1971.

The fraction of each component depends on the Debye-Waller factor that is calculated from the incident neutron energy and the thermodynamic properties of the sample.

$$W = \frac{E_n}{M} \frac{3T}{\theta_D^2} \left[1 + \alpha \left(\frac{\theta_D}{T} \right)^2 - \beta \left(\frac{\theta_D}{T} \right)^4 \right]$$

where W is the Debye-Waller factor at a neutron energy E_n at a sample temperature of T and the Debye temperature of the sample is θ_D , α and β are constants of the crystal lattice

The fraction of interactions

$$F_0 = \exp(-2W)$$
$$F_{\pm 1} = 2W \exp(-2W)$$
$$F_{\pm 2} = 1/2\ (2W)^2 \exp(-2W)$$

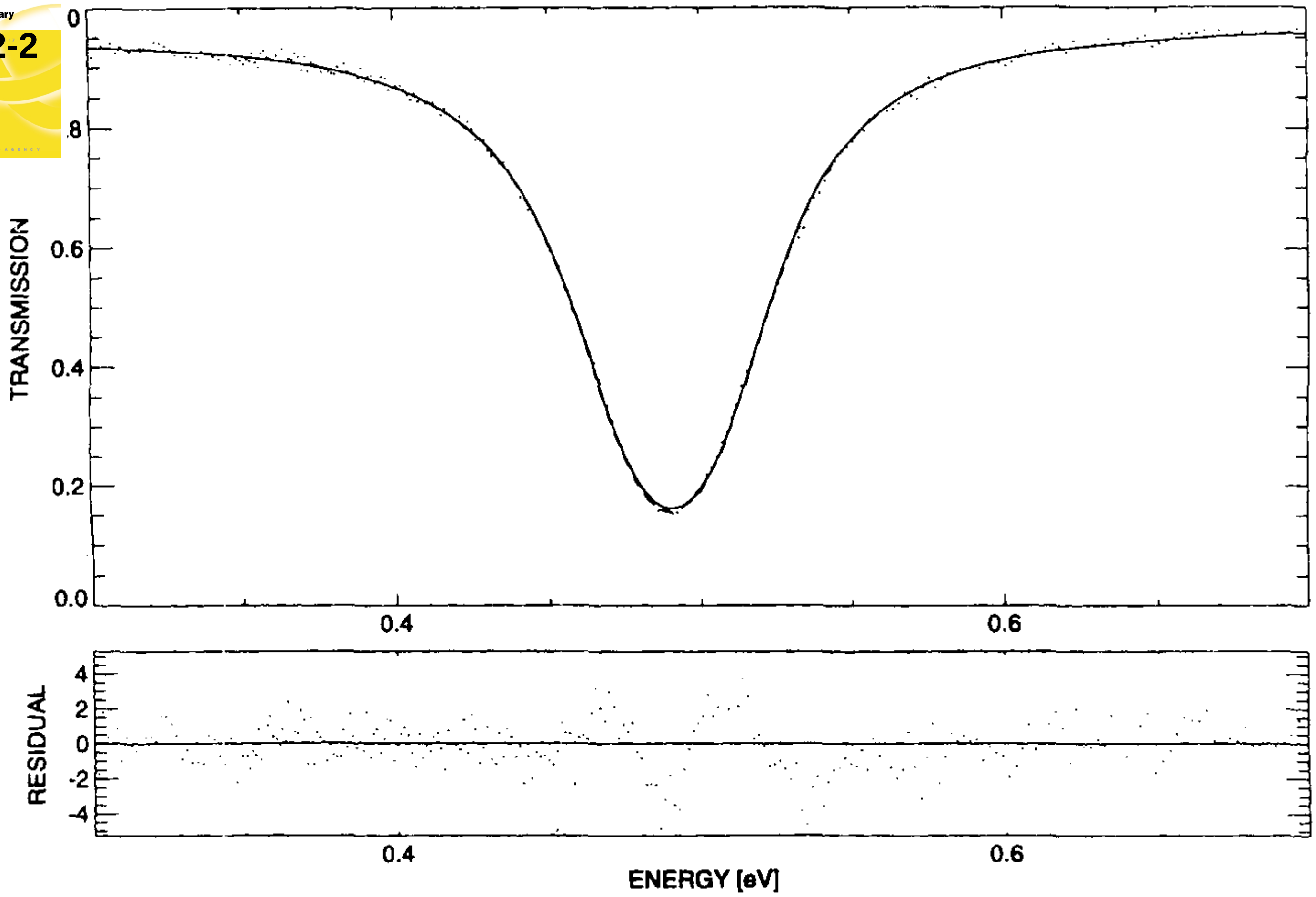
where F_0 is the fraction of neutrons that neither lose nor gain energy from the lattice and $F_{\pm I}$ is the fraction that either gains from or loses to the lattice I phonons.

The cross-section $\sigma(E_n)$ at incident neutron energy E_n is given as follows

$$\sigma(E_n) = \sum_{j=-k}^{j=+k} F_i\ \sigma(E_n + jE_p)$$

where $\sigma(E_n+jE_P)$ is the nuclear cross-section at an energy of E_n plus or minus j times the phonon energy E_P. K being the number of phonons and needs rarely to exceed 5

The ideal gas model is then used to broaden the remain fraction $1 - \sum F_j$ of neutrons.



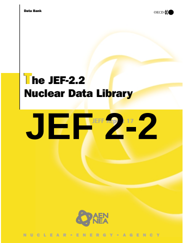
This shows the fit to the ^{237}Np transmission data in the region of the 0.5 eV resonance. These fits were carried out covering an energy range from 0.3 to 2.7 eV using the ideal gas model for the Doppler broadening.

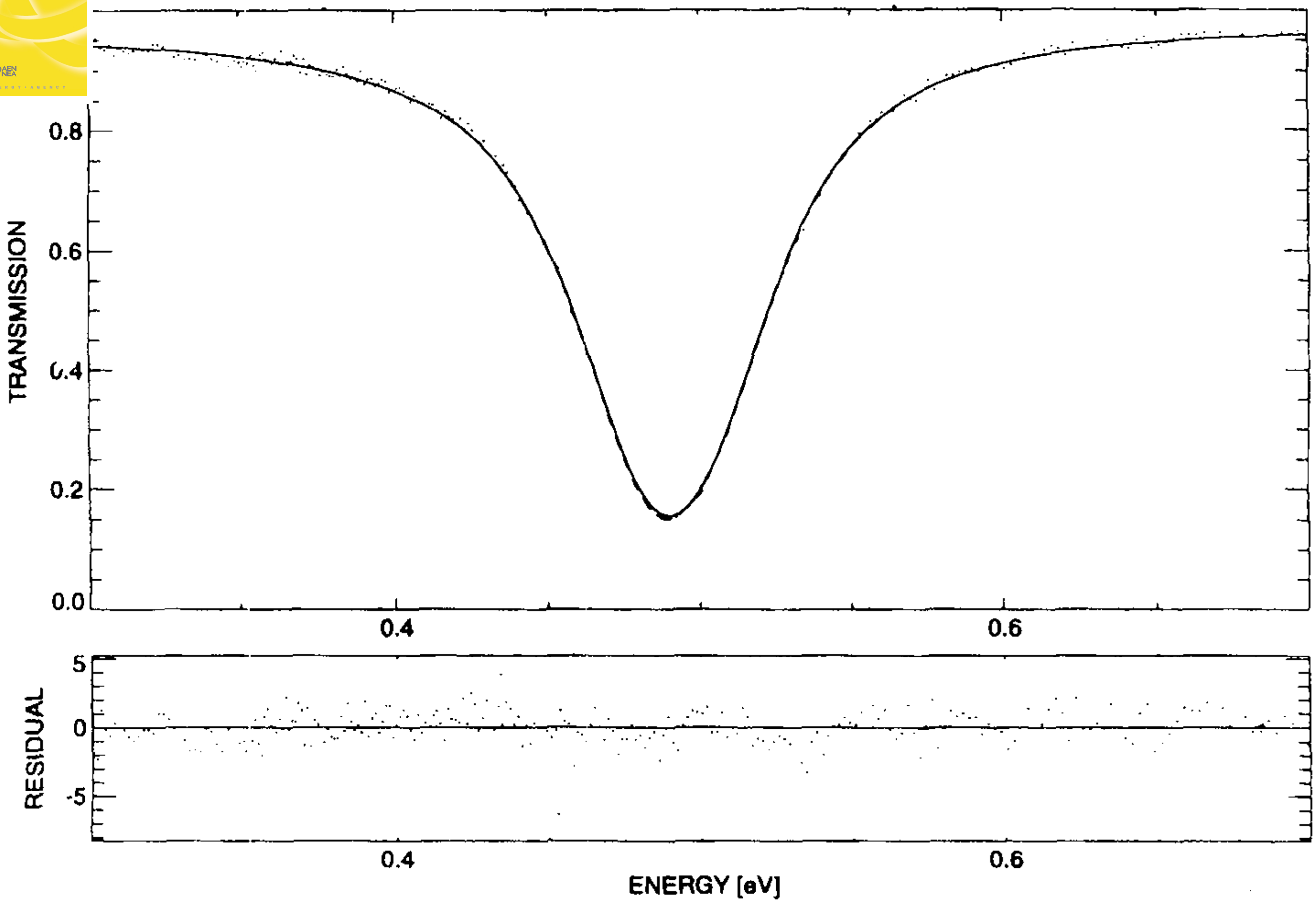
Effective sample temperature = 24.41±0.25 meV
Debye temperature = 16.2 meV fixed

Resonace energy = 0.49064±0.000307 eV
Radiation width = 38.76±0.127 meV
Neutron width = 0.0472±0.00012 meV

Number of data points = 2131
Number of variables = 20
Chi-squared per data point = 1.427

Fig. 1





This shows the fit to the ^{237}Np transmission data in the region of the 0.5 eV resonance. These fits were carried out covering an energy range from 0.3 to 2.7 eV using the phonon model for the Doppler broadening.

Effective sample temperature	= 27.40±0.63 meV
Debye temperature	= 15.26±0.78 meV
Phonon energy	= 3.98±0.94 meV
Resonace energy	= 0.49174±0.000061 eV
Radiation width	= 41.97±0.071 meV
Neutron width	= 0.0485±0.00012 meV
Number of data points	= 2131
Number of variables	= 20
Chi-squared per data point	=1.075

Fig 2.

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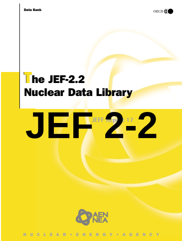
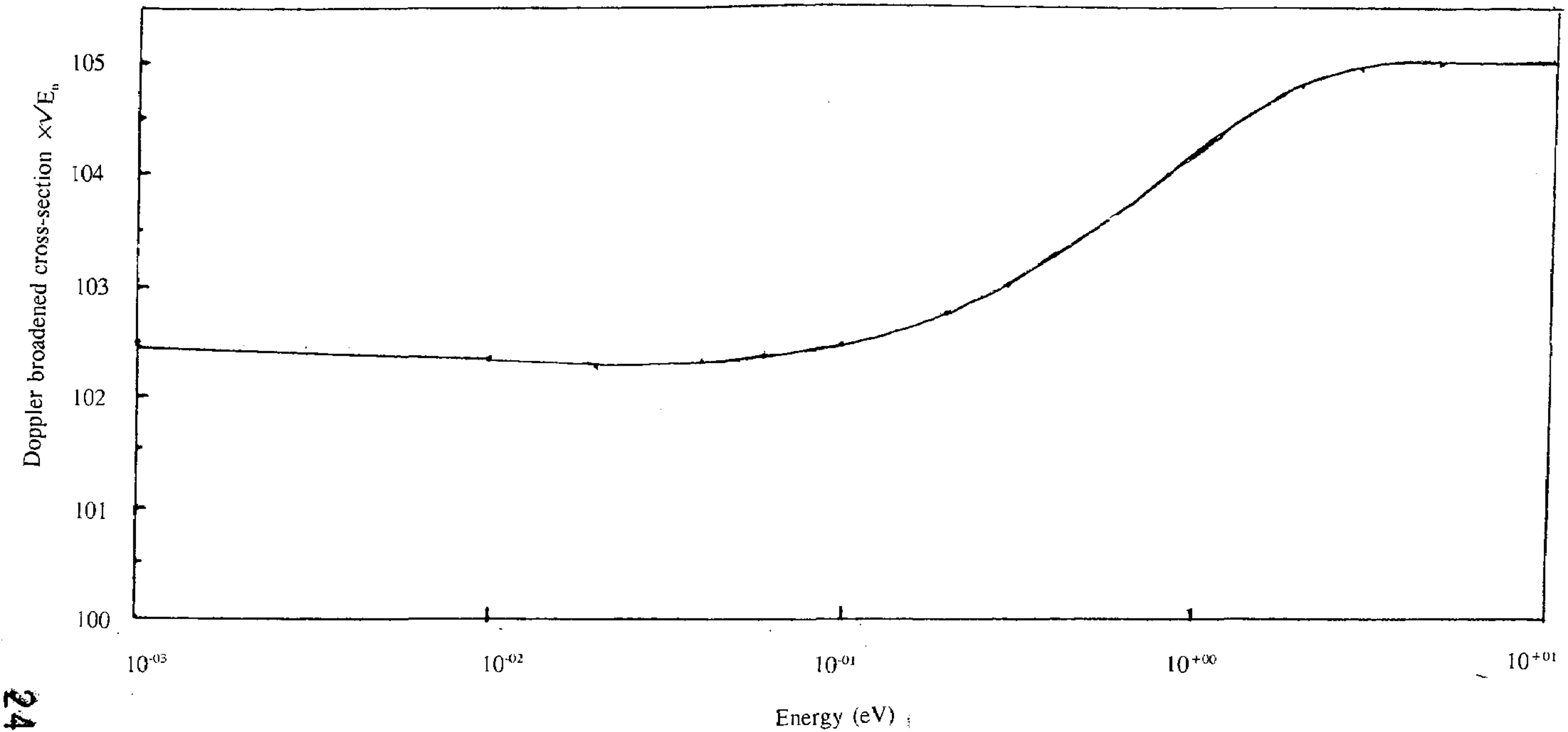
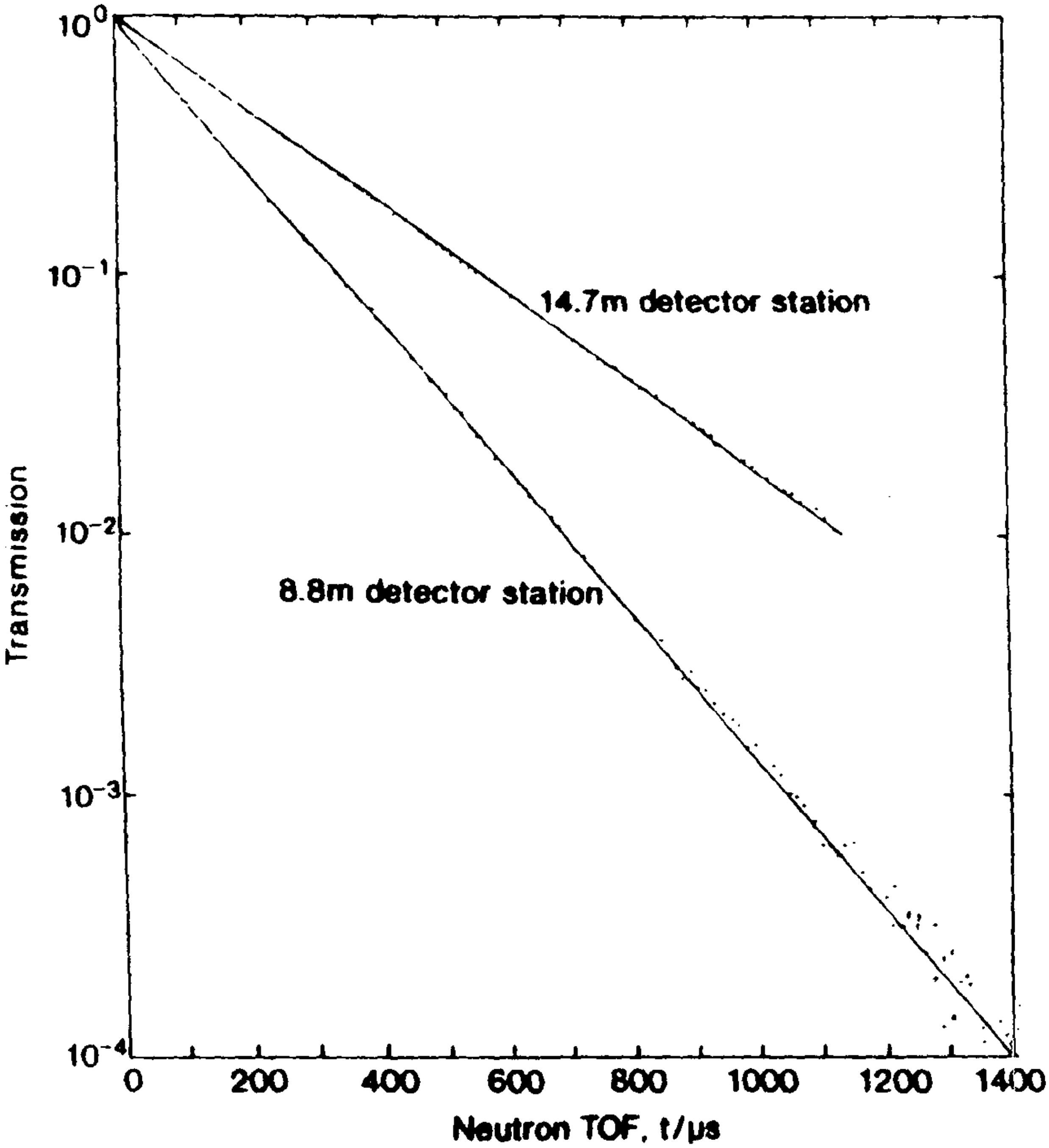




A 1/V nuclear cross-section Doppler broadened using a phonon model for nuclear mass of 10 AMU. The nuclear cross-section =100/√neutron energy barns, Sample temperature 25.3 meV; Debye temperature 25.3 meV, Phonon energy 12.0 meV

Fig 3



A plot of the measure neutron transmission for a ^{10}B metallic sample ($n = 6.941 \times 10^{-3}$) The solid line is linear fit to the region from 0.5 to 200 eV. or time 8.8m 45 to 900 μsec. and 75 to 1080 μsec.

Fig 4.