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Some results on the Doppler broadening of neutron resonances in U and UO_2 from measurements at low temperature

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Andreas Meister, CEA, CE Cadarache, France

Introduction

Within an EC Network "Study of solid state properties by neutron resonances", enclosing activities of CE Saclay, ENEA Bologna, CE Cadarache, University of Delft, IRMM Geel, ILL Grenoble, and the University of Kiel, an experiment has been set up at the GELINA pulsed neutron facility to measure low energy neutron resonances with high resolution and high accuracy to provide experimental data for studying the Doppler broadening of neutron resonances and related topics within a wide range of temperatures. The GELINA facility is besides the Dubna pulsed reactor, which has advantages in the low-energy range, the most powerful facility for such measurements in Europe.

The features of the experiment are the following:

- accelerator driven pulsed neutron source with a continuous energy spectrum and a high source strength
- neutron time-of-flight spectroscopy on a flight path of 26.5m (it is variable up to 400m)
- less than 15meV resolution (FWHM) at 6eV with 26.5m flight path, a condition for low temperature measurements
- low background (0.5 to 1% at 6eV in the actual measurements)
- equipment for cooling the samples to a stabilised temperature between 10°K and room temperature
- a furnace for high temperature measurements with an operating temperature up to about 3000°K is in preparation
- equipment for sample changing.

After doing some testing experiments with U, UO_2 and Hg_2Cl_2 samples in 1994, see [Ta94], measurements have been done during 1995 with two sets of U and UO_2 samples at 23.7°Kelvin for about 500 h. These measurements are now being analysed. Further measurements on Hg_2Cl_2, uranium compounds and NpO_2 are in process or planned.

Here, the experimental results for U and UO_2 will be presented; and some preliminary results will be discussed, which were obtained from the interpretation of the 6.67eV resonance.

The interpretation has been started with a trial to employ the commonly used Gas Model with effective temperature. While this model does not fit the experimental data, they may be described quite well by a model starting from the quantum mechanical description of the sample lattice vibration and assuming simple approximations of the lattice vibration spectrum.

The Experiment

A schematic diagram of the experiment is shown in Fig. 1. The neutrons are generated by the GELINA target device within short pulses of about 15ns duration; afterwards they are moderated by a 35mm water slab; they cross the sample and are detected after a 26.5m flight path by a Li-glass scintillator fastened between two photomultiplier tubes. The coincident pulses from both tubes are used for the time-of-flight measurement, see the middle pulse processing branch in Fig. 1. The scheme also includes the measurement of the detector pulse height spectrum, of monitor detectors, and of some counters and level settings to check and control the measurement. The data acquisition is accomplished by a PC based system.

A sample changer selects between the different samples, which are placed at about half of the flight path within a cryostat. The measurements were done with two sets of U and UO_2 samples having different area densities: about 20mg and 40mg uranium per cm^2. The samples are periodically interchanged to compensate for long term drifts.

Starting from the measured spectra, transmission spectra were determined including dead time correction, background subtraction and normalisation to the open beam spectrum. Afterwards the resonance dips were unfolded from the time-of-flight resolution, and in the case of the UO_2 samples also from the area density profiles of the uranium nuclei in the samples; and the cross-section curves were calculated.

The preparation and adjustment of the experiment were done by runs at room temperature. During this period (about 200 h of measurements) room temperature spectra were accumulated, without any pretension to analyse them afterwards; but, they can be used for comparisons.

Fig. 2: shows a time-of-flight spectrum registered with an U sample and "black resonance" filters during a 50 h period. The flat bottoms of black resonances provided by thick Bi, Co, Mo, Au, and Rh foils are used to determine the background level. The time channel widths were choosen between 16ns and 256ns according to the time resolution. In Fig. 2 the channel content is normalised to a uniform time channel width of 1 μs.

Fig. 3 to 5 show the transmissions and cross-section curves derived for the 6.67eV, 20.9eV and 36.7eV neutron resonances for the U and the UO_2 samples. The resonances have been determined with an energy grid of 5meV or 10meV respectively. The accuracy of the measured resonances is about 1.5% to about 3%; about half of them is caused by counting statistics, the other half by different experimental details.

A full documentation of the experiment and data processing is given in the IRMM Reports GE/R/ND/02/95, 01/96, and 02/96 [R1, R2, R3]. The report GE/R/ND/02/96 contains the numerical data analysed here.

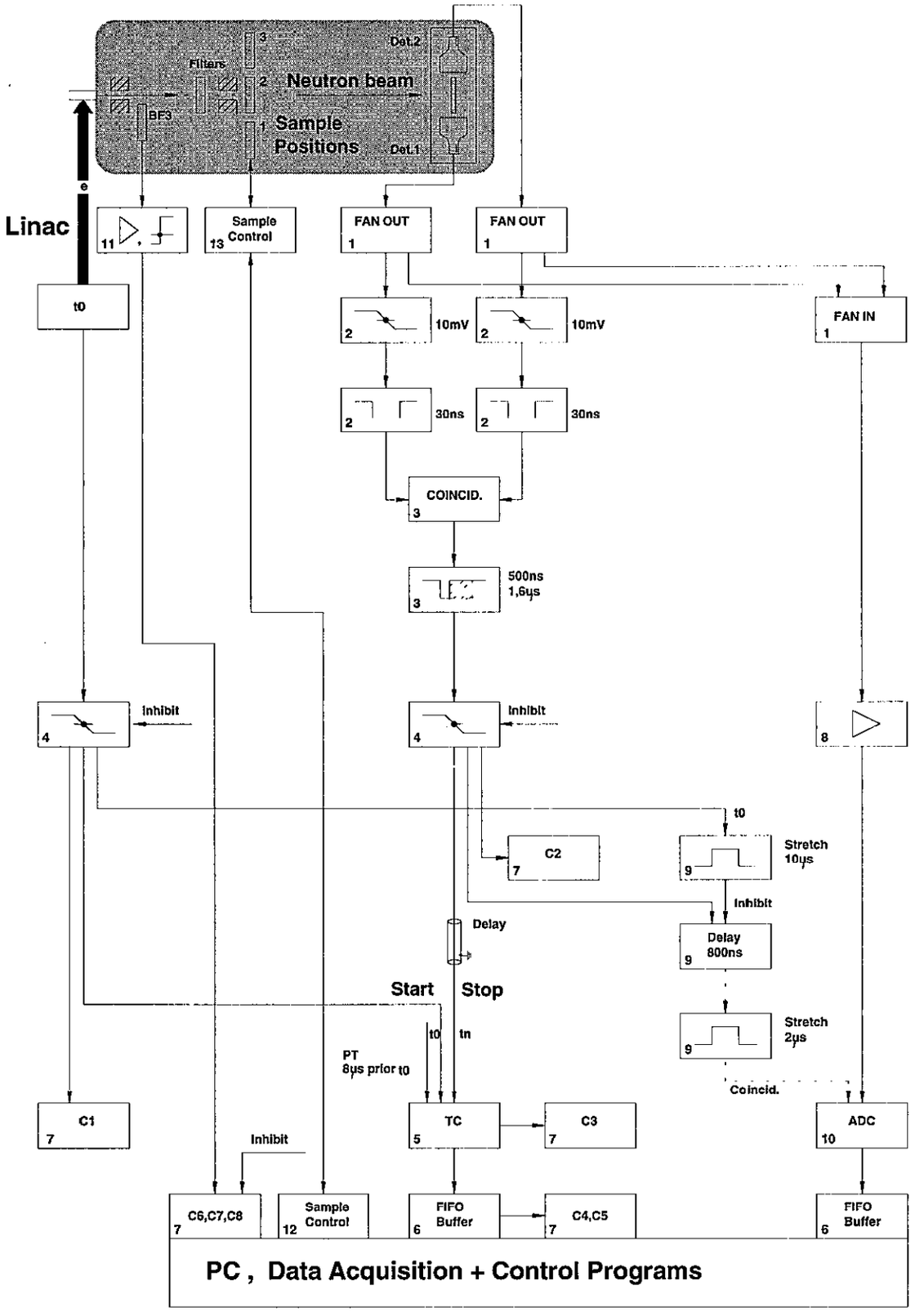

Fig. 1:
Schematic diagram of the experiment.

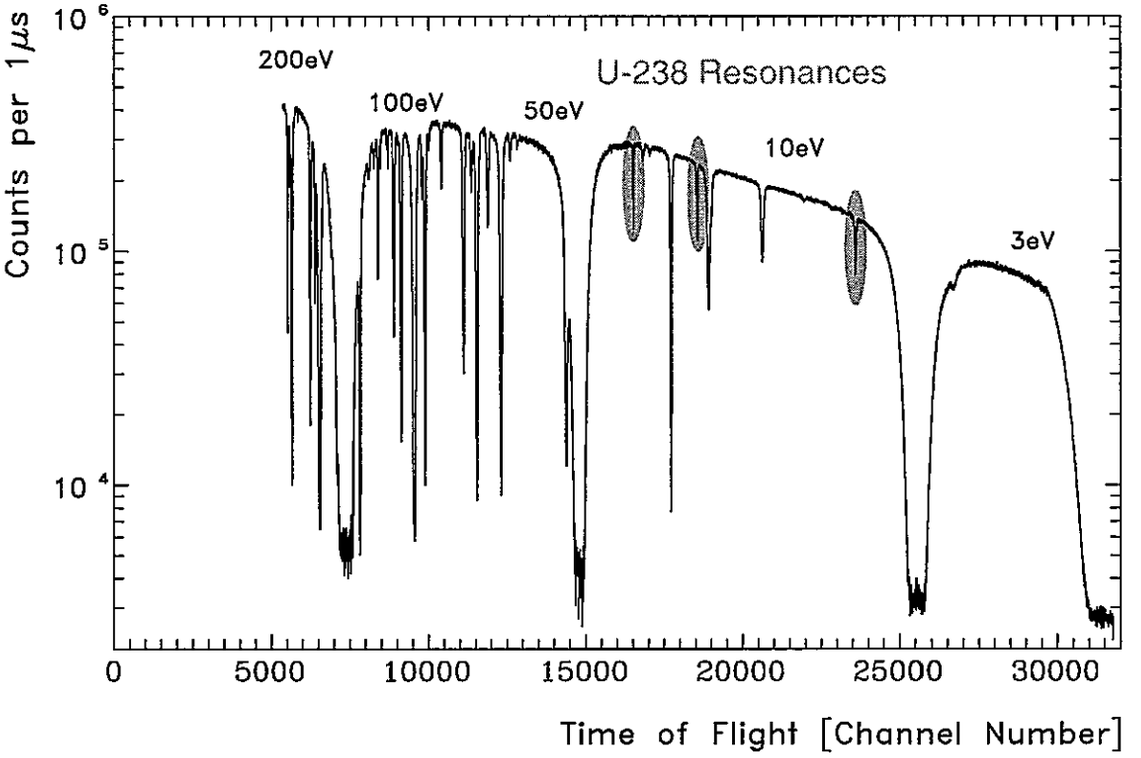

Fig. 2:
Time-of-flight spectrum registered with an U sample and "black resonance" filters.

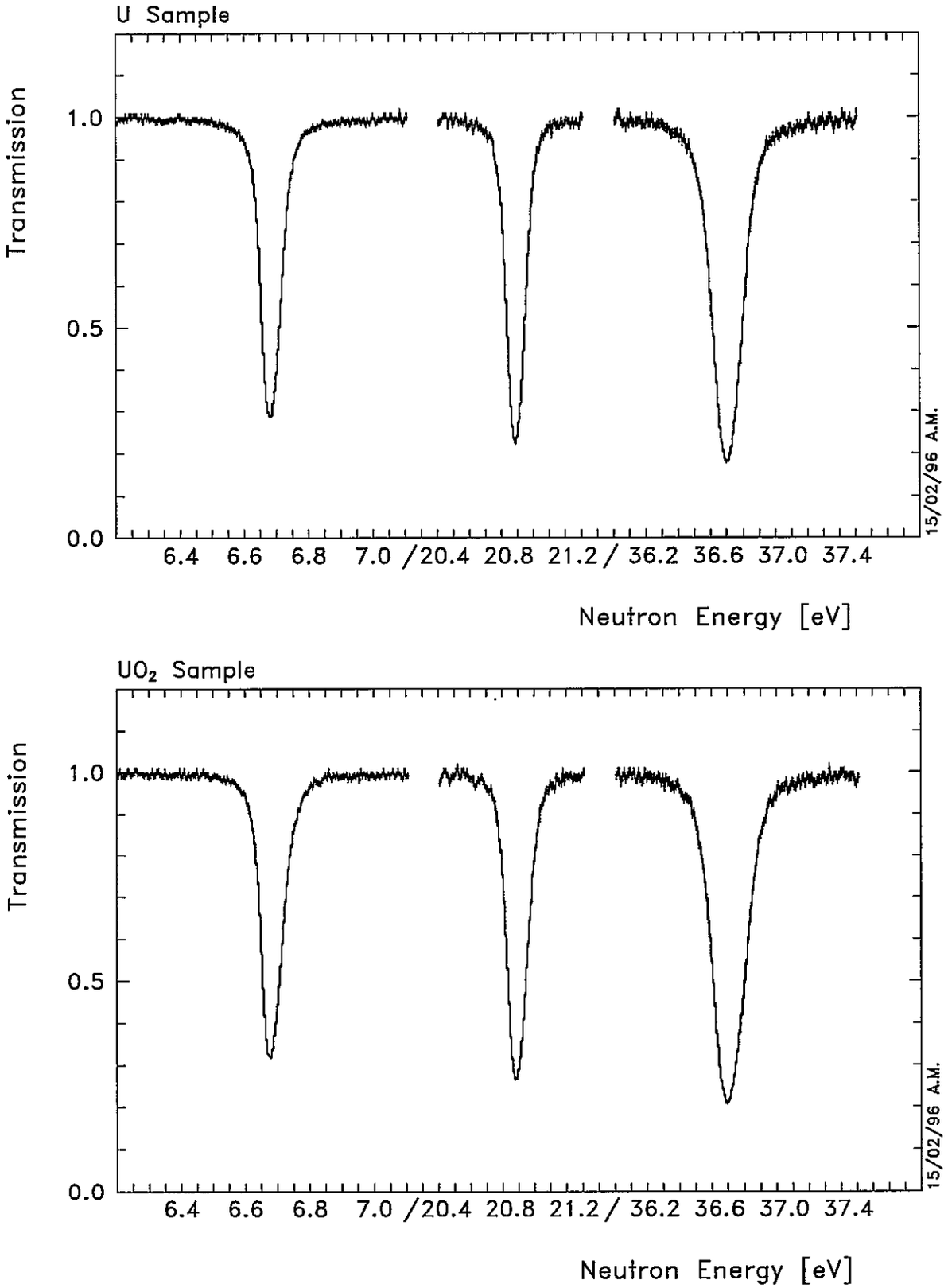

Fig. 3:
Transmission spectra in the vicinities of the 6.67eV, 20.9eV, and 36.7eV resonances for the metallic and the oxide samples, measured at 23.7°Kelvin, sample thickness: 40mg/cm^2 Uranium.

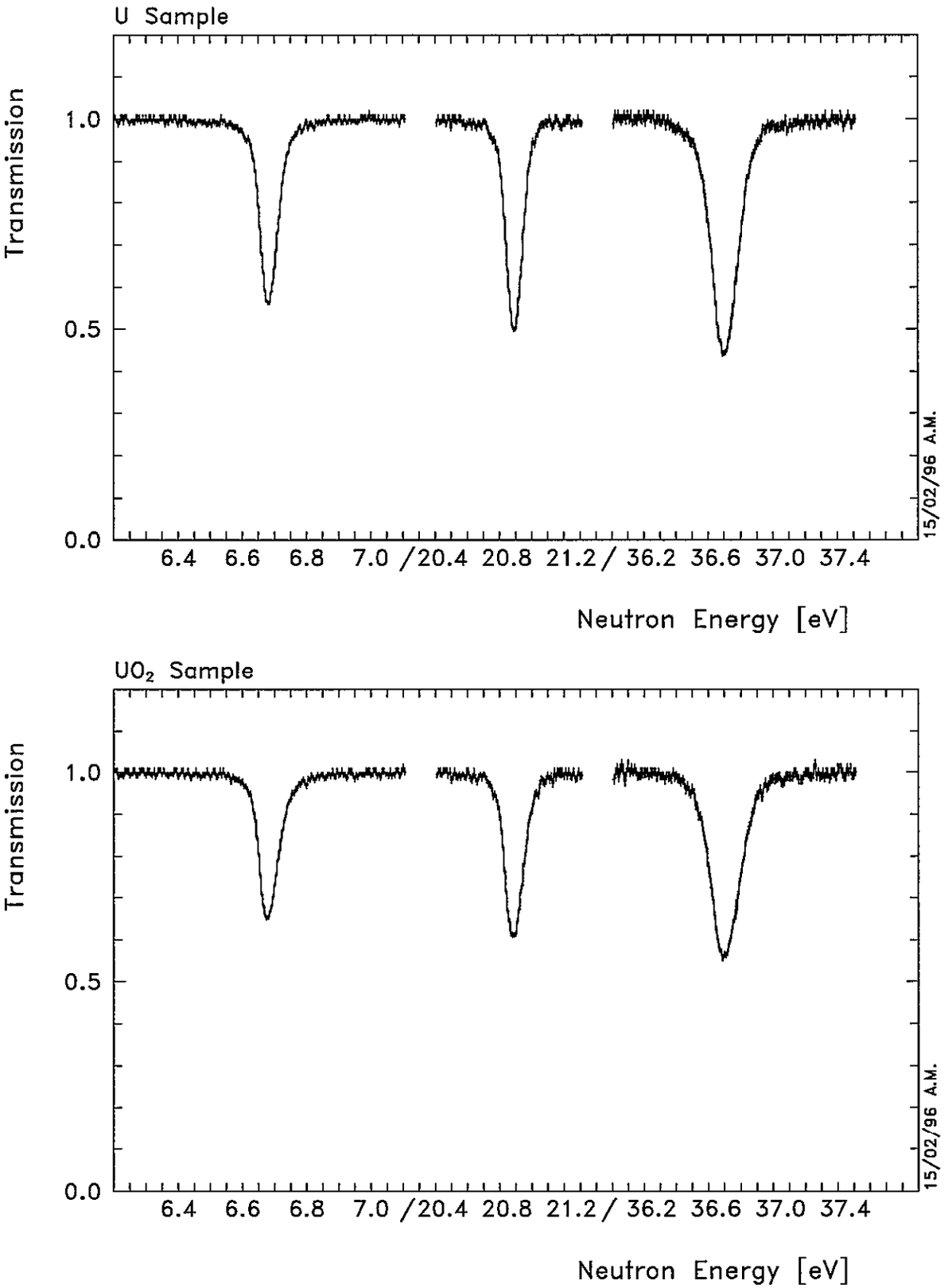

Fig. 4:
Transmission spectra in the vicinities of the 6.67eV, 20.9eV, and 36.7eV resonances for the metallic and the oxide samples, measured at 23.7°Kelvin, sample thickness: 20mg/cm^2 Uranium.

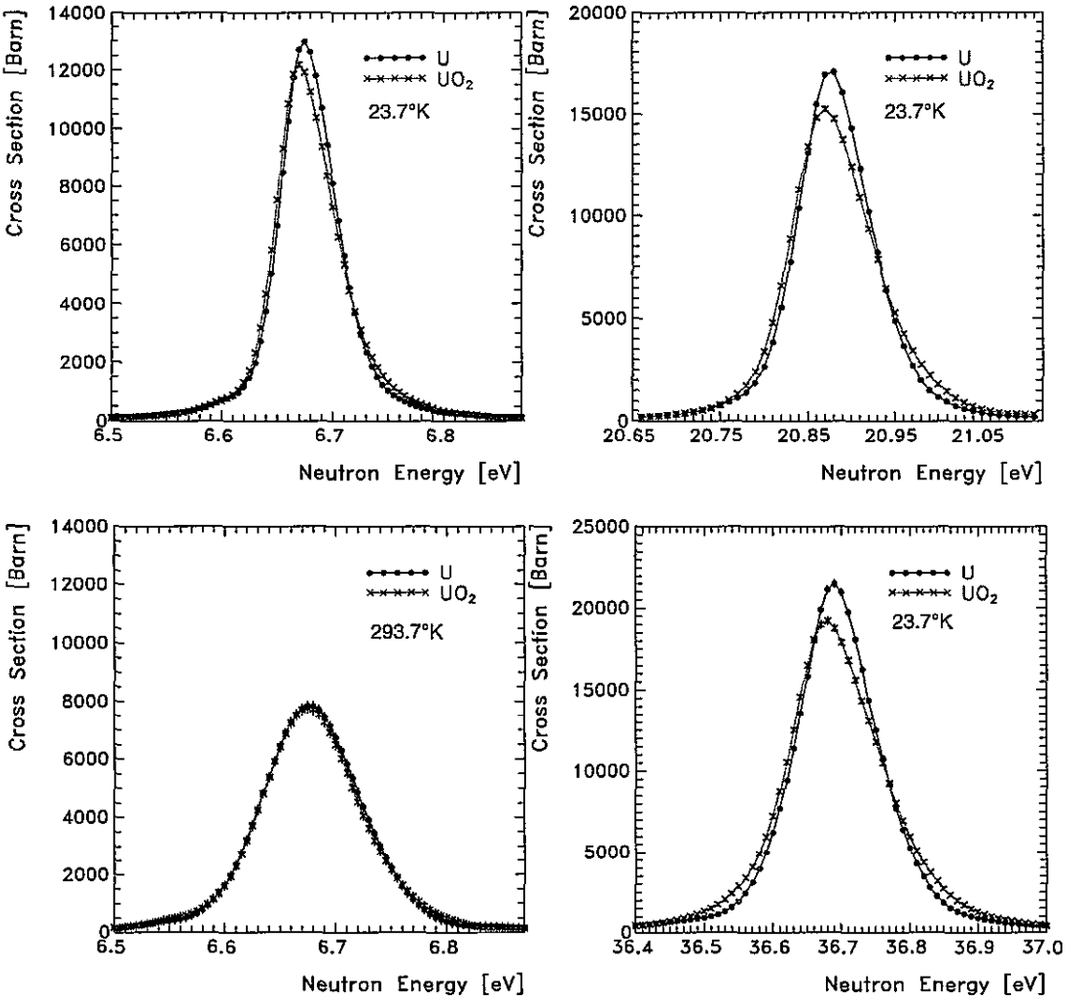

Fig. 5:
Cross-sections for the 6.67eV, 20.9eV, and 36.7eV resonances measured with metallic Uranium and with an UO_2 sample at 23.7°Kelvin, and the 6.67eV resonance measured at room temperature.

Interpretation

The resonances measured at low temperature show a pronounced difference in their shapes between the U and the UO_2 sample. This difference is in the order of 10% of the absolute cross-section value and more, far above the experimental uncertainties. The principal structure of the difference is nearly the same for all three resonances: lowering of the resonance maximum and a significant asymmetrically broadening for the UO_2 sample. The differences are smaller at room temperature.

A trial has been done to describe the neutron resonance shapes by the commonly used Gas Model with effective temperature. The cross-section curve was calculated from the single level Breit Wigner formula and the model of an ideal gas of sample atoms to describe the Doppler broadening; no further simplification was made.

The calculated curves were then fitted to the experimental cross-sections. The best fit curves are shown in the upper part of Fig. 6. The lower part of Fig. 6 shows the residuals between the experimental cross-section and the model calculation with enlarged scale. Large systematical residuals with a significant structure are evident. The structure is the same for U and for UO_2; the magnitude is larger in the case of UO_2. These residuals can not be decreased by parameter variation within the model used. They would be still larger if values according to the mean energies would be used for the effective temperatures instead of the best fit values. The error bars refer to the original statistical errors of the measured data. The fluctuation is less then predicted by these error bars; that is because of variance reduction during the data processing (by smoothing the open beam spectra and by the unfolding procedure).

The parameters determined from the fits are given in Table 1. The resonance parameters obtained agree with the JEF2.2 data within about the one Sigma error of the present data. (All resonance parameters given here refer to the c.m. system; to transform them into lab. system co-ordinates they have to be multiplied by $(M_u+m_n)/M_u$, with M_u and m_n being the atomic mass and the neutron mass, respectively.)

Table 1
Resonance data for the 6.67eV resonance at 23.7°Kelvin using Gas Model.

	U	UO_2	JEF2.2-Data
T_{eff}:	65°K	83°K	
Γ_n:	1.509±0.016 meV		1.493 meV
Γ_γ:	23.51±0.50 meV		23.0 meV

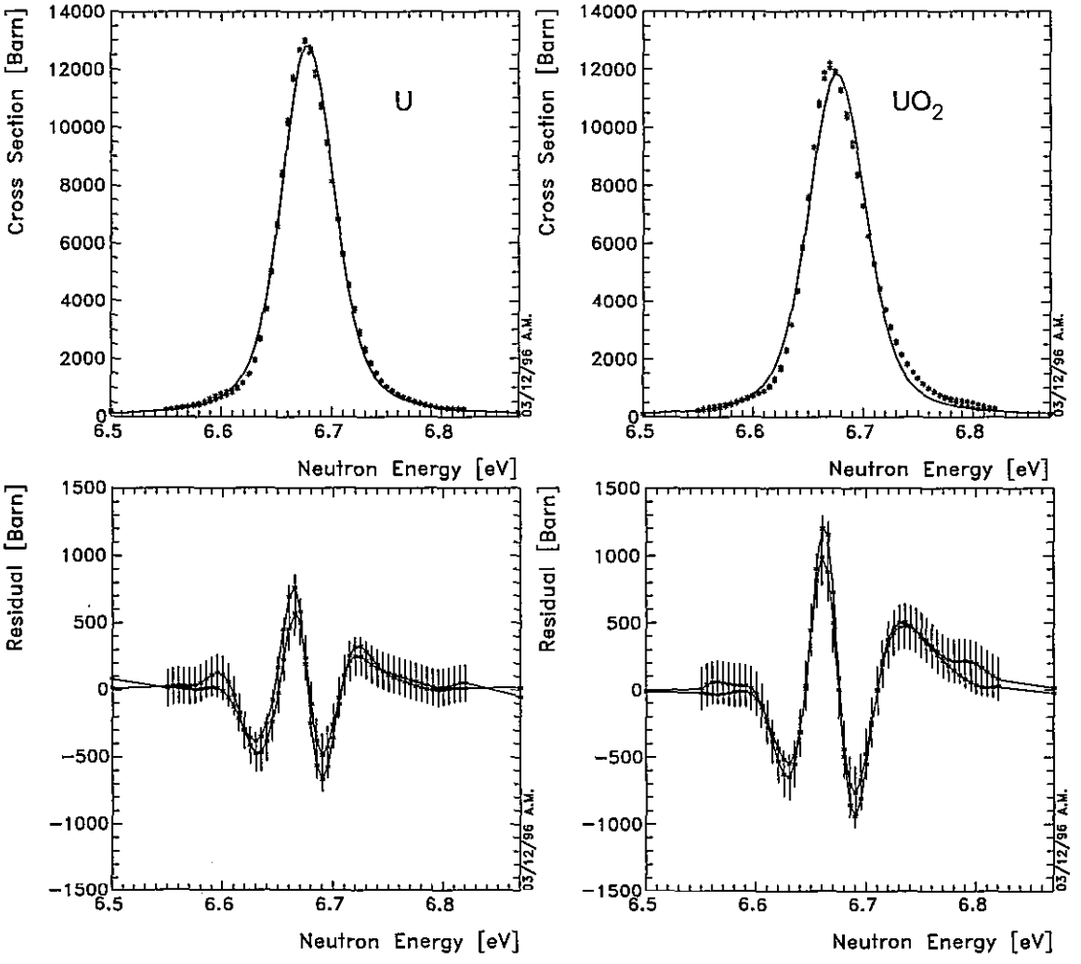

Fig. 6:
The 6.67eV neutron resonance measured with U and UO_2 samples at 23.7°Kelvin, resonance curves calculated using Breit Wigner formula and Gas Model with fitted effective temperatures (upper part), residuals between the experimental cross-section and the model calculation with enlarged scale (lower part).

Whereas the Gas Model failed in describing the experimental resonance shapes, a quite accurate description has been got using a Crystal Lattice Model for Doppler broadening. This model starts from the quantum mechanical description of the harmonical vibration of the sample crystal lattice. The spectrum of lattice vibration frequencies is simplified to one single frequency (Einstein's theory of specific heat) or to a finite number of lattice frequencies:

$$\rho(\hbar\omega) = \sum_{j=1}^{m} a_j \cdot \delta(\hbar\omega - \hbar\omega_j).$$

It is, in principle, the same model as was used by H.E. Jackson and J.E. Lynn to analyse the 6.67eV resonance in U and U_3O_8 [JL62]. Here, some improvements were done: the mass change upon neutron capture is included and the calculation is extended to more tan two lattice frequencies.

The calculation consists in adding cross-section curves shifted in energy. The energy shift is given by the energy loss or energy gain caused by the phonon creation or phonon absorption upon target atom recoil. Fig. 7 illustrates this model for one vibration frequency. This example meets the real situation of a metallic uranium sample at 23.7°K. The 6.67eV resonance is then build up of about 10 components corresponding to the excitation of 0 phonons up to about 9 phonons. The probability to absorb one phonon during the neutron nuclear interaction is very low at this temperature. Of course, the number of components will rise with increasing sample temperature or number of phonon frequencies. The amplitudes of the single components have to be calculated by quantum mechanics. The formulas and the short Fortran code used to calculate them are given in the IRMM Report GE/R/VG78/94 [Me94].

Fits of the lattice model curves to the experimental date from metallic Uranium were carried out assuming two approximations to the phonon spectrum: one single lattice vibration frequency and three frequencies. The latter simulates a Debye spectrum $\rho(\hbar\omega) \propto \omega^2$. The results are shown in Fig. 8. In both cases the residuals are in the order of the magnitude of the statistical errors. They could be further reduced if more (experimental and phonon) parameters would be adjusted. However, this has not been done, because it can not be excluded, that at least a certain part of the oscillating structure in the residuals is due to the unfolding procedure and not to further phonon processes. The parameters resulting from the curve fits are given in Table 2.

The mean energy of vibrational degree of freedom:

$$<\varepsilon> = \frac{1}{2} \int_{\hbar\omega=0}^{\hbar\omega_{max}} d(\hbar\omega) \cdot \hbar\omega \cdot \coth\left(\frac{\hbar\omega}{2kT}\right) \rho(\hbar\omega)$$

was calculated from the fitted phonon frequencies.

In reactor neutronics calculations it is convenient to use a Debye temperature to describe the solid state phonon spectrum. Therefore, a Debye temperature Θ_D which reproduces the same $<\varepsilon>$ value was determined from the equation:

$$<\varepsilon>=\frac{\int_{\hbar\omega=0}^{k\Theta_D} d(\hbar\omega)\cdot\hbar\omega^3\cdot\coth\left(\frac{\hbar\omega}{2kT}\right)}{2\cdot\int_{\hbar\omega=0}^{k\Theta_D} d(\hbar\omega)\cdot\hbar\omega^2}.$$

In Ref. [JL62] the Doppler broadening of the 6.67eV resonance in a metallic uranium sample was described by one lattice frequency $\hbar\omega_1$ = 11±2 meV; this is in agreement with the present data.

It should be mentioned, that the analysis of the 20.9eV and the 36.7eV resonances yields values for the single phonon frequency model which agree very well with the value found from the 6.67eV resonance.

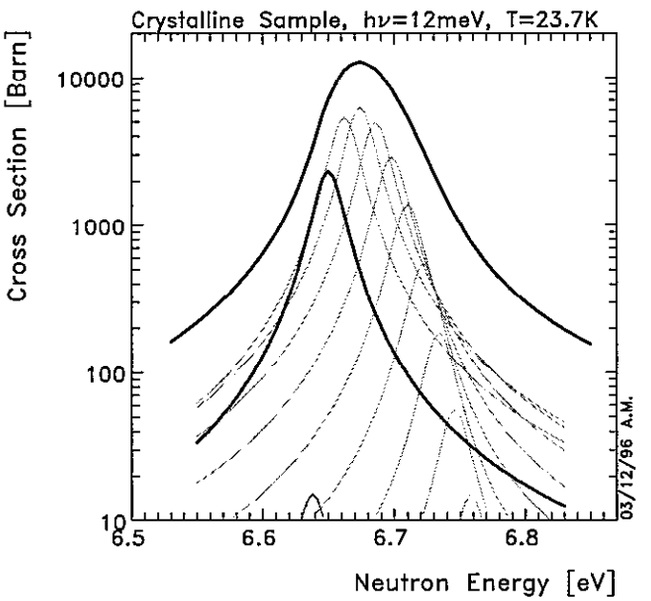

Fig. 7:
Components of the Doppler broadened 6.67eV resonance corresponding to the various phonon excitations in a crystallic sample having one lattice frequency $\hbar\omega_1$=12meV.

The analysis of the Doppler broadening in UO_2 was done supposing three different approximations of the phonon spectrum: one single frequency, two frequencies related by the square root of atomic masses and three frequencies with two of them representing the acoustical mode similar to a Debye spectrum and the third representing the optical mode. The results are shown in the Figs. 9 and 10.

The one frequency approximation leaves large residuals. If higher frequency vibrations (the optical mode vibrations) are accounted for, the residuals between experiment and calculation become less. The remainder is then again in the order of the magnitude of the statistical errors; and the same argumentation holds as in the case of the metallic sample. For comparison, the frequencies are given obtained in Ref. [Mi83] from the analysis of ^{235}U and ^{234}U resonances at 293°K in U and UO_2.

It should be mentioned, that in the case of a chemical compound, as UO_2, only that part of the full phonon spectrum of lattice vibrations can be excited by the recoil of the selected atom in which this sort of atoms is really vibrating. Therefore, the phonon spectrum responsible for Doppler broadening consists only in a part of the full phonon spectrum. The full phonon spectrum can be measured by thermal neutron scattering, see Ref. [Yo69]. If one attributes a "Debye temperature" to this spectrum part, this value will be of course different from the common Debye temperature describing the full phonon spectrum of the compound.

Conclusions

Although the analysis of the measurements is not complete yet, some results are evident. They support earlier results from Ref. [JL62], [Se88] and other authors. That is: using Gas Model description of the shape of low-energy neutron resonances results in large discrepancies to high resolution experiments, also if an effective temperature will be adjusted. Using a Crystal Lattice Model yields good agreement with the experiment, even if the phonon spectrum of lattice vibration is approximated in a very simple manner. The simplicity of this model is favourable for the practical computation of Doppler broadening.

It should be therefore suggested to think over, whether Doppler broadened cross-sections for solid state materials should be calculated by a Crystal Lattice Model instead by the Gas Model with effective temperature.

The present results support the recommedations given in the JEF-DOC 555 [Ro95] for "Debye temperature" values to be used in resonance cross-section calculations for UO_2 and U.

Table 2
Parameters of the 6.67eV resonance in a metallic uranium sample at 23.7°Kelvin using Crystal Lattice Model.

	one frequency	three frequencies	JEF2.2-Data
$\rho(\hbar\omega)$:	$\hbar\omega_1$=11.6±0.2 meV	$\hbar\omega_1$= 2.6±0.04 meV 3% $\hbar\omega_2$= 7.8 meV (3*$\hbar\omega_1$) 25% $\hbar\omega_3$=13.1 meV (5*$\hbar\omega_1$) 72%	
{Ref. [JL62]:	$\hbar\omega_1$=11±2 meV }		
<ε>:	5.84±0.08 meV	5.81±0.09 meV	
<ε>/kT:	2.86	2.85	
Θ_D:	15.3±0.2 meV 178±3°K	15.3±0.2 meV 177±3°K	
Γ_n:	1.513±0.016 meV	1.515±0.016 meV	1.493 meV
Γ_γ:	23.31±0.35 meV	23.49±0.38 meV	23.0 meV

Table 3
Parameters describing the Doppler broadening of the 6.67eV resonance in UO_2 at 23.7°Kelvin using Crystal Lattice Model. The uncertainties of <ε> and Θ_D are estimated from the fitted $\hbar\omega$ values by error propagation.

	one frequency	two frequencies	three frequencies
$\rho(\hbar\omega)$:	$\hbar\omega_1$=15 meV	$\hbar\omega_1$= 13.4±0.2 meV 90.0±0.7% $\hbar\omega_2$= 51.8 meV ($\hbar\omega_1$*√(238/16)) 10%	$\hbar\omega_1$=4.4±0.1 meV 8.5% $\hbar\omega_2$=13.1 meV (3*$\hbar\omega_1$) 76.7% $\hbar\omega_3$=40±2 meV 14.8±1%
<ε>	7.5 meV	8.66±0.2 meV	8.21±0.2 meV
<ε>/kT	3.7	4.24	4.02
Θ_D:	20 meV 230°K	23.0±0.4 meV 267±5°K	21.8±0.5 meV 253±6°K
{Ref. [Mi83]:		$\hbar\omega_1$= 12±3 meV 92±4% $\hbar\omega_2$= 46 ±4 meV ($\hbar\omega_1$*√(238/16)) 8%	
Θ_D:		260°K}	

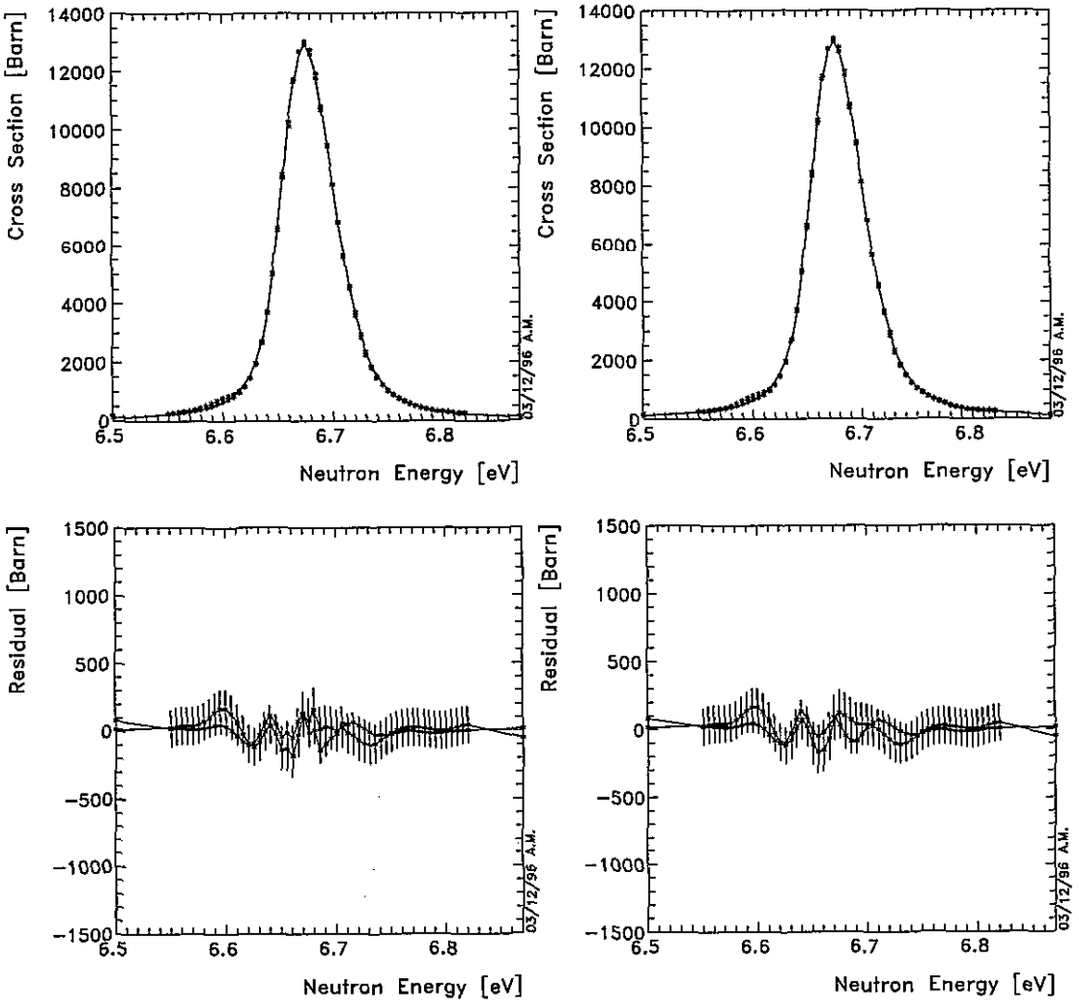

Fig. 8:
The 6.67eV neutron resonance measured with U samples at 23.7°Kelvin, resonance curves calculated using Breit Wigner formula and Crystal Lattice Model with one frequency (left hand side) or three frequencies (right hand side), residuals between the experimental cross-section and the model calculation with enlarged scale (lower part).

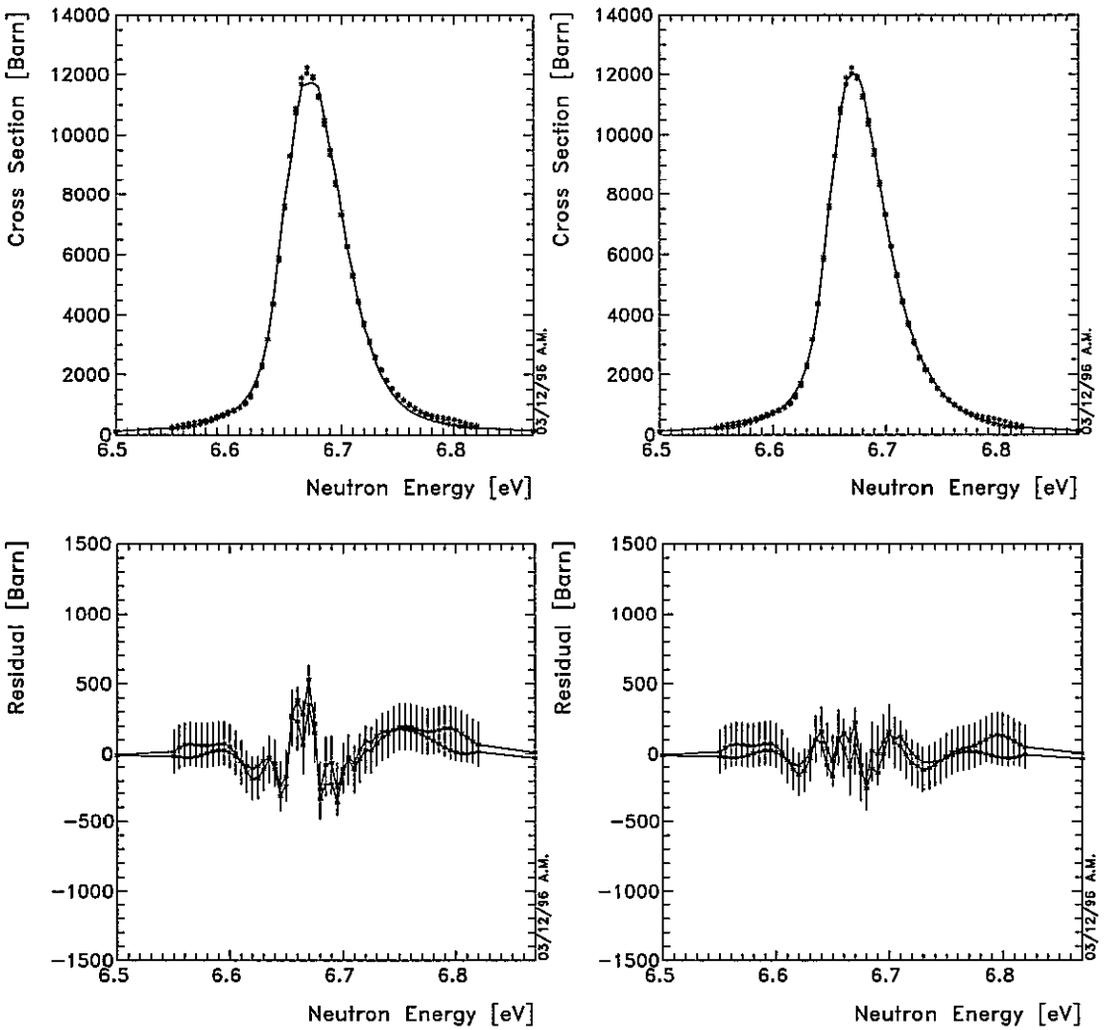

Fig. 9:
The 6.67eV neutron resonance measured with UO_2 samples at 23.7°Kelvin, resonance curves calculated using Breit Wigner formula and Crystal Lattice Model with one frequency (left hand side) or two frequencies (right hand side), residuals between the experimental cross-section and the model calculation with enlarged scale (lower part).

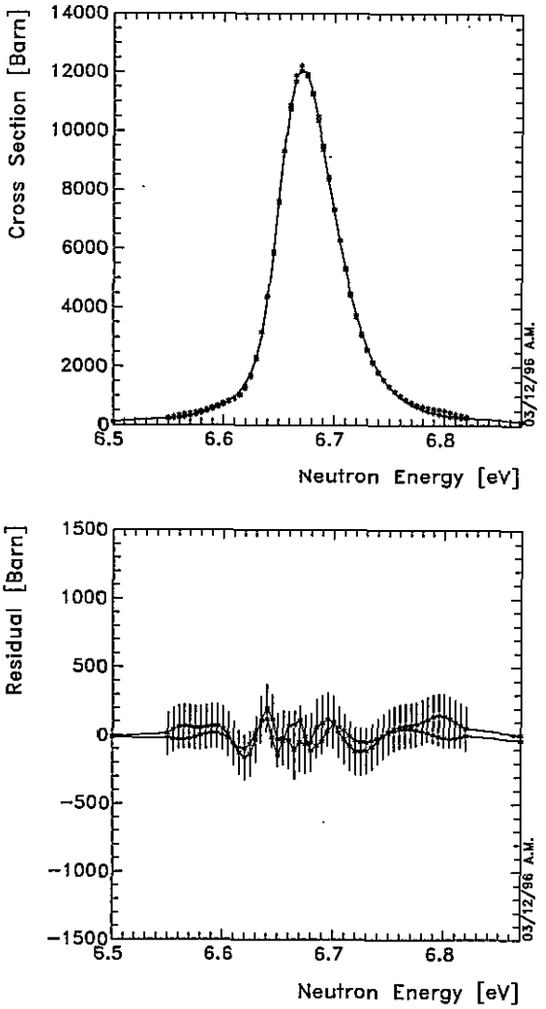

Fig. 10:
The 6.67eV neutron resonance measured with UO_2 samples at 23.7°Kelvin, resonance curves calculated using Breit Wigner formula and Crystal Lattice Model with three frequencies, residuals between the experimental cross-section and the model calculation with enlarged scale (lower part).

References

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