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**Analysis of η measurements for ^{235}U in the thermal neutron
energy region**

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ABSTRACT

Existing experimental data on η of ^{235}U in the thermal and sub-thermal energy regions are reviewed. Special attention is given to the various systematic uncertainties. When combined with recent fission cross section data, an R-matrix fit yields a good representation of the data in the energy region below about 0.2 eV. At higher energies problems remain which need further investigation.

1 Introduction

The detailed energy dependence of neutron cross sections and related parameters of fissile nuclei for sub-thermal neutron energies has recently found considerable attention because of its effect on the temperature coefficient of reactivity of thermal reactors [1]. One of the quantities thus considered is η , the number of fission neutrons emitted per neutron absorbed, of ^{235}U . Several measurements of the energy dependence of this quantity have been performed in recent years. The two earlier ones were done at Harwell [2] and Geel [3], later measurements were done at the ILL, Grenoble [4], and by a Harwell-ORNL collaboration at ORELA [5], and finally a measurement of the related quantity alpha was done at Geel [6]. Preliminary analysis of some of these data seemed to show discrepant results, although the differences were at the limit of the combined systematic uncertainties. Therefore, a subgroup of the NEA Working Group on Evaluation Cooperation was set up to further investigate this (and related) problems. In the present note we want to discuss some of our findings as of June 1993. In section 2 we will briefly discuss the experimental techniques applied and their difficulties. In section 3 we will present our findings for the thermal and sub-thermal energy regions. In section 4 we will point to still existing problems at somewhat higher energies (above ~ 0.3 eV) and finally in section 5 shortly address the question of Doppler broadening.

2 Experimental Techniques and Difficulties.

As a general statement it may be said that systematic corrections which had to be applied to the measured data are important, and so are systematic uncertainties as they are compared to the size of the effect under investigation. However, some of the experiments differ strongly in the applied techniques, and as a consequence the relative importance of the various systematic uncertainties are rather different.

The principle method to measure the energy dependence of η is simple: A beam of low energy neutrons first passes through a flux monitor before it hits a "black" metallic U sample. The transmission of this sample is almost zero for neutron energies below 0.1 eV. Fission neutron emerging from this sample are detected by a NE213 liquid scintillation detector. Pulse shape discrimination is used to distinguish neutrons from γ -rays. The shape of the neutron flux is measured by replacing the black U sample by a neutron capture sample which also is "black" for neutrons in the energy range of interest. Samples of ^{10}B and Cd have been used. Thus the shape of the neutron flux is directly obtained from the yield of γ -rays from the capture sample. The flux monitor is used only to record possible changes of the neutron flux shape between the fission measurements with the black U sample and the flux measurement with the capture sample.

The earlier Harwell and Geel as well as the more recent Harwell-ORNL measurements have been done with a linac pulsed white neutron source and conventional time-of-flight technique. The most difficult problem in these measurements is the determination of backgrounds, especially in the sub-thermal region: Every fission event in the U sample produces neutrons which may be back-scattered from the surroundings and produce a delayed fission event in the sample at a later time. A decaying background with a few ms delay is indeed seen in measurements with a Cd filter. The true background cannot be readily measured because any background filter, e.g. Cd, will take away also most of the source of this background.

In the latest analysis of the Harwell-ORNL data much effort has been devoted to reconstruct the background by folding the delayed component observed in the measurement with the Cd filter into the foreground fission rate. This and an extrapolation to zero filter thickness were best possible to be carried out on the Harwell-ORNL data. Measurements on samples of Pb and C provided an additional guideline for the determination of the background and of the effect of the filters used to determine the background.

In the measurement at the ILL reactor the flux at the thermal neutron guide was sufficient to allow the use of a double chopper set up: Two choppers, separated by about 3 m, essentially produced a pulsed monoenergetic beam. The sample for the η measurement (or the capture sample for the flux measurement) is placed another 0.9 m downstream from the second chopper. Thus the time signal from the detector can be used to separate background events due to out-of-time neutrons from true events. With this method the uncertainty in background determination could be reduced. On the other hand, instantaneous count rates during the neutron pulse are high, and thus are count loss corrections which reach a maximum of about 6% at about 60 meV neutron energy. In the original analysis no correction was applied for a count rate dependent cross talk between γ -ray and neutron output channels of the PSD circuit. Recent inclusion of this effect resulted in a slight reduction of the energy dependence of η (by 0.2% between thermal and 2 meV).

In all the η measurements corrections have to be applied for incomplete absorption and for multiple scattering of the incident neutrons in the U sample. These finite sample size corrections are very small at low neutron energies, but increase sharply as the absorption cross section drops at energies above 0.3 eV. We will have to come back to this point in section 4. In addition, a correction must be applied for absorption and multiplication of the emitted fission neutrons in the U sample: it depends on the place in the sample where the primary fission event took place and thereby on the incident neutron energy. The maximum amount of this correction amounts to about 3.5% for the Geel and Grenoble measurements, whereas it is smaller for the Harwell-ORNL measurements due to the use of a detector which subtended a larger solid angle to the sample.

In contrast to the η measurements, in the measurement of alpha performed at Geel, no count loss corrections appear, and practically no corrections for finite sample size. The method is based on the measurement of the intensity ratio of specific low energy capture γ -rays and prompt fission γ -rays with a Ge-detector. However, it rests on the assumption that the relative yields of the measured γ -rays per capture and fission event, respectively, do not vary as a function of neutron energy. This will only be fulfilled as long as the relative contributions of different resonances to the cross sections do not vary strongly, i.e. only for an energy interval which is smaller than the typical resonance width. The method is thus limited to the sub-thermal and near-thermal energy region. The main experimental uncertainty is due to a limited statistical precision.

3 Present Results.

With the improved determination of the various corrections as described above, the different data are in fairly good agreement for neutron energies below about 0.2 eV. At higher energies some problems remain which will be further discussed in section 4 below. In the "low energy region", i.e. more precisely below 0.3 eV, we have attempted to produce a "best curve" representing the general behaviour of the experimental data. This has been done by a simultaneous R-matrix fit of the fission and capture cross sections of ^{235}U obtained

Energy eV	Radiation width meV	Neutron width meV	Fission 1 width meV	Fission 2 width meV	Spin
-0.4065	35.6	133.9×10^{-03}	-1.281	-191.4	3
-0.017	30.07	5.45×10^{-06}	-3.31	0.68	4
0.2848	42.85	4.749×10^{-03}	107.4	-4.875	3
1.1418	62.92	17.03×10^{-03}	0.107	104.0	4

Table1: The resonance parameters for ^{235}U obtained from R-matrix fit.

from the following experiments:

- the fission cross sections data of Wagemans et al. [7],
- the η data from the experiments at Geel, Grenoble and the Harwell-ORNL data,
- the alpha data from Geel.

The fit was done using the Reich-Moore R-matrix routine MULTI [8]. We started from the resonance parameters as given by de Saussure et al.[9], however omitting their states at -3.49 and -1.50 eV as they were found to have very little effect on the cross sections. We then iterated on the parameters of the resonances between -1.0 and 1.5 eV. The best overall fit to the data was obtained for the parameter set given in table 1.

η -values calculated from the R-matrix fit are compared to some of the experimental data in Fig.1a. The left part of Fig.1a shows the calculated η (full curve) together with the experimental data from the Grenoble measurement (circles) and with η as calculated from the measured alpha values of the Geel alpha experiment (crosses). The broken curve represents η as calculated by NJOY from the resonance parameters of de Saussure et al. The right part of Fig.1a shows the same curves together with the experimental data from the Harwell-ORNL measurement. The error bars indicated in Fig.1a represent statistical errors only. It is to be stressed that the fit curve of Fig.1a will not be the final "best curve" because there are still problems at somewhat higher energies (see below). Further investigation of these problems may lead to a modification of the parameters of the resonances e.g. at 0.28 and 1.14 eV; such modification will also effect the low energy region, although only slightly. Therefore, the present fit curve of Fig.1a is only a first approximation.

It is seen that the R-matrix fit of Fig.1a is a good representation of both sets of data, especially at sub-thermal energies. This holds also for the data from the earlier Geel η experiment (not shown), except for the lowest energies ($E < 3\text{meV}$) where background problems were severe. There is a slight systematic difference between the fit and the data in the 70 to 100 meV region, in opposite sense for the Grenoble and the Harwell-ORNL data. However, this is within systematic uncertainties. The decrease of η from thermal energy to 2 meV as represented by the fit curve is 1.6%.

Fig.1b shows the difference between the R-matrix fit and the experimental data of Fig.1a. On the same scale, Figs.1c and 1d show the most important systematic uncertainties of the experimental data originating from the various sources discussed above. The numbers indicated on the figures have the following meaning:

- 1) uncertainty due to background errors in the measurement of fission neutrons from the U sample.
- 2) uncertainty due to background errors in flux measurement.
- 3) uncertainty due to possible changes in the shape of the incident neutron spectrum.
- 4) uncertainty in the determination of count loss corrections.
- 5) uncertainty in the correction for absorption and multiplication of fission neutrons.

6) change in the finite sample size correction due to the addition of 1 b to the ^{235}U total cross section.

7) change in the finite sample size correction due to the addition of 1 b to the ^{235}U scattering cross section.

It is seen from Fig.1 that differences between the present fit curve and the experimental data are generally smaller than the combined systematic uncertainties, with the possible exception of the lowest data point from the Grenoble experiment. Furthermore, it is seen that the most important systematic uncertainties in the left (Grenoble) and right (Harwell-ORNL) columns of the figure originate from different sources: count loss corrections for the Grenoble case, backgrounds for the Harwell-ORNL one. The fact that nevertheless there is fair agreement now between these two data sets adds additional confidence in the final result.

4 Problems at higher energies.

In the neutron energy region above 300 meV the corrections for scattering and the finite sample size become larger and dominate the uncertainty, due mainly to the lower absorption cross-section as the scattering cross-section only decreases from $\sim 15\text{b}$ down to $\sim 12\text{b}$. At these higher energies practically only the Harwell-ORNL measurements provide useful information. The earlier Geel measurement used a thinner sample, thus making the results even more sensitive to the uncertainties in the finite sample correction; At Grenoble there are no neutrons in this energy region and the Geel alpha measurements are uncertain over large energy ranges by the measurement principle as explained above.

In case of the ORELA measurement, the zero temperature cross-sections used in the calculations of the corrections were generated from the ENDFB/6 resonance parameters [12] using the program NJOY. These cross-sections were used in a modified version of REFIT to calculate the fission and capture yield taking into account the finite sample size, effective temperature and the effect of neutrons initially scattered but absorbed on subsequent collisions. From these calculations the corrections needed to obtain η from the fission neutron yield were determined. The correction for the measurements eg. at 0.8 eV varied from ~ 1.6 for the thickest sample to ~ 6.5 for the thinnest one.

The disagreement in the values of η obtained from different sample thicknesses in the energy region from ~ 0.3 to ~ 1.0 indicate that the ENDFB/6 total cross-section, in the neutron energy region from $\sim 200\text{ meV}$ to $\sim 1\text{ eV}$, may be too large by a few barns. Thus in the neutron energy region above $\sim 200\text{ meV}$ an accurate analysis of the neutron energy dependence of η must await a reassessment of the other cross-sections in that region.

5 Doppler effects

Some of the discrepancy between the integral and differential data may be due to errors in the assumptions made in the programs used in the analysis of differential data and the programs used to generate the cross-sections from the nuclear parameters. The assumption in the modeling of the Doppler effect especially at low neutron energies may be one of the contributing factors to these discrepancies.

The nuclear parameters are required because we cannot measure the neutron cross-sections under all conditions required by the user of neutron cross-section data. Shape

analysis programs are used to determine the nuclear parameters from the measured differential data, correcting for Doppler and resolution effects. The resultant nuclear parameters are then used together with a model, describing the thermal motion of the atoms, to extrapolate the cross-sections to the required conditions. Safety and economic consideration require that the parameters AND the model are correct!

A check was recently carried out at ORELA to see that the total cross-section generated from resonance parameters by two shape analysis programs (REFIT and SAMMY) agreed with those generated by the reactor code NJOY [13]. The calculated total cross-section at a temperature of zero, agreed to better than 1 part in 10^4 . All three programs use the simple gas model with an effective temperature to carry out the Doppler broadening over a neutron energy range from zero upwards. The calculated Doppler broadened total cross-section agreed to better than 1 in part 10^3 . The small differences were thought to be due to differences in the integration functions used in the programs and the energy where the programs change from the simple form of the Doppler function used at high energies to the full equation for use when the neutron velocity is comparable with the nuclear velocity. The Doppler model used in these programs is based on the free gas model with a modified temperature (see Lamb [10]), in which the nuclei in a solid behave like a free gas at an effective temperature. The effective temperature being a function of the true temperature and the Debye temperature.

The calculated cross-sections from all the programs do NOT include any solid state effects i.e. coherent and incoherent scattering etc.

The measurements by Haste et al [11] on the temperature dependence of the total cross-section of uranium oxide indicates that the gas model gives an adequate description of the Doppler effect as a function of temperature (~ 300 K to ~ 2000 K) for the resolved resonances region covered by the experiment (~ 20 eV to ~ 200 eV).

In contrast to the above there are now several measurements that point to the fact that the simple free gas model does not adequately describe the Doppler effects in the neutron energy region below a few eV. Some examples are given below.

At Dubna [14] [15] [16] neutron cross-section measurements and calculations carried out on various compounds of uranium, metallic uranium and including gaseous compounds showed that not only the simple gas model of Doppler broadening did not hold for the 6.7 eV resonance but also that the cross-section depends on the chemical compound.

In the analysis [17] used for the evaluation of the ^{238}U resonance parameters the effective temperature was determined to be 305.8 ± 0.5 K for measurements carried out at normal temperatures on metallic samples. This is slightly higher than the value of 298.2 K calculated using the simple gas model and a Debye temperature of 180 K. The fit to the neutron energy region around the 6.7 eV resonance gave a value of 2.1 for chi-square per degree of freedom, while an average value of 1.02 was obtained for the resonances up to 350 eV. This again suggests that the simple gas model for Doppler broadening is not adequate in the low energy region.

During the measurements carried out at ORELA to determine the neutron energy dependence of η for ^{235}U , two thin transmission samples (~ 0.1 and ~ 0.3 mm) of cadmium were used in the determination of the background. The subsequent shape analysis with the program REFIT of the measured transmission gave resonance parameters (see table 2) that did not agree with the evaluated values. The value of chi-squared per degree of freedom was also high ~ 6 . An analysis by N. Larson [18] using SAMMY produced almost identical parameters as REFIT and a similar high value of chi-square. This again indicates that the simple gas model for Doppler broadening breaks down when the neutron velocity

	BNL325		ORELA'92	
Energy (meV)	178.0	± 2.0	174.395	± 0.057
Neutron width (meV)	0.653	± 0.033	0.6120	± 0.0007
Radiation width (meV)	113.0	± 1.0	109.62	± 0.11
Spin	1		1	

Table2: The parameters for the resonance in ^{113}Cd at 174 meV

is comparable with that of the nucleus in the solid.

6 Conclusion.

We believe that it is fair to say that below about 200 meV there is good agreement now between the different experiments measuring the neutron energy dependence of η for ^{235}U , and that the R-matrix fit shown in Fig.1, although still subject to minor changes, is a good representation of these data. However, the higher energy region needs some further investigation.

REFERENCES

- [1] A. Santamarina and H. Tellier, Proc. Intern. Conf. on Nuclear Data for Science and Technology, Mito 1988, p.47; JAERI, Tokyo 1988.
- [2] M.C. Moxon, J.B. Brisland and D.S. Bond, Proc. Intern. Conf. on Nuclear Data for Science and Technology, Mito 1988, p.75; JAERI, Tokyo 1988; M.C. Moxon, private communication.
- [3] J.A. Wartena, H. Weigmann and C. Burkholz, report IAEA Tecdoc 491 (1987) p.123.
- [4] H. Weigmann, P. Geltenbort, B. Keck, K. Schreckenbach and J.A. Wartena, Proc. Intern. Conf. on the Physics of Reactors, Marseille 1990, Vol.3, Pl, p.33.
- [5] M.C. Moxon, J.A. Harvey and N. W. Hill, The measurement of η for ^{235}U in the neutron energy region below 300 meV. (TBP)
- [6] H. Weigmann, J.A. Wartena and C. Burkholz, Proc. Intern. Conf. on Nuclear Data for Science and Technology, Julich 1991, p.38; Springer Verlag, Berlin Heidelberg 1992.
- [7] C. Wagemans, P Schillebeecks, A.J. Deruytter and R. Barthelemy, Proc. Intern. Conf. on the Physics of Reactors, Marseille 1990, Vol.1, III, p.9.
- [8] G.F. Auchampaugh, Los Alamos National Laboratory report LA-5473-MS, 1974.
- [9] G. de Saussure, L.C. Leal and R.B. Perez, Proc. Intern. Conf. on the Physics of Reactors, Marseille 1990, Vol.1, III.
- [10] W. E. Lamb Phys. Rev. 55(1939)190
- [11] T. J. Haste and M. G. Sowerby, AERE-R 8961 A study of the temperature dependence of the neutron transmission of uranium oxide (1978)
- [12] L. C. Leal Resonance analysis and evaluation of the ^{235}U induced cross-sections ORNL/TM-11547
- [13] R. Q. Wright Comparison of the cross-sections calculated from resonance parameters by the programs NJOY and SAMMY Private communication
- [14] A. Meister, D. Pabst, L. B. Pikelner and K. Seidel Nuc. Phys. A362(1981)18
- [15] S. Mittag, D. Pabst, L. B. Pikelner, W. Pilz, R. Tschammer, A. Neister, D. Seeliger and K. Seidel Nuc. Phys A435(1985)97
- [16] K. Seidel, D. Seeliger, A. Meister, S. Mittag and W. Pilz The influence of atomic, molecular and solid state effects on the neutron resonance cross-section. INDC-056/L
- [17] M. C. Moxon and M. G. Sowerby Evaluation of the resolved resonance parameters for ^{238}U . (TBP)
- [18] N.Larson Private communication (1992)

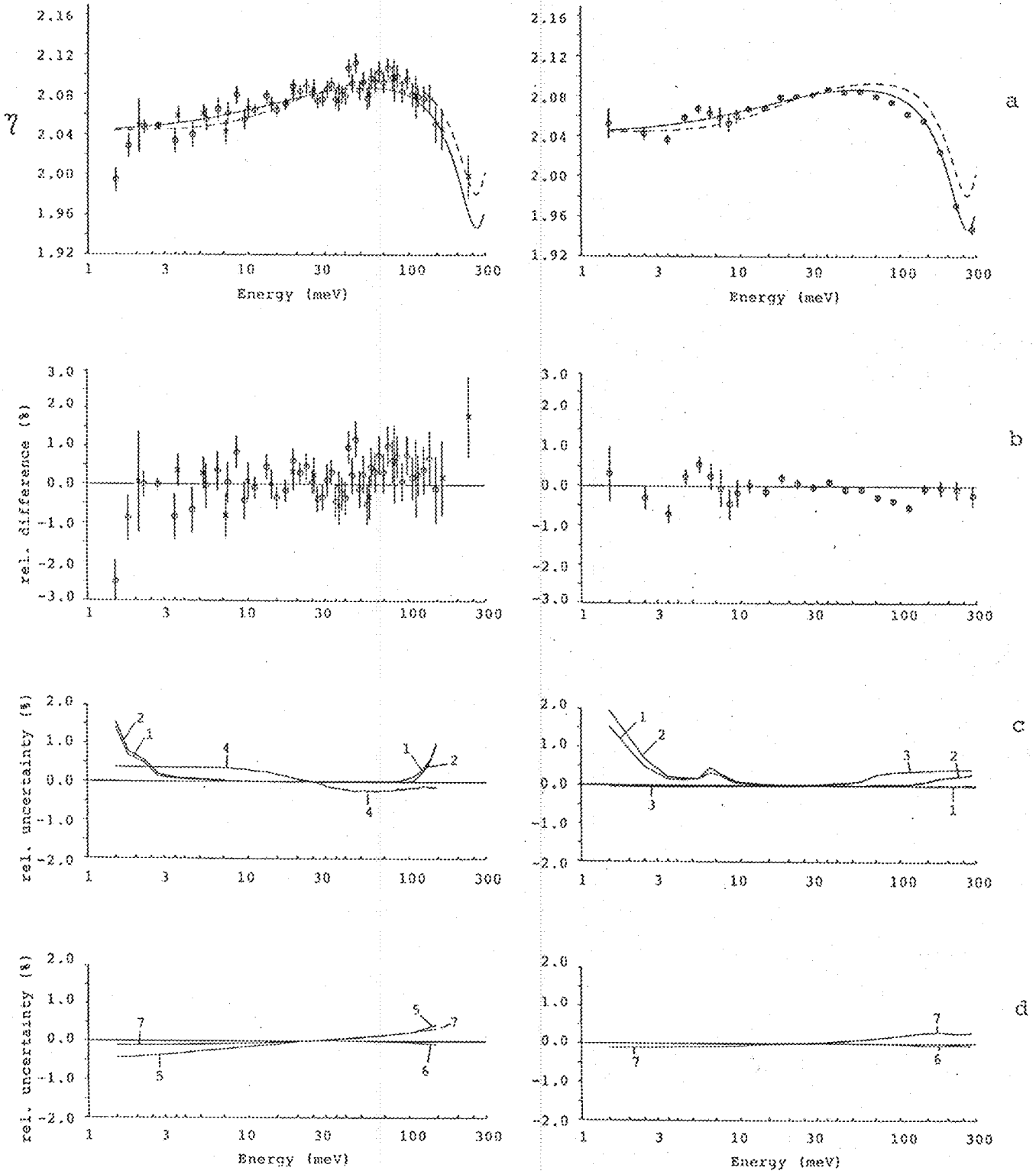


Figure 1: a Experimental η data and R-matrix fit
 b Difference between fitted curve and data
 c, d Various systematic uncertainties
 left column refers to data from Grenoble (and Geel- α) experiment
 right column to Harwell-ORNL experiment
 for details see main text