

## A "Doppler project" sub-group ?

**History** - In the frame of the European Community a "Doppler Project" was launched in 1995 on the request of the French CEA (requests supported by H. Tellier and M. Salvatores).

It involved physicists of 4 European countries : France, Germany, Italy<sup>1</sup>, Netherlands, and of two international laboratories : the ILL at Grenoble and the IRMM at Geel.

The motivation was twofold : studies of resonances at low temperatures in order to clarify the problem of uncertainties on the so-called "Debye effective temperature" (and, implicitly, the validity of the model), and the behavior of nuclear fuel near their fusion temperature<sup>2</sup>.

Furthermore the participation of ILL introduces a look towards a more fundamental approach, and related studies (Hg<sub>2</sub>Cl<sub>2</sub>).

The experimental tools are mainly GELINA (the IRMM experimental time-of-flight facility at Geel) for measurements, a cryostat provided by the Delft University of Technology, and a high temperature furnace, constructed by the ILL at Grenoble (assumed to work up to 3 000 K), which is still under tests.

Various theoretical studies were also engaged.

A 150000 ECU's budget was provided by the European Commission to this project, mainly for the furnace ( 106000 ECU's).

**Present results** - Several experiments have been done for UO<sub>2</sub>, UO<sub>3</sub>, Hg<sub>2</sub>Cl<sub>2</sub>, Ta, .... Some data have been analyzed, and results published. Many data-sets are waiting for their analysis.

We remind that similar low temperature experiments have been done in the past. In order to improve the quality of the results we are devoting a great care to the definition of the experimental resolution and of the background. Furthermore we prepare tools to analyze the reduced data by two methods :

- unfolding (what assumes no theoretical model but introduces oscillations),
- description of the broadening function by a quadrature table (which will have to be interpreted by a theoretical model)

A third approach (introduction of an Einstein or Nernst-Lindemann model in a least-square shape analysis program) has been considered, but not undertaken.

Then we recognize that the data analysis is lengthy and is our actual obstacle together with the availability of the furnace<sup>3</sup>.

Several data have been analyzed with the "effective temperature model" and showed that this model is not valid at low energies (effective temperature lower than the room temperature<sup>4</sup>). This conclusion is particularly verified for NpO<sub>2</sub> : several experiments, ranging from 1995 to 1997, analyzed by different physicists, provided low effective temperatures below 30 eV, increasing with energy. A theoretical explanation of this fact has been established for UO<sub>2</sub> : the gaz model, with an "effective temperature", is not valid at low energy, what introduces systematic errors on resonance parameters (Na98).

<sup>1</sup>The Italian participant, M. Coceva, retired and has not been officially replaced.

<sup>2</sup>In order to elucidate the question of anharmonic behavior of vibrations at high temperatures (Di88, Av91, Ro92)

<sup>3</sup>Another difficulty, the sample holder, is probably solved and has to be tested.

<sup>4</sup>The meaning of "effective temperature" has to be clearly defined : is it the temperature which allows the best description of the cross section shape, or the temperature defined by LAMB, which is always greater than the room temperature ?

Another definition is proposed at Cadarache, more linked to reactor physics.

## Our plans for the near future -

### Experiments :

- termination of the tests of the furnace, check of the behavior of UO<sub>2</sub> in the sample holder, and of this sample holder with the graphite of the furnace, and the transport of the furnace to Geel and its installation on the flight path; then ...;
- quick experiments with Hg at room temperature<sup>5</sup>;
- low temperature studies of UO<sub>2</sub> single-crystals<sup>6</sup>;

### Analysis :

- formalisation of the data reduction procedure (knowledge of the resolution function<sup>7</sup>, ...),
- analysis of the present data by different procedures,
- confrontation of these analyses, and theoretical interpretation.

It is clear that the last two points can be done in different places.

### Applications :

-study of the influence of a better knowledge of the Doppler broadening description and of the resonance's parameters on the behavior of nuclear reactors; this study has to be done (and is being done) by reactor physicists.

### Long term actions :

- improvement of resonance parameters<sup>8</sup>
- modification of processing codes for a better description of the Doppler broadening.

### Our request

The help of the European Commission has been very useful to start this action, providing funds for the most important expenses. But it seems now that the various laboratories involved can support their own expenses<sup>9</sup> for the continuation of these studies.

Nevertheless we found a necessity to continue the actual international cooperation, and even to broad the participation to other experts working, or able to work, in that field.

It is why we expect that a patronage by the NEA would lead to a wider recognition of this action and therefor be profitable for its efficiency.

### References

- Av91 : R.L.d'Avila and R.A. Karam, Ann. nucl. Energy, "Anharmonic effects on the resonance line shape of <sup>238</sup>U in UO<sub>2</sub>", 18-8, (1991)
- Di88 : D.E. Djafri, Ann. nucl. Energy, 15-2 (1988), "Effects of anharmonicity on the Doppler broadening of neutron capture resonances"
- Me95 to Me97 : Andreas Meister et al., several communications from NEUTRON DATA, GE/R/ND/02/95 to "Nuclear data for science and technology", May 1997 (Trieste);
- Na98 : Dimitri Naberejnev and Claude Mounier, to be published.
- Ro92 : John L. Rowlands, Ann. nucl. Energy, 19-7 (1992), "Solid state effects on Doppler broadening of <sup>238</sup>U resonances in UO<sub>2</sub>"

<sup>5</sup>To solve discrepancies observed with the Hg<sub>2</sub>Cl<sub>2</sub> samples;

<sup>6</sup>Not undertaken up to now, because we tried, without success, to thin them;

<sup>7</sup>This resolution function has been calculated by C. Coceva; its results are confirmed by experiments, but does not show "tails", presently unexplained; furthermore it appears some variations of this resolution, unexplained, may linked to be the accelerator set-up.

<sup>8</sup>C.Mounier remarks that the determination of correct resonance parameters at low energy for <sup>238</sup>U is possible from presently available data and has to be treated as a priority; for the main cause of error comes from the error on the resonance parameters, improperly analyzed.

<sup>9</sup>The IRMM, who supports the actual most important expenses (for experiments), did not got any money from the EEC contract.

## FINAL REPORT

"Study of Solid State properties by Neutron Resonances"  
Contract No. CHRXCT940497  
January 1996-January 1998

### Introduction

In the frame of an EC HCM network, an experiment to investigate the exact shape of Doppler-broadened neutron resonances has been set up at a neutron time-of-flight spectrometer of **Institute for Reference Materials and Measurements** (Geel, Belgium).

The principal goal of this study is to perform neutron transmission measurements at low ( $\approx 20$  K) and high temperatures (up to 3000 K) of various isotopes of which  $^{238}\text{U}$  as metal and as oxides is the most important one. During the first year (1995) of the contract, measurements at low temperature were planned and have been done on  $^{198}\text{Hg}$  in single crystal slabs of  $\text{Hg}_2\text{Cl}_2$ , on  $^{238}\text{U}$  as metal and as polycrystalline  $\text{UO}_2$ . For the measurements at low temperature, **Delft University of technology** (the Netherlands) provided cryogenic equipments and expertise in neutron spectroscopy. The **ENEA Bologna** (Italy) provided a neutron detector and performed a Monte Carlo calculation of the resolution function of the Geel neutron time-of-flight spectrometer. The  $\text{Hg}_2\text{Cl}_2$  sample, obtained from the **Institute Laue Langevin (ILL)** (France), exhibits a prototype displacive phase transition at 185 K around which the phonon dynamics are profoundly modified via the existence of a soft mode related to the transition mechanism. We used this property to test the sensitivity of the Doppler broadening experiments at the GELINA facility of IRMM. We expect a precise indication as to which lower limit phonon energy gives rise to measurable Doppler broadening. The first experiments with U-metal and  $\text{UO}_2$  samples were carried out at 24 K and at 293 K. The **Kiel University** (Germany) performed analysis of  $\text{UO}_2$  sample uniformity and provided expertise in solid state effect. The **CEA laboratories** at Saclay and Cadarache (France) developed models to take into account chemical bonds on Doppler broadening of  $^{238}\text{U}$  in  $\text{UO}_2$  and analysed effect of these models on reactor calculations. The REFIT of Moxon has been the basis of the resonance-shape analysis.

The main part of the EC funding (2/3) has been reserved for building a dedicated furnace for in-beam experiments with samples heated up to about 3000 K. The responsibility for the construction of this complicated furnace and its testing was taken up at **ILL**. This major project including operating tests at **ILL** was finished by the end of 1997; the furnace is available for experiments at IRMM.

In the mean time the neutron-beam facility at IRMM was used for further low temperature and room temperature measurements; namely of  $^{237}\text{Np}$  in  $\text{NpO}_2$ ,  $^{238}\text{U}$  in  $\text{UO}_2$  and again  $^{198}\text{Hg}$  in  $\text{Hg}_2\text{Cl}_2$  and for improving the measurements and analysis procedures substantially.

### Results

The main results, obtained up to now, can be found in the proceeding of the Trieste conference held in May 1997. This paper synthesise the work and the results of the low and room temperature measurements for uranium metal and uranium dioxide. Implementation of crystal Doppler kernel have been done in resonance analysis code besides the classical free gas model to analyse these new measurements. Concerning the reactor physics consequences from these experiments, the results shall be published in 1998 in a review specialised to nuclear reactor engineering.

The 3000 K furnace is ready and available at **ILL** since the end of 1997. Work is going concerning a convenient sample holder. Considerable attention is given to prevent chemical reactions between the uranium sample and its surroundings inside the furnace. With this solved the high temperature measurements on  $\text{UO}_2$  could begin during this year at IRMM (Geel, Belgium).

### Networking and training activities

Two network meetings have been arranged during the contract period. The first was held on May 31, 1996 in Grenoble, where the network discussed mainly the goals of the high temperature experiments and the status of the low temperature measurements. The second network meeting was held on November 23, 1997 at Saclay, where Zeyen from ILL showed the advance of the furnace design. The 3000 K furnace design was well engaged by the ILL high temperature team. Some difficulties were encountered due to the large sample size required for the Doppler experiments. The heating power and water cooling the outer aluminium parts had to be carefully optimised to achieve the required performance. We discussed the problem of the sample holder which should not chemically react with the  $\text{UO}_2$  sample heated to very high temperature. The Doppler Network met CEA specialists of  $\text{UO}_2$  to help us to find the best solution. The planning of the high temperature measurements was considered in some detail including the first heating tests at Grenoble of the furnace and thereafter with a  $\text{UO}_2$  sample. If these tests are successful the furnace shall be transported to the Geel (Belgium) neutron time-of-flight spectrometer to start the measurements at high temperature. During the Saclay meeting reactor physicists joined the Doppler Network to explain what can be learned from our measurements in terms of reactor stability calculations.

Two young scientists (A.Royer and A.Tagziria) were involved as postdoc's in the experimental data taking and analysis.

### Conclusions

The low temperature measurements has been finished according to the network program but the results are not yet fully analysed. Each partner participated as planned in the work program. All the results are published or will soon be published in the open literature. For the high temperature measurements, the main achievement according to the network program is the availability of the 3000 K furnace. The high temperature measurements on  $\text{UO}_2$  samples shall start at IRMM (Geel, Belgium) during 1998.

### List of publications

- [1] "Measurements to investigate the Doppler-broadening of  $^{238}\text{U}$  resonances, part 1"  
A. Meister, H. Tagziria, A. Royer, H. Weigmann, C. Burkholz, C. Van der Vorst  
NEUTRON DATA, GE/R/ND/02/95, November 1995
- [2] "Measurements to investigate the Doppler-broadening of  $^{238}\text{U}$  resonances, part 2"  
A. Meister, H. Tagziria, A. Royer, H. Weigmann, C. Burkholz, C. Van der Vorst  
NEUTRON DATA, GE/R/ND/01/96, March 1996
- [3] "Precise measurement of  $^{238}\text{U}$  resonances at low temperature"  
EC Network Co-operation : "Study of Solid State properties by Neutron Resonances"  
A. Meister, H. Tagziria, A. Royer, H. Weigmann, C. Burkholz, C. Van der Vorst  
P. Ribon, C. Mounier (CEN Saclay), C. Coceva (ENEA Bologna), H. Postma (TU Delft),  
C. Zeyen (ILL Grenoble), H.-G. Priestmeyer (University Kiel)  
conference Spring Meeting of the German Society of Physics (DPG), "Physics of Hadrons  
and Nuclei" Stuttgart, 25-29 March 1996.
- [4] "Study of Uranium neutron resonances at low temperature", A.Meister, P.Ribon,  
H.Tagziria, A.Royer, A.Brusegan, H.Postma, H.-G.Priestmeyer, International  
Conference on "Nuclear data for science and technology" May 19-24, 1997, Trieste, Italy
- [5] "Experimental setups for the study of Doppler broadening of neutron resonances from  
13 to 3000 K", A.Meister, P.Ribon, P.Siegler, C.Bürkholz, C.van der Vorst,  
A.Brusegan, M.Moxon, C.Coceva, H.Postma, C.Zeyen, H.-G.Priestmeyer, International  
Conference on "Nuclear data for science and technology" May 19-24, 1997, Trieste, Italy
- [6] paper (TITLE?) to be submitted in 1998 to Nuclear Science and Engineering »