

# Experimental Tests of the Crystal Lattice Model of the R-Matrix Code SAMMY

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**Abstract.** In the evaluation of low-energy cross-sections, the broadening of neutron resonances due to the motion of the target nucleus was thoroughly studied. The widely used model for Doppler Broadening is the Free Gas Model (FGM) that neglects atomic interaction between target atoms. When the target atoms are bound to a molecule or a crystal, the incident neutron energy can change the atomic vibrational or rotational quantum state and a more realistic treatment is needed. In the early days of cross-section theories, Lamb [1] derived a theoretical model, known as the Crystal Lattice Model (CLM), to account for the emission or absorption of one or more phonons (quanta of vibrational energy). This model was recently implemented in the Bayesian R-matrix fitting computer code SAMMY [2]. In the present work, the Crystal Lattice Model of SAMMY is tested against transmission experiments performed at GELINA (GEel LINEar Acelerator) on  $^{238}\text{U}$  [3] with U and  $\text{UO}_2$  samples at 23.7K and at room temperature. The CLM of SAMMY is then used to improve the evaluation of  $^{238}\text{U}$  resonance parameters in progress at the Oak Ridge National Laboratory (ORNL).

## INTRODUCTION

A new evaluation of  $^{238}\text{U}$  resonance parameters is underway at the ORNL laboratory [4] primarily to extend the resonance range up to 20 keV and to investigate discrepancies in thermal integral systems calculations in the framework of the Working Party on International Nuclear Data Evaluation Cooperation (WPEC/subgroup-22). The use of the Free Gas Model (FGM) for Doppler broadening of the low-energy resonances was again questioned. The Crystal Lattice Model (CLM) was recently put into the SAMMY code in order to account for the presence of chemical binding in the sample. The main goals of this paper are to test the CLM of SAMMY on the low temperature uranium experiments performed at GELINA where the solid state effects are strong and to include the model in the new  $^{238}\text{U}$  evaluation in progress at ORNL.

## THE CRYSTAL LATTICE MODEL

The first theory of neutron cross-section with bound nuclei was made by E. Fermi under the Born Approximation and using the "pseudopotential" method, primarily to describe potential neutron scattering. For resonant capture cross-sections, a derivation was proposed by W. E. Lamb [1] in 1939 shortly after the pioneering paper of Breit and Wigner on the shape of neutron resonances. Lamb's approach [1] used the quantum first-

order perturbation formula to express the probability for the system in its initial state (neutron of mass  $m$  and target nucleus of mass  $M$  bound in a crystal) to form the intermediate state (excited compound nucleus and crystal) and decay into a final state. In the case of a free target and isolated resonance, perturbation theory<sup>1</sup> gives straightforwardly the Single-Level Breit-Wigner (SLBW) resonance shape for the capture cross-section.  $\sigma_\gamma(E) \propto \Gamma_n \Gamma_\gamma / ((E - E_0)^2 + \Gamma^2/4)$ . When the nucleus is chemically bound, assuming the internal motion of the nucleons in the nucleus to be independent of the center of mass motion, the quantum perturbation formula gives a modified SLBW shape that accounts for the transfer of neutron momentum and energy to the crystal :

$$\sigma_\gamma(E) \propto \Gamma_n \Gamma_\gamma \sum_{a,b} \frac{g(a) | \langle b | e^{i\vec{k}\cdot\vec{r}} | a \rangle |^2}{(E - E_0 + \epsilon_a - \epsilon_b)^2 + \Gamma^2/4} \quad (1)$$

Here  $|a\rangle$  and  $|b\rangle$  stand for the initial and intermediate quantum states of the crystal with energy  $\epsilon_a$  and  $\epsilon_b$ .  $E$  and  $\vec{k}$  are the energy and momentum of the incident neutron and  $E_0$  the resonance energy. The capture cross-section  $\sigma_\gamma(E)$  is averaged over the distribution of initial crystal states  $g(a)$  at temperature  $T$  and summed over intermediate states  $b$ . Equation (1), using integral

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<sup>1</sup> Not valid, in principle, for compound nucleus process

representation of the Dirac function, can be written into a suitable form :

$$\sigma_\gamma(E) = \int_{-\infty}^{\infty} S(\vec{k}, E') \sigma_\gamma^{T=0}(E - E') dE' \quad (2)$$

in which  $\sigma_\gamma^{T=0}(E)$  represents the SLBW form of the un-broadened capture cross-section. The function  $S(\vec{k}, E')$  is the well known "scattering kernel" used in neutron thermal scattering theory and is usually presented as the Fourier transform of the so-called "intermediate function"  $\chi(\vec{k}, t)$ ,

$$S(\vec{k}, E') = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iE't} \chi(\vec{k}, t) dt. \quad (3)$$

Calculation of  $\chi(\vec{k}, t)$  requires knowledge of the displacement operator  $\vec{u}$  of the target atoms around the equilibrium position, as well as the crystal wave functions  $|a\rangle$  and  $|b\rangle$ . In lattice dynamic theory, a crystal is described through normal modes of vibrations or phonons (labeled by wave vector  $\vec{K}$  and branch index  $j$ ). We note  $q = \vec{K}, j$ . The energy of mode  $q$  is then quantized as follow:  $\varepsilon_q = (n_q + 1/2) \hbar\omega_q$ . An elegant method avoiding explicit calculation of the harmonic-oscillator wave functions and matrix elements was first presented by Van Hove [5] using the Heisenberg time representation of operators. It can be shown that  $\chi(k, t)$  is a simple function of the displacement operator,

$$\chi(\vec{k}, t) = e^{-\langle (\vec{k} \cdot \vec{u}(0))^2 \rangle_T} e^{\langle \vec{k} \cdot \vec{u}(0) \vec{k} \cdot \vec{u}(t) \rangle_T}. \quad (4)$$

The notation  $\langle \dots \rangle_T$  stands for the average of matrix elements over the distribution of crystal states,  $\langle X \rangle_T = \sum_a g(a) \langle a | X | b \rangle$ , taken at temperature T. The thermal average is performed with the Bose-Einstein distribution. The time-independent factor (Debye-Waller factor) and the time-dependent factor in Equation (4) are more easily calculated for cubic Bravais lattices where the average value of  $\vec{k} \cdot \vec{e}_q$  over all the relative orientations of the crystal and neutron beam is  $|\vec{k} \cdot \vec{e}_q|^2 = 1/3 k^2 |\vec{e}_q|^2$ . In addition, the discrete sum over the excitation modes  $q$  can be replaced by an integral assuming a continuous description of the weighted phonon frequency spectrum  $\rho(\hbar\omega)$ .

The final result for  $S(\vec{k}, E')$  becomes :

$$S(E, E') = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{iE't} e^{\frac{mE}{M+m}[\gamma(t) - \gamma(0)]} dt \quad (5)$$

and

$$\gamma(t) = \int_0^{+\infty} \frac{\rho(\hbar\omega)}{\hbar\omega} \left[ \coth\left(\frac{\hbar\omega}{2kT}\right) \cos(\omega t) + i \sin(\omega t) \right] d\hbar\omega \quad (6)$$

These formulae have been shown to be valid for resonant scattering (in the single collision approximation that neglects multiple scattering on the same scatterer) [6], fission [7], and total cross-section. However, from a theoretical point of view, the first-order perturbation used by Lamb for the description of nuclear resonance is questionable. Despite additional work of Trammel et al. [6] for the scattering process, a rigorous multilevel and multichannel formulation for resonant neutron-crystal interaction is still missing. Other approximations have been considered: the dependence of the barrier penetrabilities on the phonon transfers is neglected in Lamb's derivation. It was assumed that  $\Gamma_n$  and  $\Gamma_t$  are not significantly modified by phonon creation or absorption. This assumption has been studied by several authors and appears to be accurate for heavy nuclides in practical applications. Furthermore, anharmonic effects (giving a finite phonon lifetime) have been investigated, but contradictory results have been obtained. The effects of anharmonicity might be important at high temperature.

The CLM of SAMMY is based on the DOPUSH [8] code. The DOPUSH code is similar to the LEAPR code of NJOY [9] which does not compute directly the integral in the  $\gamma(t)$  function (because of its strongly oscillating behavior) but calculates  $S(E, E')$  with the usual phonon expansion methods.

## ANALYSIS OF U AND UO<sub>2</sub> GELINA TRANSMISSION MEASUREMENTS

The experiments were performed at the GELINA facility by Meister et al. [3] and the main features are presented in Table 1. The data (transmission as a function of neutron energy) were taken from the EXFOR database and have been corrected for various experimental conditions such as energy calibration, dead time correction, background subtraction with black resonances technique and normalization.

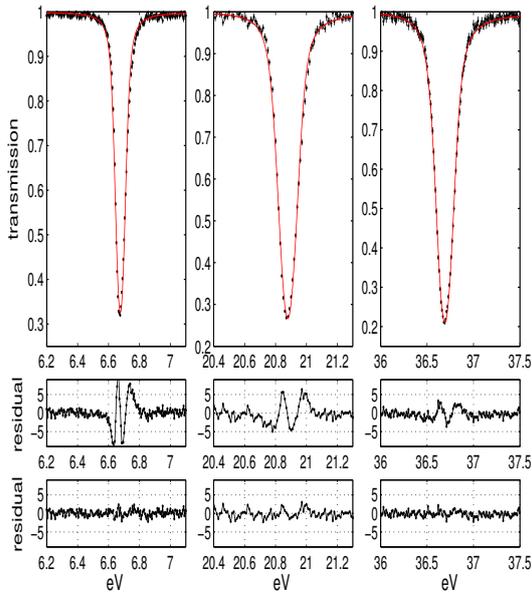
**TABLE 1.** Summary of the GEEL measurements

Flight path number 2	l=26.48 m
Flight path angle	9°
Moderator	Be + water of 3.6 cm thickness
Electron burst FWHM	15 ns
neutron detector	<sup>6</sup> Li (NE905)

Four samples (thin and thick *U* and *UO<sub>2</sub>* samples) have been measured at 293K and 23.7K. The temperature was controlled by a cryostat to achieve good accuracy. Data analysis was performed with the pre-released M7 version of the SAMMY code that includes CLM. The comprehensive experimental reports by Meister et al. permit construction of the full resolution function (energy spread mainly due the shape of fast neutron pulse,

the moderation in water, the beam geometry and angle of the flight path as well as the path of neutron in the  ${}^6\text{Li}$  detector) which was input as a pointwise distribution in the SAMMY code.

The Reich-Moore fits were performed using a priori resonance parameters of Moxon et al. [10], and the effective scattering radius is not adjusted ( $R_{eff} = 9.43 \text{ fm}$ ). The most severe test of CLM is the measurement of the  $UO_2$  sample at 23.7K. The asymmetry of the resonance is pronounced and cannot be reproduced with FGM as shown in Figure 1. The CLM used as input phonon spectrum data measured by Dolling et al. [11] with the one-phonon coherent scattering method. The *weighted* phonon spectrum is deduced from normalized amplitude of the uranium atoms ( ${}^{238}\text{U}$  in  $UO_2$ ) and features two acoustic modes at about 14 meV and 21 meV and a broader peak at around 35 meV. As displayed in Figure 1, the CLM gives a very good representation of the solid state effect in the resonance at 6.67 eV, 20.8 eV and 36.7 eV. For the metallic samples, a single peak centered around 11 meV was found to be sufficient to describe the solid state effect.



**FIGURE 1.** SAMMY fit of  $UO_2$  transmission data from Meister et al. at GELINA ( $T=23.7\text{K}$ ). The middle figure shows the residual for the FGM fit with adjusted temperature; the bottom figure is the residual for the CLM fit.

## THE 6.7 EV RESONANCE OF ${}^{238}\text{U}$

For Doppler broadening when solid state effects are present, two methods are commonly employed :

- The Free Gas Model is used with an effective temperature deduced from the phonon spectrum when the "Lamb condition" (weak binding) is fulfilled i.e; when  $\Gamma + \Delta \gg \hbar\omega_m$ .  $\Gamma$  is the total nuclear resonance width,  $\Delta = \sqrt{4ET/A}$  is the Doppler width, and  $\hbar\omega_m$  is the maximum energy of the phonon spectrum. This condition is not met for the low resonances of  ${}^{238}\text{U}$  below 100 eV.
- Alternatively, the effective temperature can be treated as a fitted parameter and subject to vary from resonance to resonance.

When neither of these alternatives is adequate, then the CLM must be used. In order to assess the impact of Crystal Lattice Model in real applications, the CLM was studied for the ORNL evaluation of the  ${}^{238}\text{U}$  resonances. Below 250 eV, the ORNL evaluation of radiative and neutron widths is based on the Olsen et al. transmission data [12] and part of the de Saussure capture data [13]; both measurements were performed at ORELA using metallic uranium samples at room temperature. In the Olsen experiments, from 5 eV to 4 keV, seven sample thicknesses were measured using a 42 m flight path.

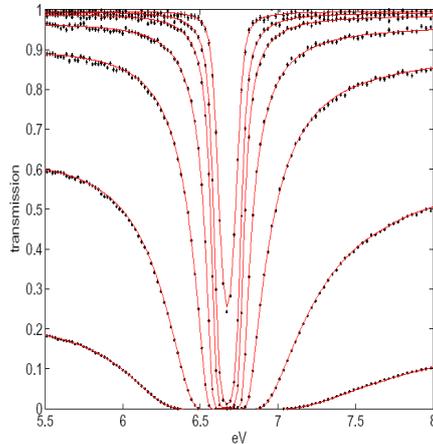
First as in [8], a numerical study was carried out. Using resonance parameters from Moxon et al., transmission data were generated with CLM (phonon spectrum featuring a peak at 11 meV) and fitted with FGM. In this numerical experiment, only uncertainty values were taken from the Olsen data. As shown in Table 2, when using metallic samples at room temperature, the most significant bias is observed for thinnest samples on radiative widths. Bias appears to be negligible for thick samples (note that the resonance is black).

**TABLE 2.** Numerical experiments : fit of transmission generated by CLM with FGM and free temperature for several thicknesses.

Sample thickness (at/b)	T (K)	$\Gamma_\gamma$ (meV)	$\Gamma_n$ (meV)
<b>Ref : Crystal Lattice Model</b>			
	293.0K	23.00	1.493
<b>Free Gas Model + fitted Temperature</b>			
0.000185	288.54K	23.64	1.494
0.001239	296.51K	23.48	1.478
0.01239	302.04K	23.30	1.475
0.17536	292.36K	22.99	1.494

Then, sequential SAMMY fits of Olsen data from 0.5 eV to 16 eV were performed with (1) FGM using a fixed temperature  $T = 300 \text{ K}$ , (2) FGM with a fitted temperature, (3) CLM with thermodynamical tempera-

ture  $T = 293K$ . The three fits were performed under the same conditions: resolution function, prior uncertainty on normalisation and background correction. The effective scattering radius was not adjusted ( $R_{eff} = 9.45fm$ ).



**FIGURE 2.** SAMMY Fit of the seven metallic samples data of Olsen et al. with the Crystal Lattice Model

**TABLE 3.** Comparison of FGM versus CLM resonance parameters of the 6.7 eV resonance from sequential fits of Olsen et al. [12] transmission data. Uncertainty values take into account only statistical errors and are strongly underestimated

Method	$E_r$ eV	$\Gamma_\gamma$ meV	$\Gamma_n$ meV
$R' = 9.45fm$			
FGM $T = 300K$	6.6742	23.09	1.471
FGM $T_{fit} = 296.8K$	6.6741	23.08	1.473
CLM $T = 293K$	6.6734	23.02	1.476
Statistical uncertainty	0.0001	0.02	0.001

Table 3 shows that the resonance parameters are not changed significantly with CLM, mainly because of the use of metallic samples at 293K. Moreover, despite the use of 7 sample thicknesses in the fitting process, the present value of the radiative width is almost entirely given by the thick sample data (0.175 atom/barns) where the impact of CLM is the smallest.

## CONCLUSIONS

The Crystal Lattice Model implemented in SAMMY was tested on  $^{238}U$  transmission measurements performed by Meister and co-workers at GELINA. The CLM accurately reproduced the shape of the first three resonances of  $^{238}U$  when solid state effect are strong.

To improve the evaluation of  $^{238}U$  resonances, in progress at ORNL, the CLM was also used to analyse the ORELA Olsen measurements. Compared with the usual

Free Gas approach, a small impact has been found on extracted resonance parameters.

The great advantage of the crystal broadening model is to avoid the problem of choosing the effective temperature; however, it assumes that the phonon spectrum of the crystal is known. Its major drawback is a significant increase of computation time.

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