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On the Influence of Crystalline Binding on Resonant Absorption and Reaction Rates

by

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Abstract

At present time, experimental transmission data are analyzed with codes like REFIT or SAMMY which use the free gas model to fit the form of the resonances. The use of the resonance parameters issued from such analysis for further reconstruction of the cross section with codes like NJOY can result in non negligible errors in the cross-sections as well as in the reaction rates.

To analyze the bias introduced on resonance parameters by the use of the free gas model and its consequences on reaction rates we set up a numerical experiment that closely follows the actual scheme of the nuclear data evaluation

First, we use resonance parameters from JEF2.2 nuclear library to calculate our reference cross section with Lamb's harmonic crystal model. This cross section is then used to simulate a transmission coefficients, and a new set of resonance parameters are obtained using the code REFIT to fit the shape of the transmission with the help of the free gas model. These resonance parameters are used to estimate the errors on the reaction rates.

We conclude that the free gas model does not ensure reaction rate conservation. A comparison of the capture rates showed that the discrepancy between this model (with the bias on the resonance parameters described above) and the harmonic crystal model (with initial JEF2.2 parameters) is important for reactor physics. For the first resonance of uranium, which represents 30% of the absorption in a thermal nuclear reactor, the error in the capture reaction rates reaches 3% for the biased resonance parameters issued from UO_2 analysis, and up to 1% for the biased resonance parameters issued from metallic uranium analysis. Such discrepancy could be corrected using a crystal model for the experimental data analysis.

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0INTRODUCTION

Solid state effects on the shape of the cross section at epithermal energies are customarily taken into account by using the monatomic gas model with an effective temperature instead of the thermodynamic temperature (EFG model). As suggested by Lamb,[i] for a weakly bound crystal this effective temperature can be deduced from a appropriate phonon spectrum. It has been tacitly assumed that this procedure gives good results for most reactor materials.

It was soon recognized that the chemical compound dynamics has to be considered in resonance shape analysis of differential measurements. Fundamental works like that of Jackson and Lyn[ii] showed that the free gas (FG) approximation is insufficient for tight bonded materials like U_3O_8. The implication is that experimental data should be analyzed with more realistic models like Lamb's harmonic crystal (HC) model that we have chosen for the present study. Jackson and Lyn also emphasised that good agreement between the experimental data and the FG model was only achieved for weakly bound compounds, like metallic uranium. However in the nuclear libraries,[iii] that are used for reactor applications, the resonance parameters are usually calculated by fitting the experimental data to the FG model. Codes for resonance analysis, such a REFIT[iv] and SAMMY[v] use a least square adjustment to determine the values of resonance parameters (partial widths and resonance energy) and the best temperature T_A to be used in the FG model. The consequences of the use of the FG model at this stage are twofold. First, one can obtain the wrong (biased) resonance parameters which will introduce errors in the cross-section values as well as in the reaction rates. Second, the temperature T_A can be very different from the effective temperature T_{eff} deduced from a phonon spectrum. The bias on the resonance parameters related to the use of the FG model, present in today's nuclear data evaluation, deserves to be considered in detail.

In the next stage the code NJOY[vi] is widely used for nuclear data processing. In the resonance domain the cross sections are calculated in different ways. The FG model is applied for all types of cross sections with the exception of the transfer cross sections. The later are calculated with a more naive approach, that of the free nuclei at rest. The use of the FG model in the code NJOY is coherent with the model employed for experimental data analysis. What we question here is the use of T_{eff} in reactor neutronics. We note that neither T_A nor T_{eff} do not ensure reaction rates conservation which is of paramount importance in reactor physics : the temperature that actually interests the reactor physicists is the one that preserves the reaction rates. This temperature, which will be noted hereafter as T_R, can be obtained by minimizing the difference between the reaction rates issued from the FG model and that issued from the crystal model.

The purpose of the present paper is to assess the errors introduced by the use of the FG model at different stages of nuclear data evaluation, from the differential measurements to the reaction rate calculation. The principal points to be investigated are the following. First, what is the error introduced on the resonant parameters by the use of the FG model in resonance shape analysis ? Second, what is the error on the reaction rates due to the use of the EFG model instead of Lamb's crystal model ?

To answer the first question we will numerically simulate a transmission experiment, using the HC model with resonance parameters from JEF2.2 as a reference. Then with the help of the code REFIT we will fit our reference transmission coefficients to the FG model to deduce a new set of resonance parameters and the temperature T_A that gives the best agreement. Such simulation represents somewhat the actual scheme of the nuclear data evaluation and gives us an insight on the bias on the resonance parameters inferred from the use of the free gas model.

To address the second question we will compare the capture reaction rates, calculated with the HC model, with initial JEF2.2 parameters, with those calculated with the FG model with biased parameters.

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0METHOD OF CALCULATION

From the numerical point of view, the computation of the scattering function for an harmonic crystal is strictly equivalent to that for thermal neutron scattering. Therefore we have used for its calculation the code LEAPR[vii] recently implemented in the code NJOY by MacFarlane. Some minor modification were necessary, however, to adapt this code to our purposes. The code LEAPR calculates integral (4) by expanding the integrand in a power series, each term of which gives the contribution of the corresponding phonon process. A program was written to calculate convolution integral (3) ; the convolution with the elastic contribution, which is especially important at low temperatures and energies, requires no computational effort because it corresponds to a recoilless interaction. The precision of the calculation was attributed by the verification of the well known sum rule and of the normalization condition. In our calculation the precision was fixed at 0.1%.

1RESULTS

1NUMERICAL SIMULATION OF A TRANSMISSION EXPERIMENT

To investigate the errors introduced by the use the free gas model in resonance analysis we have simulated several transmission experiments for two compounds of ^{238}U, uranium dioxide and metallic uranium. In this « thought » experiment an initial set of resonance parameters from JEF2.2 library and an appropriate phonon spectrum were used in Lamb's crystal model to generate cross sections at a given temperature ; from these cross sections one straightforwardly computes transmission coefficients for different sample thicknesses. This numerical simulation eliminates the errors arising from experimental resolution broadening. Statistical errors, however, were considered and taken proportional to the square of the transmission coefficients.

For each one of these « experiments » a new set of resonance parameters was obtained from a shape fit carried out with the code REFIT. The analysis of the transmissions in this code is done with the help of the FG model. Therefore, the new set of resonance parameters is somewhat biased and the errors can be quantified by comparing these parameters with the original ones used in the simulation.

To control our calculational scheme we repeated each calculation but this time with transmission coefficients derived from the EFG model. In these conditions, the shape investigation with REFIT reproduced the initial JEF2.2 values of the resonance parameters and the effective temperature $T_{eff}=T_A$.

We have checked our reference calculations by comparing the total cross section calculated with the crystal model to experimental values. In the Figures 1 and 2 we present a comparison between our calculations and the recent measurements of ^{238}U in UO_2 and metallic uranium done at GEEL facility.[viii] The resonance parameters used in our calculation are the following : Γ_n=1.493 meV, Γ_γ=23 meV, E_r=6.674 eV.

The values of the resonance parameters obtained from the shape analysis of our simulated data for UO_2 and metallic uranium are given in Tables I through III for different temperatures and sample thicknesses. These tables show that the adjusted temperature T_A is thickness dependent. At room temperature the value of T_A is always lower than the thermodynamic temperature and, consequently, than the effective temperature T_{eff}. The explanation of this fact lies on the use of the FG model, instead of the more accurate crystal model, and is revealed by the violation of Lamb's condition at low temperatures and energies. We note that the use of the more elaborated anharmonic crystal model, proposed by Karam and d'Avila[ix] gives following inequality $T_{eff}<T$.

2REACTION RATES

The use of the free gas model may induce errors on the values of the reaction rates in two different ways. First, because of the bias on the resonance parameters obtained from shape analysis of transmission experimental data, and second because of the use of the

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gas model in the nuclear data processing stage done with codes such NJOY. In this section we separately analyze both sources of errors.

The bias on resonance parameters depends on the chemical compound (uranium dioxide or metallic uranium), the thickness of the sample and the temperature. A realistic estimation of the errors is rather difficult because the body of experimental data used in the evaluations is quite heterogeneous : transmissions or capture yields of uranium for different compounds and sample thickness. Therefore, the estimations given here ought to be considered as an indication of a trend. We have derived upper and lower limits for the errors on the reaction rates from two different sets of resonance parameters issued from REFIT analysis for UO_2 (Table I) and for metallic uranium (Table III) for temperature of 293.16 K and a sample thickness of 2.86 10^{-4} atoms/barn. Figures 3 and 4 show a comparison of the capture reaction rates obtained for these biased parameters with the two free gas models (FG and EFG) with those calculated with the initial JEF2.2 parameters with the HC model. We see in these figures that the free gas models overestimate the reaction rates at 293.16 K, up to 3% for the biased resonance parameters issued from UO_2 analysis, and up to 0.9% for the biased resonance parameters issued from the analysis of metallic uranium. Similar trends, less pronounced, are still present at 973.16 K.

Next, we assume that the initial resonance parameters are correct and compare the capture reaction rates issued from the nuclear data processing stage with the HC and the two free gas models. We have analyzed the first resonance at temperatures of 293.16 and 973.16 K with the WR (wide resonance) approximation. The relative errors between each gas models and the crystal model are given in Figures 5 and 6 versus the value of the dilution cross section. The figures show that the gas model values are within 1% of the crystal values. At room temperature and higher, the EFG model is closer to the HC model, and the error of the FG model is 2 to 3 times higher than that of EFG model. For the second resonance we found that the discrepancy between the HC and the FG models is smaller due to a better verification of Lamb's condition. We have noted, as well, that the HC model is not too sensitive to the values of the resonance parameters : the use of biased parameters does not produce large errors in the reaction rates.

2CONCLUSIONS

In this work we have analyzed the errors introduced on the reaction rates by the use of the gas model instead of the more realistic crystal model. Our study shows that the major source of error comes from the resonance shape analysis stage. We remaind the reader that, for uranium in PWR with a typical dilution of 50 barn, the errors in reaction rates caused by a biased resonance shape analysis are nearly ten times greater that the errors caused by a biased Doppler model itself. Therefore, the predicted values of the reaction rates would be improved by accounting for crystalline effects in the evaluation of resonance parameters, even though the nuclear data processing code NJOY still uses the free gas model. Further, if one wants to keep using the free gas model in NJOY, then a supplementary improvement at the reactor computational level could be gained by using the free gas model with the temperature T_R that preserves the reaction rates predicted by the crystal model.

To draw a more complete picture of the influence of chemical binding on the reaction rates and to determine the temperature T_R to be used in reactor neutronics, further study taking into account the effect of harmonic crystalline bindings in the values of the transfer cross section will be necessary. These cross-sections could then be used for solving the slowing-down equation in the multigroup approximation, for instance with the APOLLO2$^{\times}$ transport code. This approach will permit us to get a clearer view of crystalline effects on the reaction rate and on the determination of T_R. We will reserve the conclusion on the value of this temperature to a future study in which the influence of crystalline binding will be taken into account for all cross sections.

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TABLE I

Adjusted resonance parameters (Γ_n, Γ_γ, E_r) and gas temperature (T_A) obtained from shape analysis of samples of different thicknesses (n) from a simulated transmission experiment for UO2 at 293.16 K. The effective temperature derived from the corresponding phonon spectrum is T_{eff}=307.16 K.

n(atoms/barn)	Γ_n (meV)	Γ_γ (meV)	E_r (eV)	T_A (K)
10^{-4}	1.5011	25.848	6.6731	276.6
2.8610^{-4}	1.4999	25.346	6.6735	281.0
$6.0\ 10^{-4}$	1.4962	24.704	6.6742	287.9

TABLE II

Adjusted resonance parameters (Γ_n, Γ_γ, E_r) and gas temperature (T_A) obtained from shape analysis of samples of different thicknesses (n) from a simulated transmission experiment for UO2 at 77 K. The effective temperature derived from the corresponding phonon spectrum is T_{eff}=119.6 K.

n (atoms/barn)	Γ_n (meV)	Γ_γ (meV)	E_r (eV)	T_A (K)
10^{-4}	1.5043	25.944	6.6719	94.6
2.8610^{-4}	1.5117	25.291	6.6731	97.1
$6.0\ 10^{-4}$	1.5593	24.091	6.6750	96.9

TABLE III

Adjusted resonance parameters (Γ_n, Γ_γ, E_r) and gas temperature (T_A) obtained from shape analysis of samples of different thicknesses (n) from a simulated transmission experiment for metallic uranium at 293.16 K. The effective temperature derived from the corresponding phonon spectrum is T_{eff}=298 K.

n (atoms/barn)	Γ_n (meV	Γ_γ (meV)	E_r (eV)	T_A (K)

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10^{-4}	1.4968	23.968	6.6738	287.3
2.8610^{-4}	1.4961	23.761	6.6740	289.0
$6.0\ 10^{-4}$	1.4941	23.538	6.6742	291.6

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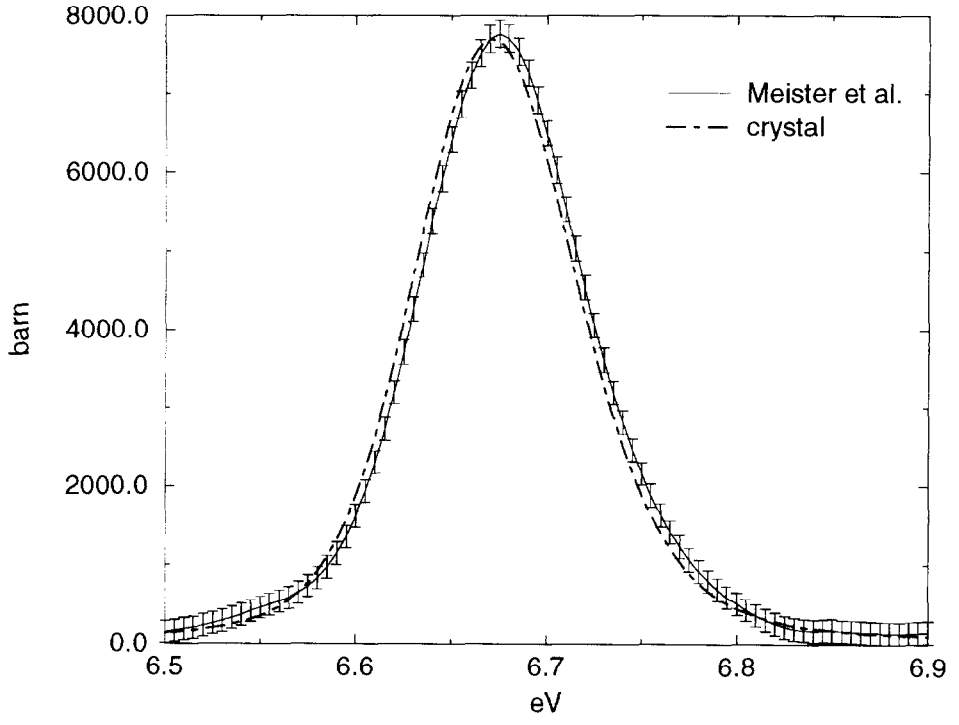

Figure 1
Total cross-section of 238U in UO2. for the first 6.674 eV resonance at T=293.6 K. Comparison between Lamb's harmonic crystal model and experiment (Meister et al.).

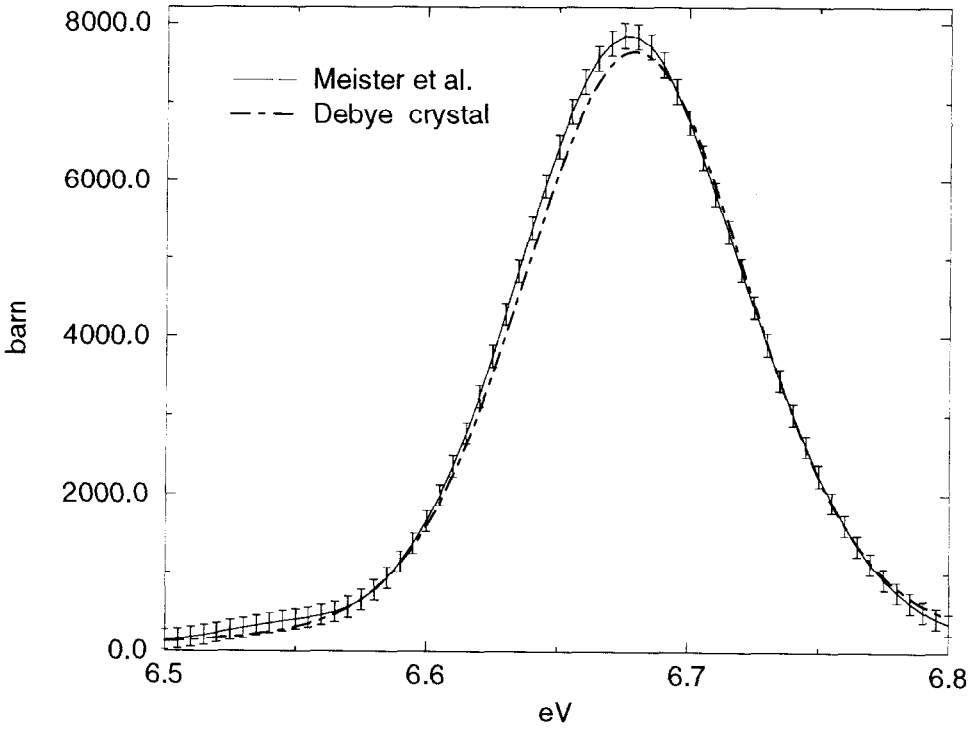

Figure 2
Total cross-section of metallic ^{238}U for the first 6.674 eV resonance at T=293.6 K. Comparison between Lamb's harmonic crystal model with Debye's spectrum and experiment (Meister et al.).

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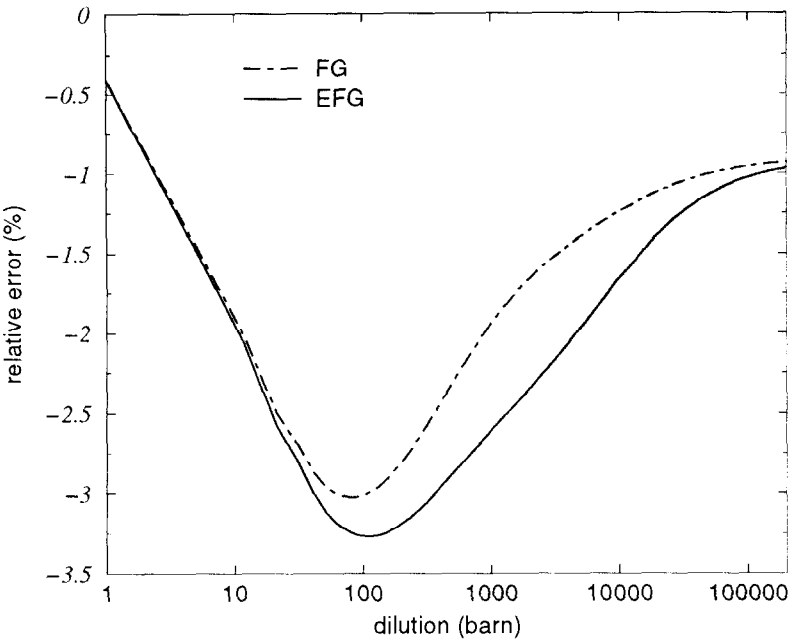

Figure 3

Relative errors for the capture reaction rates of 238U in UO2 versus the dilution cross section . Values calculated using the wide resonance approximation for the first resonance at 6.674 eV at 293.16 K. Sample thickness 2.86 atoms/barn. The relative error is defined as (ref-val)/ref where « ref » is the reference value obtained with the harmonic crystal model with the initial JEF2.2 parameters, and « val » denotes the value calculated with the free gas models (FG=free gas, EFG=free gas with effective temperature) with the biased resonance parameters taken from Table I.

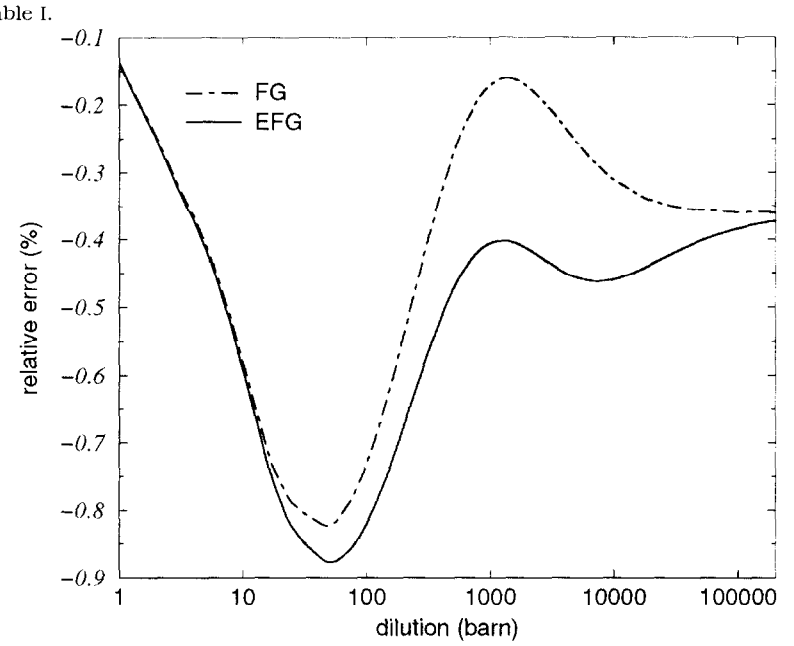

Figure 4

Relative errors for the capture reaction rates of ^{238}U in UO_2 versus the dilution cross section . Values calculated using the wide resonance approximation for the first resonance at 6.674 eV at 293.16 K. Sample thickness 2.86 atoms/barn. The relative error is defined as (ref-val)/ref where « ref » is the reference value obtained with the harmonic crystal model with the initial JEF2.2 resonance resonance parameters, and « val » denotes the value calculated with the free gas models (FG=free gas, EFG=free gas with effective temperature) with the biased resonance parameters taken from Table III.

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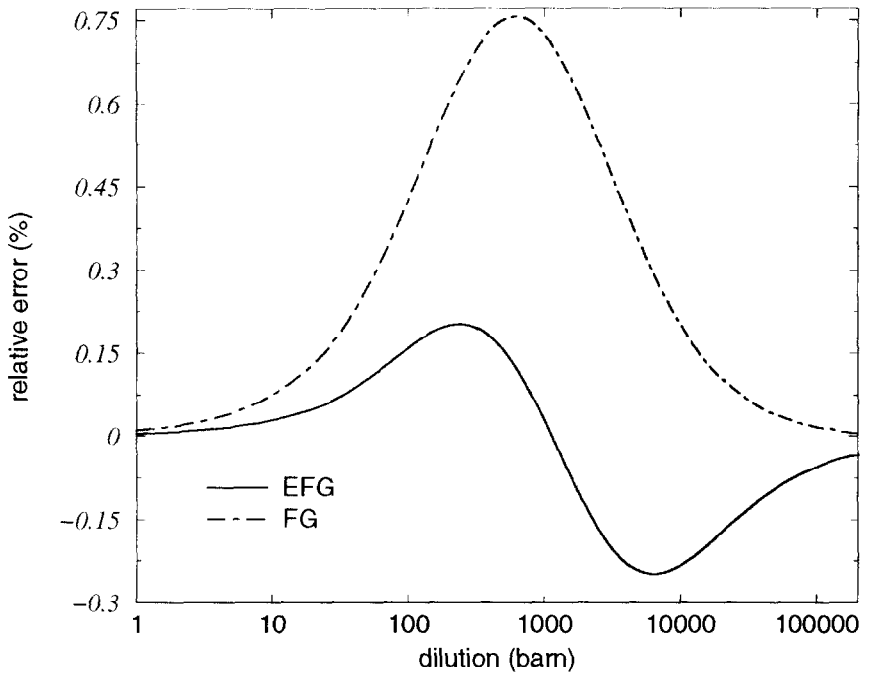

Figure 5

Relative errors for the capture reaction rates of ^{238}U in UO_2 versus the dilution cross section . Values calculated using the wide resonance approximation for the first resonance at 6.674 eV at 293.16 K. The relative error is defined as (ref-val)/ref where « ref » is the reference value obtained with the harmonic crystal model and « val » denotes the value calculated with the free gas models (FG=free gas, EFG=free gas with effective temperature) with the initial resonance parameters taken from JEF2.2.

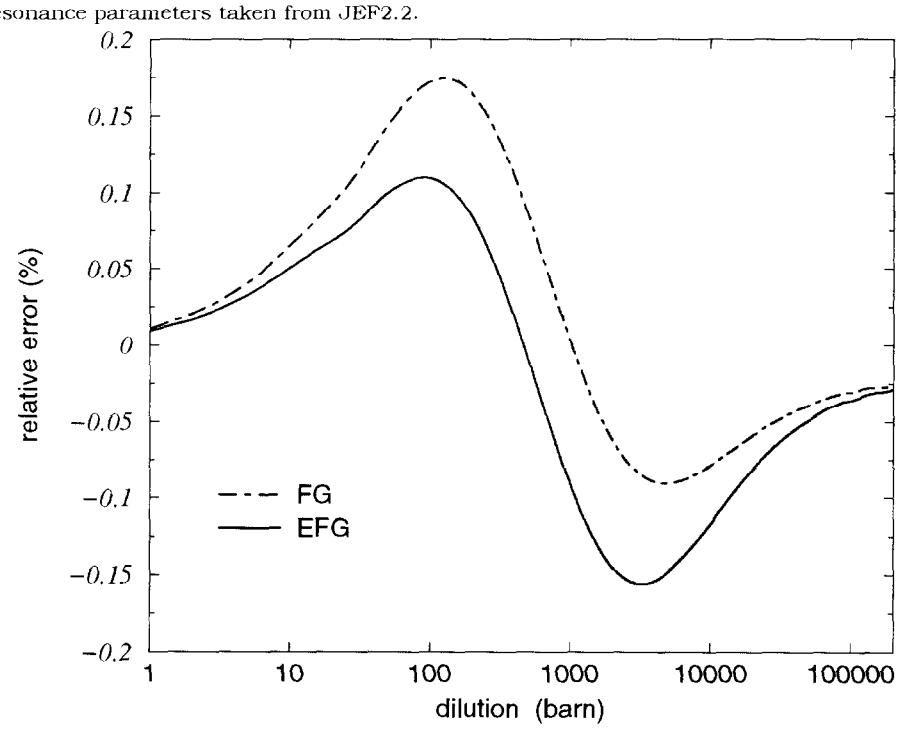

Figure 6

Relative errors for the capture reaction rates of ^{238}U in UO_2 versus the dilution cross section . Values calculated using the wide resonance approximation for the first resonance at 6.674 eV at 973.16 K. The relative error is defined as (ref-val)/ref where « ref » is the reference value obtained with the harmonic crystal model and « val » denotes the value calculated with the free gas models (FG=free gas, EFG=free gas with effective temperature) with the initial resonance parameters taken from JEF2.2.

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