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# INVESTIGATION OF THE FUSION-FISSION REACTION $^{208}\text{Pb} + ^{16}\text{O}$ AT SUBBARRIER ENERGIES

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A fission cross section of a  $^{224}$ Th compound nucleus has been measured in the reaction  $^{16}$ O +  $^{208}$ Pb in four energy points in the interval of 5—15 MeV below the fusion barrier, which made it possible to measure cross sections by 8 orders of magnitude lower than the geometrical cross section of heavy ion interaction with nuclei. It is evident that for ion energies of  $^{16}$ O  $\leq$  72 MeV the behaviour of the fission cross section deviates from the exponential drop, which testifies to the fact that structural effects have influence on the fusion cross section in the deep subbarrier case of the interaction of two spherical nuclei. The analysis performed made it possible to describe the fusion cross sections down to the lowest energies and to make certain statements on the necessity to consider in the calculation of the fusion cross sections the lowest vibration states of coupled nuclei. The sensitivity of the method enables one to go down in the cross section by four orders of magnitude to  $10^{-36}$  cm<sup>2</sup> which is by 12 orders of magnitude less than the geometric cross section of the nuclei interaction. The interest to further investigations in this energy region is explained by a possibility of obtaining additional information on the compaund states responsible for the cluster decay of nuclei.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR in collaboration with the INFN (Catania, Italy) and Istanbul University (Istanbul, Turkey).

## Исследование реакции слияния-деления $^{208}{ m Pb} + ^{16}{ m O}$ при подбарьерных энергиях

## Ю.Ц.Оганесян и др.

Измерено сечение деления составного ядра  $^{244}$ Th в реакции  $^{16}$ O +  $^{208}$ Pb в четырех точках по энергии в интервале от 5 до 15 MэВ ниже барьера слияния ядер, что позволило опуститься на 8 порядков вниз от геометрического сечения взаимодействия тяжелых ионов с ядрами. Видно, что для энергий  $^{16}$ O  $\leq$  72 MэВ ход сечения деления отклоняется от экспоненциальной зависимости, что свидетельствует о влиянии структурных эффектов на сечение слияния в глубокоподбарьерном случае взаимодействия двух сферических ядер. Проведенный анализ позволил описать сечения слияния вплоть до самых низких энергий и сделать определенные утверждения о необходимости включения в расчеты сечений слияния нижайших вибрационных состояний ядер партнеров. Чувствительность

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методики позволяет опуститься еще на четыре порядка по сечению вплоть до  $10^{-36}$  см<sup>2</sup>, что на 12 порядков меньше геометрического сечения взаимодействия ядер. Интерес к дальнейшим исследованиям в этой области энергий определяется возможностью получения добавочной информации о компаунд-состояниях ответственных за кластерный распал ядер.

Работа выполнена в Лаборатории ядерных реакций им. Г.Н.Флерова ОИЯИ в коллаборации с Национальным институтом ядерной физики (Катания, Италия) и Стамбульским университетом (Стамбул, Турция).

## Introduction

A great number of experimental data obtained in recent years on subbarrier fusion cross sections of heavy ions with nuclei in a wide range of A and Z compound nuclei has stimulated an extensive discussion on the mechanism of subbarrier reactions enhancement, on quantum effects and correlation between the dynamics of the process and nuclear structure of interacting nuclei. For the explanation of a substantial enhancement of the subbarrier reaction cross sections different models have been developed and widely used which take into consideration not only static deformation of nuclei, but also a possibility of weak fluctuation of nuclear surface in the interaction process, the possibility of vibrational states excitation and, finally, the influence of nucleon transfer channels on the fusion probability.

However, in our opinion, in a number of cases the existing experimental material is not sufficient for a more in-depth analysis of the possibility of different models' employment, especially in the case of spherical nuclei interaction, where the main enhancement effect, connected with the presence of static nuclei deformation, is absent. For example, there are many works, devoted to the study of subbarrier fusion in the reaction  $^{16}O + ^{208}Pb$ , however in all these investigations fusion-fission cross sections are obtained within a wide high-energy range, while in the subbarrier region there are only several points obtained at energies which are below the barrier by 1-8 MeV.

In the present work a fission cross section of a <sup>224</sup>Th compound nucleus has been measured in the reaction <sup>16</sup>O + <sup>208</sup>Pb in four energy points in the interval of 5—15 MeV below the fusion barrier, which made it possible to measure cross sections by 8 orders of magnitude lower than the geometrical cross section of heavy ion interaction with nuclei. Our interest in the study of such deep subbarrier interactions is connected not only with a possibility of investigating the fusion mechanism, but also with a possibility of investigating structural effects in different fission modes of compound nuclei, produced in reactions with heavy ions, whereas these investigations have been conducted only in reactions with light charged particles [1].

And finally, the study of deep subbarrier reactions using the nuclei of Pb and Bi is of special interest, since they make a basis for the synthesis of superheavy elements in cold fusion reactions on the one hand, and on the other hand, are daughter nuclei in the case of cluster decay of nuclei from Ra to U. The employed methods allow one to investigate deep subbarrier reactions up to the cross section level of  $10^{-36}$ , which opens up new perspectives for the analysis of the possibility of new elements synthesis as well as for the study of the inverse, with respect to cluster decay, reactions.

## Experimental Procedure

Beams of  $^{16}\mathrm{O}$  obtained from the tandem accelerator (NFIN, Catania, Italy) were focused onto a  $^{208}\mathrm{Pb}$  target located in the center of a diam scattering chamber (Fig.1). Beam intensities were typically of the order of 70 nA. A target of 270  $\mu \mathrm{g/cm^2}$  of  $^{208}\mathrm{Pb}$  (99.1% enrichment) was evaporated onto ~30  $\mu \mathrm{g/cm^2}$  carbon backing. The target was oriented with a carbon side facing the beam.

In this experiment the fusion-fission cross section for  $^{16}{\rm O} + ^{208}{\rm Pb} \rightarrow ^{224}{\rm Th}$  was measured for energies  $E_{\rm lab} = 78$ , 75, 73 and 68 MeV, which corresponded a range from 5 to 15 MeV below the Coulomb barrier. Single fission fragments were detected in the backward angular range of 90—164° and 198—270° by using mica dielectric detectors with an area of 170 cm<sup>2</sup>.

The mica detectors were located 13 cm away from target onto metallic backing covered with plastic film (Melinex), which detected background fission fragments arising onto mica after interaction of U-Th mica contamination with neutrons during irradiation. For determination of the neutron fluence, arising during each  $^{16}\mathrm{O}$  irradiation, the calibrated  $^{235}\mathrm{U}$  (0.5  $\mu\mathrm{g/cm^2}$ ) and  $^{238}\mathrm{U}$  (9.8  $\mu\mathrm{g/cm^2}$ ) sources in contact with plastic dielectric detectors (Melinex) were used. These «sandwiches» were located in the backward angles (90°; 135°; 180°) of the scattering chamber. Fissile element contaminations into target and support were determined by neutron radiography.

After each irradiation the <sup>208</sup>Pb quantity in the target was controlled by Induced Roentgen Fluorescence method.

The irradiated mica detectors were annealed during 6 hours at 460°C for decreasing the background events which arise in mica when the scattering <sup>16</sup>O ions interact with mica atoms and their compound nuclei give registered tracks. For scanning under optical microscope mica and plastic detectors were etched in 40% HF and 20% NaOH at 60°C, respectively.

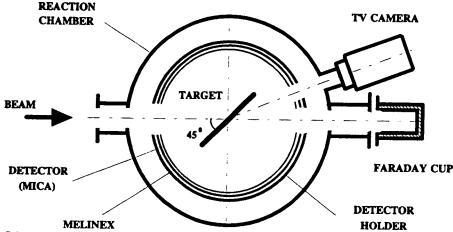


Fig.1. Scheme of the reaction chamber used in the fission cross-section measurements of  $^{224}$ Th isotope obtained by the  $^{208}$ Pb +  $^{16}$ O reaction

#### Results

The energies and fluences of accelerated  $^{16}O$  beams, used for this experiment, the loss energy interval for passing through the target of  $^{16}O$  beam, the obtained cross sections of the  $^{224}Th$  fission are presented in Table 1.

During irradiation with 73 MeV energy, when a 150 nA beam was used, the <sup>208</sup>Pb target lost 40% of initial matter.

Background from fissile element contanimation in the target, backing and mica was less than  $10^{-3}$  events for the whole detector area. After annealing the background mica tracks have not been observed.

Energy, MeV	Loss energy interval of <sup>16</sup> O for target, MeV	Fluence of <sup>16</sup> O, ions/cm <sup>2</sup>	Cross-section of <sup>224</sup> Th fission, mb	
78*	77.8—77.0	1.4 · 10 <sup>14</sup>	$7.8 \pm 0.1$	
75	74.8—74.2	$1.35\cdot 10^{15}$	$(3.7 \pm 0.1)10^{-2}$	
73	72.8—72.4	4.1 · 10 <sup>15</sup>	$(8.5 \pm 0.1)10^{-4}$	
68	67.8—67.2	5.6 · 10 <sup>15</sup>	$(6 \pm 3)10^{-6}$	

Table 1

## Analysis

This section we shall tentatively split into two parts. In the first one we shall present in brief the main ratios used at the description of subbarrier fusion cross sections, having skipped for the sake of simplicity the case of static deformation of partner nuclei, and in the second we shall consider the issues related with the description of the nuclei deexcitation process and with the cross sections calculation.

The fusion cross sections and the mean-square angular momentum are equal respectively to

$$\sigma_f = \sum_{l=0}^{l_{cr}} \sigma_l(E),$$

$$\langle l^2 \rangle = \frac{\sum_{l=0}^{l} \sigma_l(E)}{\sum_{l=0}^{l} \sigma_l(E)}.$$

<sup>\*</sup>Data was obtained using 300 µg/cm<sup>2</sup> <sup>208</sup>Pb evaporated onto a 40 µg/cm<sup>2</sup> aluminium backing

The partial wave cross section of fusion can be expressed in the following way

$$\sigma_l = \pi \cdot \chi^2 \cdot (2l+1) \cdot T_l$$

For the one-dimensional fusion barrier within the parabolic approximation for the total potential of interaction, taken as a sudden approximation, the transmission coefficient can be written down as

$$T_{l} = \left\{ 1 + \exp\left[\frac{2\pi}{\hbar\omega_{l}} (V_{bl} - E)\right] \right\}^{-1},$$

where

$$\hbar\omega_{l} = \left| \frac{\hbar^{2}}{2\mu} \cdot \frac{d^{2}V_{bl}}{dr^{2}} \right|_{r=R_{bl}}^{1/2}.$$

At the calculation of the subbarrier fusion cross sections on can use the Wong [2] approximation as the simplest one, in which it is assumed that  $(\hbar\omega_l = \hbar\omega_0, R_{bl} = R_{b0})$ 

$$\sigma_{\text{fus}}(E) = \frac{\hbar \omega_0 \cdot R_{b0}^2}{2E} \ln \left( 1 + \exp \frac{2\pi}{\hbar \omega_0} \left( E - V_{b0} \right) \right).$$

At the analysis of the subbarrier fusion cross sections, along with these equations some expressions linked with additional degrees of freedom are also used, in particular: the inclusion of the adiabatic potential, accounting of the vibrational zero-point motion of the surface, coupled-channels approach, etc. In the simplest approximation the accounting of the vibrational zero-point motion of the surface (Z.P.V.) can be described by the following expression

$$\sigma_l(E) = \frac{1}{2\Delta E} \int_{-\Delta E}^{\Delta E} \sigma_l(E - B_{\text{fus}} + \varepsilon) d\varepsilon,$$

where within the Esbensen model [3] the quality  $\Delta E$  can be related with the vibrational zero-point motion of the spherical surface with a Gaussian distribution of the interaction radius with a standard deviation

$$\sigma_{\lambda} = \frac{R}{Z \cdot (\lambda + 3)} \left[ (2\lambda + 1) \frac{B(E\lambda)}{B_{\rm sp}(E\lambda)} \right]^{1/2},$$

where  $\lambda$  is the vibration multipolarity or this quantity can be used as a free parameter. Calculations of the fusion cross sections were tested: (a) within the Wong approximation;

Table 2

σ fusion (Mb)								
E <sub>L,S</sub> (MeV)	experiment	CCFUS <sup>1)</sup>		Wong*)	Z, P, V*)	Igo		
		uncoupled	coupled					
67.5	6 · 10 <sup>-6 **)</sup>	10 <sup>-7</sup>	1.5 · 10 <sup>-6</sup>	6 · 10 <sup>-8</sup>	$1.2 \cdot 10^{-6}$	2 · 10 <sup>-4</sup>		
72.5	1.1 · 10 <sup>-3 ••)</sup>	1.2 · 10 <sup>-4</sup>	$1.6 \cdot 10^{-3}$	10 <sup>-4</sup>	$2.\cdot 10^{-3}$	$4\cdot 10^{-3}$		
74.5	4.2 · 10 <sup>-2 ••)</sup>	3 · 10 <sup>-3</sup>	4 · 10 <sup>-2</sup>		· <del>-</del>	10 <sup>-1</sup>		
75.5	3.0 · 10 <sup>-1</sup>	$2.5 \cdot 10^{-2}$	2.3 · 10 <sup>-1</sup>	1.1 · 10 <sup>-2</sup>	$2.5 \cdot 10^{-1}$	4 · 10 <sup>-1</sup>		
77.5	8.0**)	7.0 · 10 <sup>-1</sup>	5.7	$1.5\cdot 10^{-1}$	3	3.6		
80	40.0	18	57	5.8	30	43		
82	100.0	88	123	58	84	125		

<sup>1)</sup>  $^{208}\text{Pb}$ :  $\beta_2 = 0.06$ ,  $E_2 = 4.1$ ;  $\beta_3 = 0.1$ ,  $E_3 = 2.6$ ;  $^{16}\text{O}$ :  $\beta_2 = 0.4$ ,  $E_2 = 6.9$ ;  $\beta_3 = 0.5$ ,  $E_3 = 6.1$ ; parameter DV = -10

(b) within the standard approximation of an inverted parabola with a nuclear potential in the form of Igo with the following parameters:  $V_0 = 50$  MeV, d = 0.7 fm,  $r_0 = 1.23$  fm; (c) by the introduction of parameter  $\Delta E$  and selection of three parameters conditioned by the best agreement with the experimental data [4]; (d) by the method of coupled channels using a standard software package CCFUS [5]. Table 2 presents our experimental data, data from Ref.[6] and the results of calculating the fusion cross sections under these models.

It is evident that at an ion energy of  $^{16}\text{O} \le 75$  MeV the subbarrier fusion cross sections calculated using the Wong approximation or using the CCFUS software package disregarding the channel coupling are going down much sharper and at an energy of  $E^* = 67.8$  MeV are by nearly an order of magnitude lower than the experimental value of fusion cross section whereas the accounting of channel coupling or of zero-point vibrations leads to the increase of the cross section. That is why, from our point of view, further measurements of the fusion-fission cross sections at yet lower energies are of great interest bearing in mind the fact that the sensitivity limit for the solid state detector method is  $\sigma \le 10^{-36}$  cm<sup>2</sup>.

For the analysis of the fission cross sections we have used a statistical model which, for the sake of universality, uses the minimum number of physical assumptions and parameters, which, naturally, makes the model somewhat coarser, but enables to make less

<sup>\*)</sup> The calculations are made by Shilov V.M.  $V_b = 75.4$  MeV,  $\Delta E = 3.6$  MeV,  $h\omega = 3.95$ 

<sup>\*\*)</sup> This work points, corrected on the evaporation cross section

ambiguous conclusions. The most important quantity in the calculations under the statistical model is the nuclear level density. In our calculations we are using the ratios of the Fermigas model (without taking into account the effect of collective enhancement) with the phenomenological consideration of shell effects  $(\Delta W_{\nu})$  in the level density parameter according to Ignatyuk [7].

$$a_{v}(E^{*}) + \tilde{a}_{v} \{1 + [1 - \exp(-0.054E^{*})] \Delta W_{v}(A, Z) / E^{*}\},$$

where  $\tilde{a}_v = (0.11A - 6.3 \cdot 10^{-5} \cdot A^2)$ ,  $E^*$  is the excitation energy of the compound nucleus and  $\Delta W_v(A, Z)$  is the nuclear shell mass correction after evaporation of the v-particle (neutron, proton or  $\alpha$ -particles). We consider the parameter of level density  $\tilde{a}_f$  in the fission channel to be constant, independent of the excitation energy and proportional to the asymptotic value of the level density parameter in the channel of  $\tilde{a}_v$ -particle evaporation (an assumption that the shell correction in the saddle point is negligibly small). The fission barriers for nuclei were calculated using the formula

$$B_f(l) = CB_f^{LD}(l) + \Delta W^{\exp},$$

where C is a free parameter;  $B_f^{LD}(l)$ , the fission barrier in the rotating liquid drop model CPS;  $\Delta W^{\rm exp}$ , a correction to the compound nucleus fission barrier equal to the shell correction of its ground state mass. In the fission barrier we are neglecting the small value of the shell correction in the saddle point.

The calculations of evaporation widths are based on the Weiskopf-Eving formalism

$$\Gamma_{v}^{l}(E) = \frac{(2l+1)(2s_{v}+1) m_{v}}{\pi^{2} \rho_{c}(E_{c})} \int_{0}^{E-E_{rot}(l)-E_{v}} \rho_{v}(E-E_{rot}(l)-E_{v}-\varepsilon) \varepsilon \sigma_{v}(\varepsilon) d\varepsilon,$$

where  $S_{\rm v}$ ,  $E_{\rm v}$  and  $m_{\rm v}$  are the spin, the binding energy and the reduced mass of the v particle;  $\sigma_{\rm v}(\epsilon)$  is the cross section of the inverse reaction of capture of the v particle with energy  $\epsilon$  calculated according to the optical model using the parameters suggested in Ref.[8]. The fission width is calculated using the classical Bohr and Willer formula

$$\Gamma_f^l(E) = \frac{2l+1}{2\pi\rho_c(E_c)} \int_0^{E-E_{\text{rot}}^{\text{sp}}(l)-B_f(l)} \rho_f(E-E_{\text{rot}}^{\text{sp}}(l)-B_f(l)-\varepsilon) d\varepsilon,$$

where  $E_{\text{rot}}^{\text{sp}}(l)$  is the rotation energy at the saddle point.

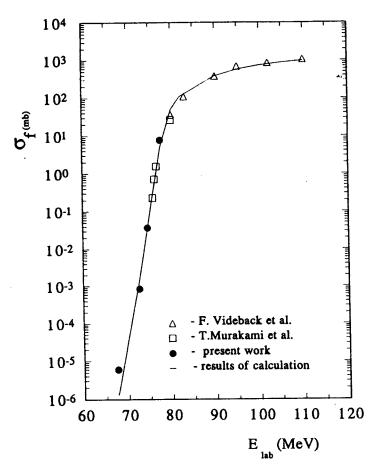


Fig.2. <sup>224</sup>Th fussion-fission excitation function: points — experimental data, solid line — results of calculation

Within such an approach the main parameters of the statistical calculations are the ratio of asymptotic values of the level density in the fission and evaporation channels  $\tilde{a}_f/\tilde{a}_v$  and a free parameter in the formula for the fission barrier, i.e., coefficient C.

The problem of choosing the  $\tilde{a}_f/\tilde{a}_v$  value was considered in a number of papers at the analysis of the deexcitation process in preactinide compound nuclei (for example, review [9]). In different models its value varies from 0.95 to 1.1, and all the papers note a weak dependence of  $\tilde{a}_f/\tilde{a}_v$  on the mass number. It should be noted also that within such an approach there was obtained earlier a good description of evaporation reaction cross sections and of fissilities for a wide range of compound nuclei from Bi to U using a fixed value of the parameter  $\tilde{a}_f/\tilde{a}_v=1.0$ . The parameter C for this compound nucleus  $^{224}$ Th is equal to C=0.7 and  $B_f(l=0)=5.7$  MeV.

Figure 2 presents experimental fission cross sections and results of calculations in coupled-channels approach. It is evident, that there is a satisfactory agreement of the cross sections through the whole of the region, and at energies exceeding 75 MeV the calculated yield of evaporation residues is in good agreement with those measured in Ref.[6]. In an interval of deexcitation energies of 20—30 MeV, the contributions from the two fission chances into the total cross section are approximately equal, which has to be taken into account at the analysis of different characteristics of the fission process.

#### Conclusion

There have been measured fission cross sections of  $^{224}$ Th compound nuclei in the reaction  $^{16}$ O  $\pm$   $^{208}$ Pb deep in the subbarrier energy region. It is evident that for ion energies of  $^{16}$ O  $\leq$  72 MeV the behaviour of the fission cross section deviates from the exponential drop, which testifies to the fact that structural effects have influence on the fusion cross section in the deep subbarrier case of the interaction of two spherical nuclei. The analysis performed made it possible to describe the fusion cross sections down to the lowest energies and to make certain statements on the necessity to consider in the calculation of the fusion cross sections the lowest vibration states of coupled nuclei. The sensitivity of the method enables one to go down in the cross section by two orders of magnitude to  $10^{-36}$  cm<sup>2</sup> which is by 12 orders of magnitude less than the geometric cross section of the nuclei interaction. The interest to further investigations in this energy region is explained by a possibility of obtaining additional information on the compound-states responsible for the cluster decay of nuclei.

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## References

- Itkis M.G. et al. In: Proc. 15 Nucl. Phys. Divisional Conf. (LEND-95), St.Petersburg (Russia), April 18-22, 1995, ed. by Yu.Oganessian, W. von Oertzen, R.Kalpakchieva. World Scientific. Singapore, p.177, 1995.
- 2. Wong C.Y. Phys.Rev.Lett., 1973, 31, p.766.
- 3. Esbensen H. Nucl. Phys., 1981, A352, p.147.
- 4. Shilov V.M. In: Proc 15 Nucl. Phys. Divisional Conf. (LEND-95), St.Petersburg (Russia), April 18-22, 1995, ed. by Yu.Oganessian, W. von Oertzen, R.Kalpakchieva. World Scientific. Singapore, p.460, 1995.

- 5. Dasso C.H., Landowne S. Comput. Phys. Commun., 1987, 46, p.187.
- 6. Morton et al. Phys.Rev., 1995, C52, p.243.
- 7. Ignatyuk A.V., Smirenkin G.N., Tischin A.S. Yadernaya Fizika, 1975, 21, p.485.
- 8. Perey C.M., Perey F.G. Atomic Data and Nucl. Data Tables, 1976, vol.17, p.2.
- 9. Andreyev A.N. et al. In: JINR Rapid Commun., 1995, No.4[72]-95, p.43.