

# INVESTIGATIONS OF THE ANGULAR DEPENDENCES OF NEUTRON-NEUTRON COINCIDENCES FROM $^{252}\text{Cf}$ , $^{235}\text{U}$ , $^{233}\text{U}$ , AND $^{239}\text{Pu}$ FISSION IN SEARCH OF SCISSION NEUTRONS

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## ABSTRACT

The aim of the researches of prompt neutrons angular distributions in fission is neutron emission mechanism investigation and estimation of scission neutrons contribution. The neutron-neutron (**n-n**) angular correlation experiment does not require the detection of fission fragments. The observations are automatically averaged over all orientations of the fission axis, but the correlation is sensitive to the characteristics of neutron emission. At first, we have measured (2006) in PNPI (Gatchina, Russia) the (n-n) angular correlations of prompt neutrons from  $^{252}\text{Cf}$  spontaneous fission for different angles and fission neutrons energy thresholds. The same measurements (2007-2009) from reactor thermal neutrons fission of  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  have been finished at the moment. From the measurements at angles of  $12.5^\circ$  to  $180^\circ$  between the directions of the fission prompt neutrons it was found that the number of coincidences has minimum near  $90^\circ$  and increases by about a factor  $\sim (1.5 - 3)$  to  $0^\circ$  and  $180^\circ$  for different fission neutrons energy thresholds  $\sim (400 - 2000)$  keV. In order to compare our results with predictions based on the evaporation model we have made Monte-Carlo calculations of our data. The scission neutrons contributions were estimated. Theoretical analysis with use of different thresholds for fission neutron energies the correlation could be adequately described by a simple evaporation model, assuming isotropic neutron emission from the fully accelerated fragments frames in their centre of mass together with  $\sim (5-15)\%$  isotropic scission component for different investigated heavy nuclei.

## 1. INTRODUCTION

A large fraction of the neutrons emitted promptly in thermal neutron fission can be accounted for in terms of evaporation from the fully accelerated fission fragments. However the origin of the remaining part  $\sim (10-25)\%$  of the neutron emission remains in question [1-7]. It has been suggested that these neutrons may be emitted at the instant of scission [8,9] or during the acceleration period of the fragments [10,11]. Models of the neutron emission rest mainly on experimental observation of the velocity and angular distributions of the prompt neutrons. The angular distributions being referred to the axis defined by the direction of the light fragment.

The neutron-neutron (n-n) angular correlation experiment does not require the detection of fission fragments. The observations are automatically averaged over all orientations of the fission axis. Nevertheless the correlation is sensitive to the characteristics of neutron emission and provides a useful additional method for testing models of emission process.

There are a few works [5, 12, 13] devoted to the investigations of the (n-n) angular correlation in nuclear fission. But there are some vagueness and questions in obtained results.

At first, we have measured the (n-n) – angular correlation of prompt neutrons from the spontaneous fission of  $^{252}\text{Cf}$  for different angles [14, 15, 17-20]. The measurements from thermal neutrons fission of  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  have been finished at the moment and results of the experimental researches are presented [14, 16, 18-20].

The aim of the researches is investigation of the neutron emission mechanism. In order to compare results with predictions based on the evaporation model we have made Monte-Carlo calculations simulating of all obtained data.

## **2. EXPERIMENT**

The (n-n) angular correlation researches with  $\sim 1$  mkg of spontaneously fissile  $^{252}\text{Cf}$  were performed (2006) in laboratory room. The measurements in  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  fission were carried out (2007-2009) with using a targets of  $\sim 1\text{g}$  arranged into Al cylinder containers. The (n-n) angular correlations measurements were performed at the thermal neutron beam ( $3 \times 10^6$  n/cm<sup>2</sup>·s) of the WWR-M Reactor of PNPI.

We used two identical photo-multiplier detectors [14, 17] with stilbene crystal scintillators measured the coincidences rate of pairs of fission neutrons (or  $\gamma$ -quanta, or n- $\gamma$ ) emitted at relative angles  $\theta$ . The range is from  $\sim 12.5^\circ$  (minimal possible) to  $180^\circ$  in  $2.5^\circ$  intervals. The different distances  $\sim (30-110)$  cm from the target to the two stilbene scintillators ( $\varnothing 60 \times 40$  mm) are used in the measurements. The detectors were into n, $\gamma$ -protection consisted of polyethylene and lead. Besides, TiH<sub>2</sub> scattered prompt neutrons screen between detectors was applied for small angles in order to be protected from neutron cross-talk. Magnetic screens are used also.

The main task of the experiment consists in comparison of the fission prompt neutron-neutron (n-n) coincidences rate from two detectors for different angles between them.

In this experiment we use technique of time-of-flight measurements [14, 17], where “start” signals are prompt neutron (or  $\gamma$ -quantum) ones from first detector, and “stops” signals are ones from other detector. In our experiment the gamma-ray events rejection was realized by neutron and gamma pulse shape discrimination [14, 17]. After sorting and analysis of all experimental coincidences spectra we obtain the separated contributions in total time spectrum for (n-n), (n- $\gamma$ ), and ( $\gamma$ - $\gamma$ ) coincidences [14, 17]. As result we obtain experimental angular dependences for all separated coincidences.

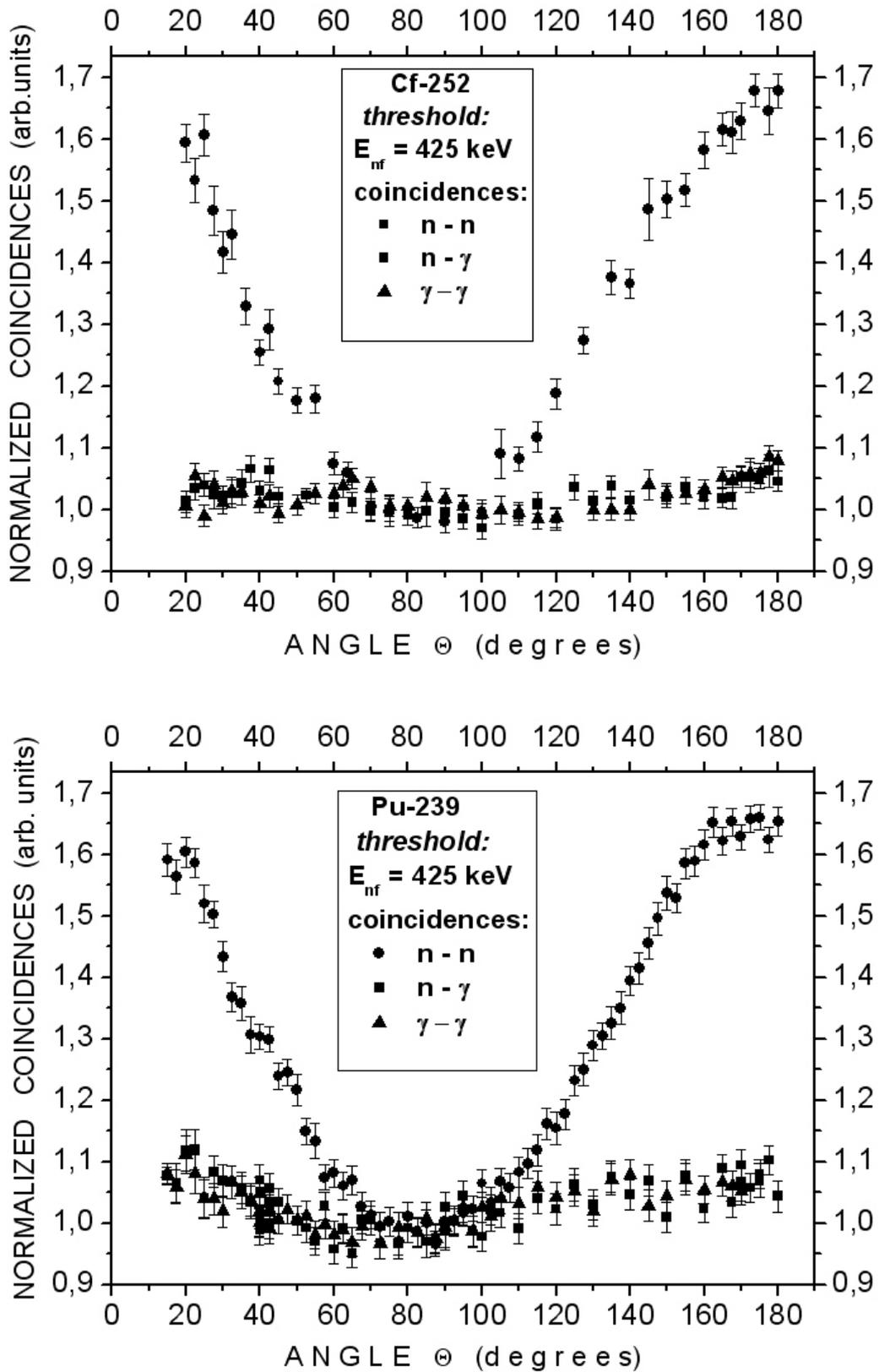
## **3. EXPERIMENTAL RESULTS**

### **3.1 Experimental angular dependences of (n-n), (n- $\gamma$ ), and ( $\gamma$ - $\gamma$ ) coincidences for threshold of prompt neutrons energy $E_{nf}=425$ keV in $^{252}\text{Cf}$ , $^{235}\text{U}$ , $^{233}\text{U}$ , and $^{239}\text{Pu}$ fission**

In Fig. 1 (above) the experimental angle dependences of (n-n), ( $\gamma$ - $\gamma$ ), and (n- $\gamma$ ) coincidences in  $^{252}\text{Cf}$  fission for prompt neutrons having energies higher then about **425 keV** (minimal experimental threshold [14, 15, 17]) are presented. All coincidences in  $^{252}\text{Cf}$  fission were normalized to count rates in two detectors. Besides, the minimums of the normalized coincidences dependences in the arbitrary units are located near one for convenience in the observation. Presented errors contain statistical and accidental ones.

From the measurements at angles of  $20^\circ$  to  $180^\circ$  between the directions of the fission neutrons it was found that the number of (n-n) coincidences in  $^{252}\text{Cf}$  has minimum near  $90^\circ$  and increases by a factor about **1.65** to near  $0^\circ$  and  $180^\circ$ .

For (n- $\gamma$ ) and ( $\gamma$ - $\gamma$ ) coincidences angular dependences some clear dependence practically is absent in comparison with (n-n) coincidences in limit of errors (see Fig.1). But the dependences are similar with (n-n) one. The ratio factor of (n- $\gamma$ ), ( $\gamma$ - $\gamma$ ) coincidences maximum (near  $0^\circ$  and  $180^\circ$ ) to minimum (near  $90^\circ$ ) is about **(1.05 - 1.10)** and it is not



**Fig. 1.** The experimental angle dependence of (n-n), (n- $\gamma$ ), and ( $\gamma$ - $\gamma$ ) coincidences in  $^{252}\text{Cf}$  (above) and  $^{239}\text{Pu}$  (below) fission for prompt fission neutrons having energies higher than about 425 keV.

depended from prompt neutrons and gamma quanta energies thresholds as the analysis of the obtained results for all investigated nuclei showed.

In **Fig. 1** (below) it can be seen the comparative angular dependences of the (n-n), (n- $\gamma$ ), and ( $\gamma$ - $\gamma$ ) coincidences in  $^{239}\text{Pu}$  for the same lower threshold  $E_{\text{nf}} = 425 \text{ keV}$ . It is necessary to note that all coincidences obtained at the reactor beam were normalized to neutron monitor count rates. We observe analogous dependences. Number of (n-n) coincidences has minimum near  $90^\circ$  also and increases by factor  $\sim 1.65$  to near  $0^\circ$  and  $180^\circ$ . It is similar with  $^{252}\text{Cf}$ .

As before, for the (n- $\gamma$ ) and ( $\gamma$ - $\gamma$ ) coincidences such angular dependences are very small and the factor is equal  $\sim 1.1$ . Only in case of ( $\gamma$ - $\gamma$ ) coincidences dependence in  $^{239}\text{Pu}$  (and in  $^{235}\text{U}$ ,  $^{233}\text{U}$  also) we observe their sharp increasing near  $180^\circ$  connected with annihilation gamma-quanta, but these experimental points are not given here for the nuclei.

Analogous dependences for  $^{235}\text{U}$  and  $^{233}\text{U}$  are presented in **Fig. 2** for the same threshold  $425 \text{ keV}$ . The number of (n-n) coincidences has minimum near  $90^\circ$  and increases by factor  $\sim 1.85$  to near  $0^\circ$  and  $180^\circ$ . For the (n- $\gamma$ ) and ( $\gamma$ - $\gamma$ ) coincidences this angular dependence is very small (the factor  $\sim 1.1$ ) in the comparison with (n-n) coincidences one.

### 3.2 Theoretical approach to description of the experimental results

The Monte Carlo simulation [15-17] based on a simple evaporation model with some admixture of the other possible mechanisms of neutron emission was used for theoretical description of the experimental data for the  $^{252}\text{Cf}$ ,  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$ :

At the first stage of this calculations it was simulated anisotropical neutron emission with individual multiplicity from both fragments with the addition of the necessary isotropical contribution of scission neutrons in laboratory system. The averaged total multiplicity of emitted neutrons was taken into account.

The actual number of neutrons evaporated by each fragments was chosen randomly by two-dimensional Gaussian distribution with known ratio of fragment multiplicities and experimentally defined covariance. We have got this ratio and neutron spectra in fragment centre-of-mass due to neutron-fragment experiment. It was taken into account the presence of neutron emission anisotropy concerned with the angular momentum of each fragment. In process of calculation were used final velocities for light and heavy fission fragments with the most probable masses.

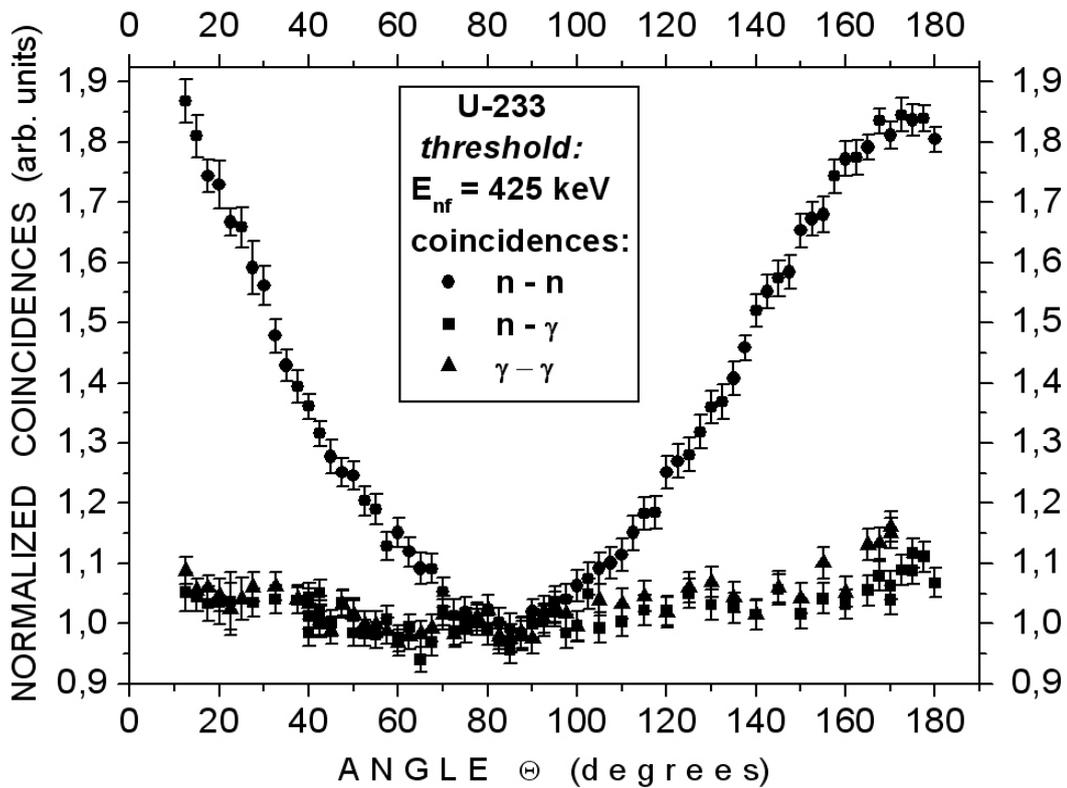
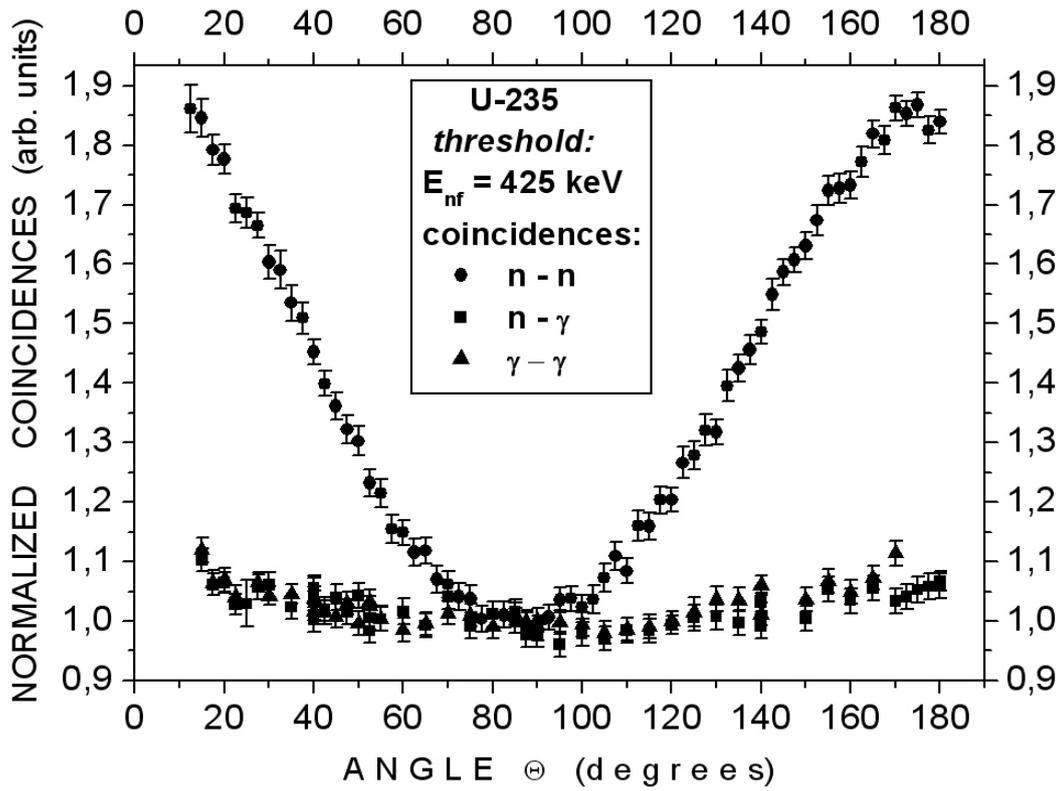
The neutron spectrum of each fragment was assumed to be Maxwellian form with fixed temperature for light and heavy fragments  $T_{L,H} \sim 1 \text{ MeV}$  in fragment system:

$$N(E) = \left( E^{1/2} / T^{3/2} \right) \cdot \exp(-E/T) .$$

In process of calculation were used final velocities of light and heavy fission fragments with the most probable masses. The Weisskopf distribution for scission neutron spectrum was assumed:

$$N(E) = \left( E/T^2 \right) \cdot \exp(-E/T) .$$

Thus we had two free parameters (the contribution of scission neutrons and temperature) to fit experimental data of (n-n) angular distributions measured with six different thresholds of fission neutrons energy. The detailed theoretical analysis [15-17] and interpretation of the results was made.



**Fig. 2.** The experimental angle dependence of (n-n), (n- $\gamma$ ), and ( $\gamma$ - $\gamma$ ) coincidences in  $^{235}\text{U}$  (above) and  $^{233}\text{U}$  (below) fission for prompt fission neutrons having energies higher than about 425 keV.

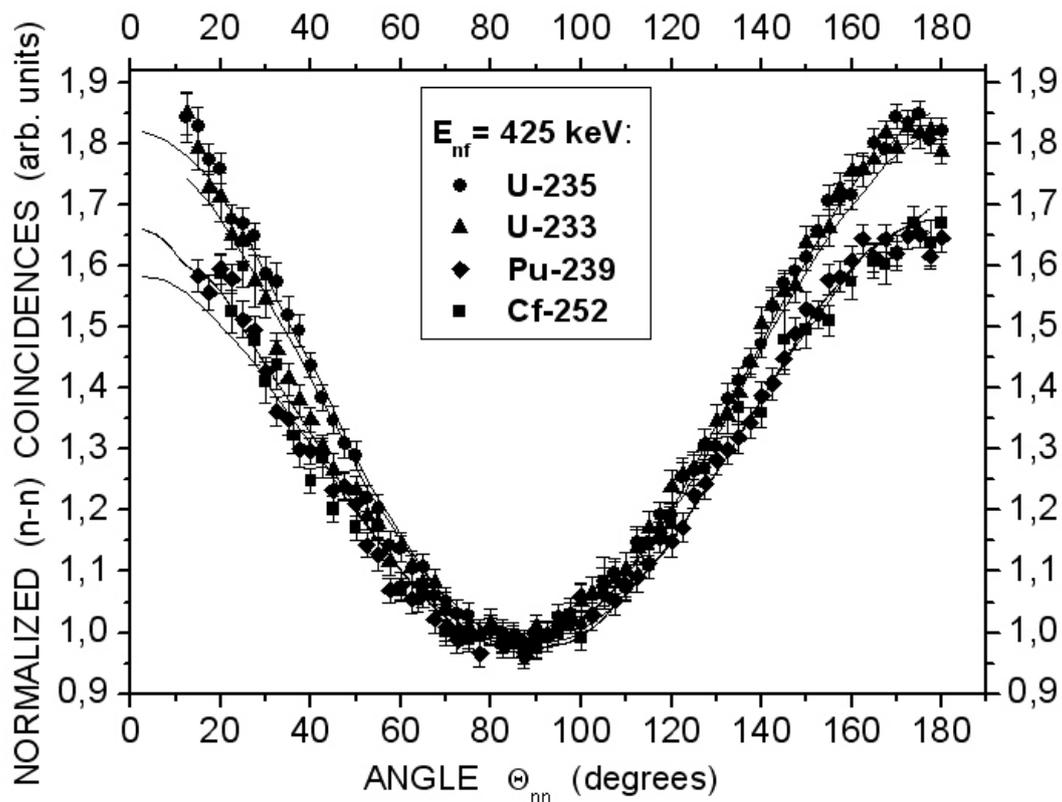
### 3.3 Comparison of (n-n) coincidences angular dependences for the investigated nuclei (threshold $E_{nf} = 425$ keV)

The comparison of (n-n) coincidences angular dependences for the investigated nuclei with minimal threshold  $E_{nf} = 425$  keV are presented in **Fig. 3**. Experimental points and theoretical curves as the results of the Monte-Carlo simulations are presented. In general we observe highly satisfactory description of the experimental data.

The angular dependences of (n-n) coincidences for  $^{235}\text{U}$  and  $^{233}\text{U}$  are similar. The same can be said about pair of  $^{252}\text{Cf}$  and  $^{239}\text{Pu}$ , but it is more plane with lesser inclination.

As already mentioned, the ratio factor of (n-n) coincidences of maximum to minimum is about **1.85** for both uraniums ( $^{235}\text{U}$ ,  $^{233}\text{U}$ ) and it is different for other pair ( $^{252}\text{Cf}$  and  $^{239}\text{Pu}$ ), as it was noticed above. For these nuclei the ratio are some less and it is equal about **1.65**.

It is necessary to mention systematic excess of the experimental count rates at small angles ( $20^\circ$ - $30^\circ$ ) for  $^{252}\text{Cf}$  and deficiency ( $30^\circ$ - $50^\circ$ ) for  $^{233}\text{U}$  in case of the low thresholds. A reason for a slight discrepancy between experimental and calculated data can be some inaccuracy of the theoretical calculation model and may be some difficulties of the measurements in the angular region. For example, the small fission neutrons cross-talk and scattering from the elements of radioactive protection of the detectors are possible. Besides, it is necessary to change and make longer distances from the target to detectors at small angles between them.



**Fig. 3.** Comparison of (n-n) coincidences angular dependences for the investigated nuclei  $^{252}\text{Cf}$ ,  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  (minimal experimental threshold  $E_{nf} = 425$  keV). The curves are result of theoretical interpretation and analysis of the experimental data.

### 3.4 Comparison of (n-n) coincidences angular dependences for the different prompt neutron energy thresholds in $^{252}\text{Cf}$ , $^{235}\text{U}$ , $^{233}\text{U}$ , and $^{239}\text{Pu}$ fission

Comparison of (n-n) coincidences angular dependences for the different prompt neutron energy thresholds  $\sim (400-2000)$  keV in  $^{252}\text{Cf}$ ,  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  fission are shown in **Figs. 4,5** [18-20].

In **Fig. 4** (above) the comparative angular dependences of (n-n) coincidences in  $^{252}\text{Cf}$  are shown for different more high thresholds of prompt neutrons energies: 425 keV, 550 keV, 800 keV, 1200 keV, and 1600 keV [17]. With rise of the neutron energy threshold the ratio of (n-n) coincidences maximum to minimum is increasing up to factor about **2.5** for the very high threshold  $E_{\text{nf}} = 1600$  keV.

The (n-n) correlation in  $^{252}\text{Cf}$  fission could be adequately described by a simple evaporation model, assuming anisotropic neutron emission from the fully accelerated fragments frames in their centre of mass together with **8 %** of isotropic scission component (see theoretical curves in the **Fig. 4**, above). This value is somewhat different from the result presented in [15, 17, 18-20].  $^{252}\text{Cf}$  was the first element for our treatment of (n-n) correlations and its investigation stood slightly out from subsequent elements. In process of new recalculation the energy depended anisotropy of neutron emission in fragment centre-of mass instead of averaged value was taken into account. Also the simulation of randomly choice for neutron multiplicity was performed more correctly.

Let us note that with an increase in the energy threshold the deviation of experimental points from the theoretical curve in the region of small angles disappears. Possibly, this speaks about the presence of the small cross-talk with the low thresholds connected with rescattering of the fission neutrons since first  $^{252}\text{Cf}$  measurements were performed without  $\text{TiH}_2$  screens.

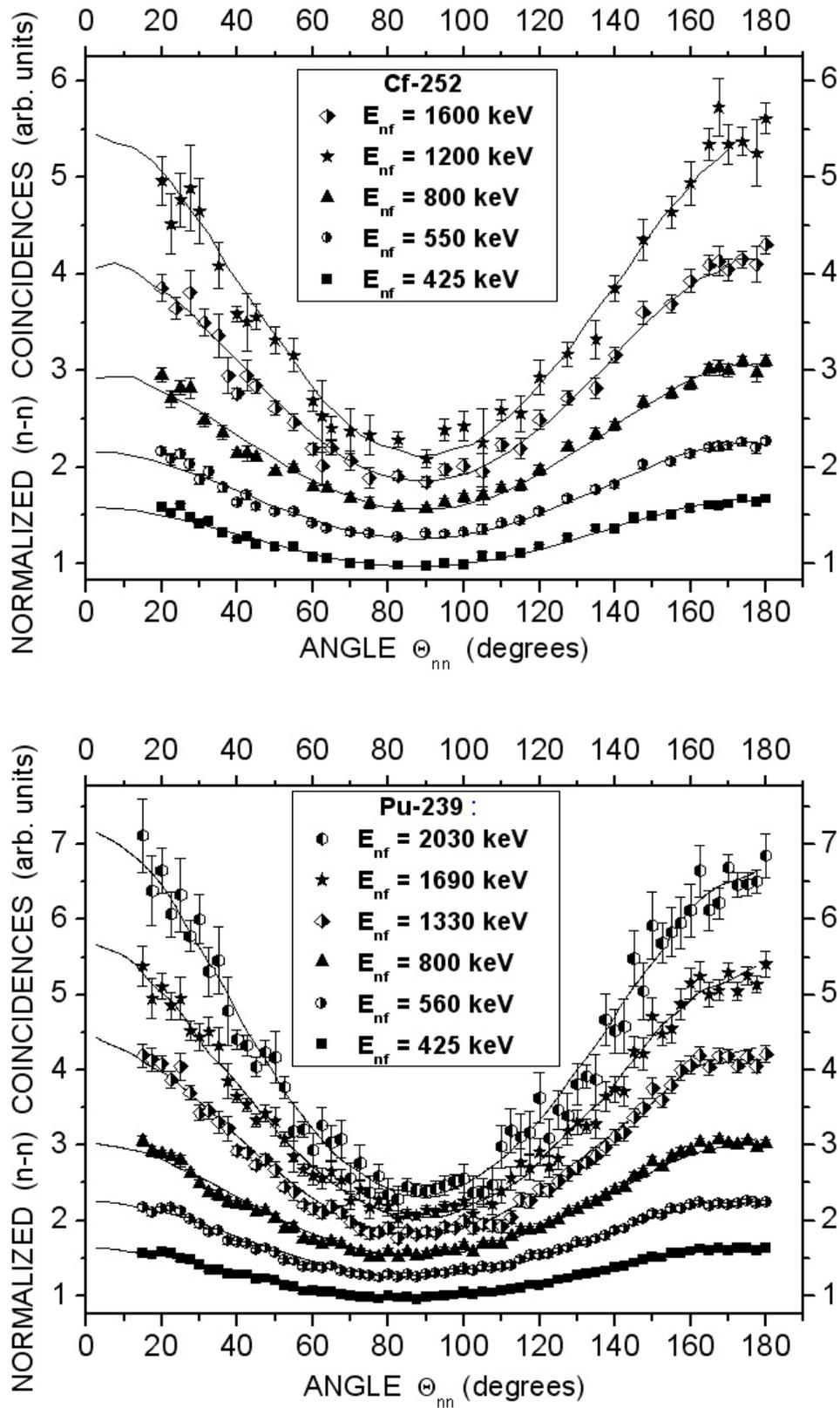
Comparison of (n-n) coincidences angular dependences for the different prompt neutron energy thresholds in  $^{239}\text{Pu}$  fission are presented in **Fig. 4** (below). With rise of the energy threshold the ratio is increasing up to  $\sim$  **2.6** for the most high threshold. The (n-n) correlation in  $^{239}\text{Pu}$  fission could be very good described for all 6 thresholds simultaneously by a simple evaporation model with  $\sim$  **14 %** scission component.

For (n-n) coincidences in  $^{235}\text{U}$  and  $^{233}\text{U}$  (**Fig. 5**) with raising of the thresholds it is observed rise of the ratio of maximum to minimum up to  $\sim$  **3** for maximal threshold  $\sim$  2000 keV. The curves are result of our theoretical interpretation and analysis of the experimental data together with  $\sim$  **(5-7) %** isotropic scission component simultaneously for all six the thresholds from  $\sim$  400 keV up to  $\sim$  2000 keV. We can see that in general the curves are in good accordance with experimental points.

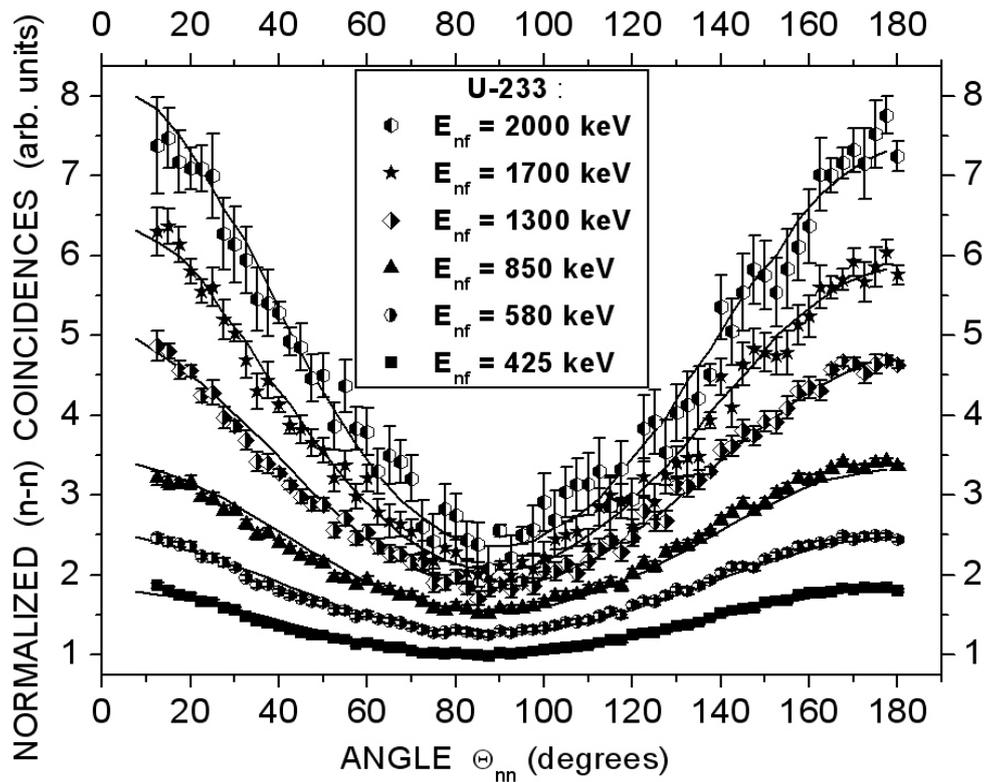
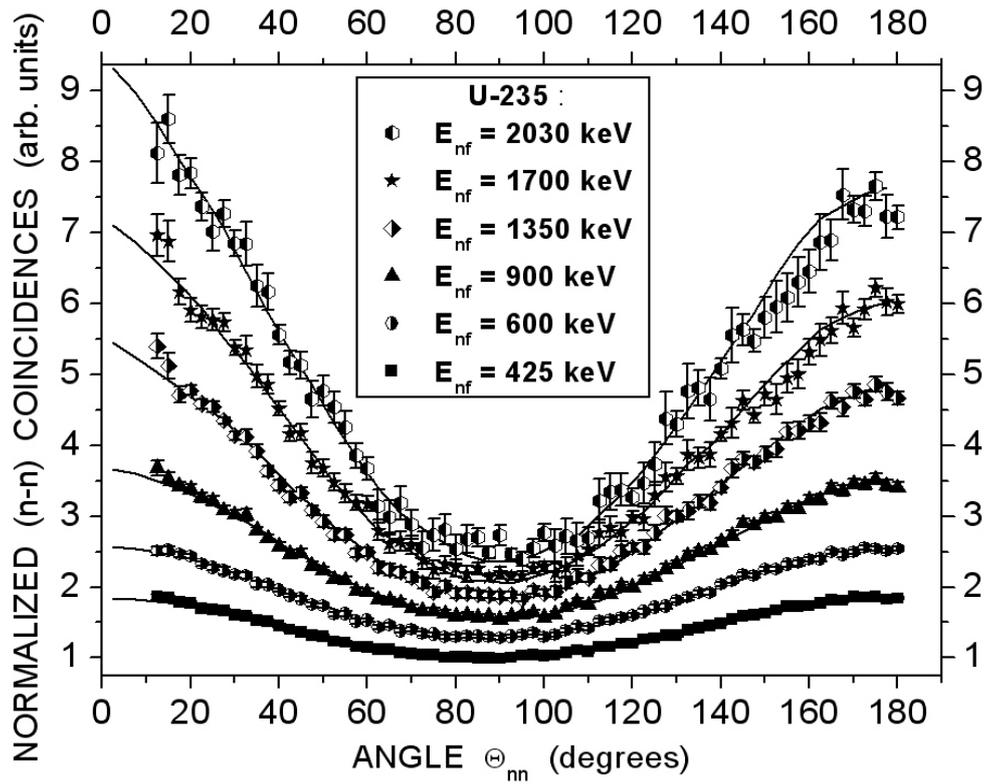
## 4. CONCLUSIONS

The angular dependences of neutron-neutron coincidences rate in fission of  $^{252}\text{Cf}$ ,  $^{235}\text{U}$ ,  $^{233}\text{U}$ , and  $^{239}\text{Pu}$  from our experiments were compared with the results of calculations based on the Monte-Carlo method for different neutron registration thresholds in the range  $\sim(400\div 2000)$  keV [14-20].

The main part of fission neutrons are prompt neutrons, emitted anisotropically from accelerated fragments after scission of nucleus. But from the comparison and theoretical analysis it was concluded that  $\sim$  **(5-15) %** of total number of neutrons in the nuclei are emitted isotropically (in laboratory system) and probably can be attributed as “scission neutrons”, arising just at rupture moment.



**Fig. 4.** The comparative experimental angular dependences of (n-n) coincidences in  $^{252}\text{Cf}$  (above) and  $^{239}\text{Pu}$  (below) for different thresholds of prompt fission neutrons energies. The curves are result of theoretical interpretation and analysis of the experimental data .



**Fig. 5.** The comparative experimental angular dependences of (n-n) coincidences in  $^{235}\text{U}$  (above) and  $^{233}\text{U}$  (below) for six different thresholds of prompt fission neutrons energies. The curves are result of theoretical interpretation and analysis of the experimental data .

The presented analysis has allowed also to obtain energy distribution of this isotropic component.

Next estimations are obtained for **scission neutrons yields** and their temperatures  $T_{sc}$  of the spectrums :

- $^{252}\text{Cf}$  :  $(8 \pm 3) \%$  ,  $T_{sc} = (1.0 \pm 0.1) \text{ MeV}$  ,
- $^{235}\text{U}$  :  $(7 \pm 2) \%$  ,  $T_{sc} = (1.0 \pm 0.1) \text{ MeV}$  ,
- $^{233}\text{U}$  :  $(5 \pm 3) \%$  ,  $T_{sc} = (1.1 \pm 0.1) \text{ MeV}$  ,
- $^{239}\text{Pu}$  :  $(14 \pm 3) \%$  ,  $T_{sc} = (0.9 \pm 0.1) \text{ MeV}$  .

All the pointed errors are statistical ones only.

For obtaining of more reliable results for estimations of scission neutrons contribution and their spectrum form in fission of heavy nuclei it is desirable to analyze this data together with results [21], obtained by my Gatchina colleagues A.S.Vorobyev and O.A.Shcherbakov at the WWR-M Reactor in PNPI (Gatchina), when their measurements will be finished.

The researches apply to fission prompt neutrons and light fragments directions angular correlations in these nuclei.

And finally, the represented cycle of our works on a study of the (n- n) coincidences angular dependences in fission of the heavy nuclei made a qualitative and quantitative step forward in comparison with researches before:

- the circle of the investigated nuclei is extended,
- accuracy and quality of our measurements are increased,
- quantitative estimations of the contribution of the scission neutrons are given.

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УЧРЕЖДЕНИЕ  
РОССИЙСКОЙ АКАДЕМИИ НАУК

ПЕТЕРБУРГСКИЙ ИНСТИТУТ  
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### РАЗРЕШЕНИЕ № 76

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