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A. V. Isaev* et al.

SPONTANEOUS FISSION OF ^{246}Fm

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* E-mail: isaev@jinr.ru

Исаев А. В. и др. Е7-2022-12

Спонтанное деление ^{246}Fm

На сепараторе SHELS завершился эксперимент по изучению спонтанного деления ^{246}Fm. Изотоп синтезировался в реакции полного слияния ионов пучка ^{40}Ar с ядрами мишени ^{208}Pb. С использованием детектирующей системы SFiNx были получены выходы мгновенных нейтронов спонтанного деления ^{246}Fm ($\overline{\nu} = 3{,}79 \pm 0{,}30$, $\sigma^2_\nu = 2{,}1$). Распределение испущенных нейтронов по множественностям восстановлено с помощью метода статистической регуляризации Тихонова ($\overline{\nu}_r = 3{,}79 \pm 0{,}20$, $\sigma^2_{\nu r} = 2{,}8$). Для изотопа были определены коэффициент ветвления по пути спонтанного деления ($b_{\rm SF} = 0{,}061 \pm 0{,}005$) и период полураспада ($T_{1/2} = 1{,}50^{+0{,}08}_{-0{,}07}$ с). Полученные в работе экспериментальные данные сравнивались с предсказаниями в рамках улучшенной модели точки разрыва. Наблюдается отличная сходимость по среднему числу нейтронов в акте деления, однако формы экспериментального и модельного распределений мгновенных нейтронов по множественностям существенно различаются.

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Spontaneous Fission of ^{246}Fm

An experiment on the study of the ^{246}Fm spontaneous fission was conducted using the SHELS separator. The isotope was synthesized in the complete fusion reaction of ^{40}Ar beam ions and ^{208}Pb target nuclei. The neutron yields of ^{246}Fm spontaneous fission ($\overline{\nu} = 3.79 \pm 0.30$, $\sigma^2_\nu = 2.1$) were obtained using the SFiNx detector system. The multiplicity distribution of emitted prompt neutrons was restored using the Tikhonov method of statistical regularisation ($\overline{\nu}_r = 3.79 \pm 0.20$, $\sigma^2_{\nu r} = 2.8$). The spontaneous fission branching ratio ($b_{\rm SF} = 0.061 \pm 0.005$) and the half-life ($T_{1/2} = 1.50^{+0.08}_{-0.07}$ s) of the isotope were determined. The experimental data were compared with scission point model predictions. Excellent convergence was observed in the average number of neutrons per spontaneous fission process. However, the forms of the experimental and model prompt neutron multiplicity distributions differ significantly.

The investigation has been performed at the Flerov Laboratory of Nuclear Reactions, JINR.

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A. V. Isaev[1], R. S. Mukhin[1], A. V. Andreev[1], M. A. Bychkov[1], M. L. Chelnokov[1], V. I. Chepigin[1], H. M. Devaraja[1], O. Dorvaux[2], M. Forge[2], B. Gall[2], K. Hauschild[3], I. N. Izosimov[1], K. Kessaci[2], A. A. Kuznetsova[1], A. Lopez-Martens[3], O. N. Malyshev[1,4], A. G. Popeko[1,4], Yu. A. Popov[1,4], A. Rahmatinejad[1], B. Sailaubekov[1,5,6], T. M. Shneidman[1,7], E. A. Sokol[1], A. I. Svirikhin[1,4], D. A. Testov[1], M. S. Tezekbayeva[1,5], A. V. Yeremin[1,4], N. I. Zamyatin[1], K. Sh. Zhumadilov[6]

[1] Joint Institute for Nuclear Research, Dubna
[2] Université de Strasbourg, CNRS, Strasbourg, France
[3] IJCLab, IN2P3-CNRS, Université Paris-Saclay, Orsay, France
[4] Dubna State University, Dubna, Russia
[5] Institute of Nuclear Physics, Almaty, Kazakhstan
[6] L. N. Gumilyov Eurasian National University, Nur-Sultan
[7] Kazan Federal University, Kazan, Russia

INTRODUCTION

Spontaneous fission is a common decay mode for heavy atomic nuclei and defines the limits of existence of chemical elements in this region. The mass and energy distributions of fission fragments and prompt neutron yields are important characteristics describing this process. However, the study of the properties of the heaviest nuclei is complicated by their short lifetimes and small formation cross sections, which requires the use of advanced experimental methods.

Data on prompt neutron yields from spontaneous fission were accumulated earlier for many isotopes with $Z < 100$, mainly in offline experiments. For fermium ($Z = 100$), such data were obtained both for neutron-rich isotopes ^{254,256,257}Fm [1–3] and neutron-deficient ones (online experiments) ^{244,246}Fm [4, 5]. The ^{246}Fm prompt γ-ray spectroscopy was performed at the University of Jyväskylä [6] and is the lowest cross-section reaction where this was possible for such heavy nuclei. The ^{246}Fm prompt neutron yields are the most poorly known because only 108 [5] fission events were obtained. Therefore, the purpose of the present work was to repeat the synthesis of ^{246}Fm in order to obtain more statistics and refine its prompt neutron yield data.

1. EXPERIMENT

An experiment aimed at studying the spontaneous fission properties of the ^{246}Fm isotope was carried out at FLNR JINR using the SHELS separator [7] and the SFiNx detection system [8] (Fig. 1). The system includes 116 ^{3}He-neutron counters that allow registering multiple prompt neutrons emitted in the spontaneous fission process of the nucleus, as well as an assembly (“well” like) of double-sided Si detectors with a 128×128-strip focal-plane detector and 8 tunnel 16×16-strip detectors for fission-fragment and α-particle registration (Fig. 2). The high granularity of the SFiNx neutron detector makes it possible to register multiple neutron events with a negligible probability that a single ^{3}He counter will detect several neutrons simultaneously within the coincidence time window [8]. The neutron registration efficiency, measured with a ^{248}Cm source, is (54.8 ± 0.1)%, and the average neutron lifetime in the array is (18.4 ± 0.2) μs. The detection efficiency of the focal-plane Si detector for α particles emitted by implanted nuclei is ~ 50% and 100% for at least one of the two fission fragments.

Fig. 1. Photograph of the SFiNx detector system at the focal plane of the SHELS

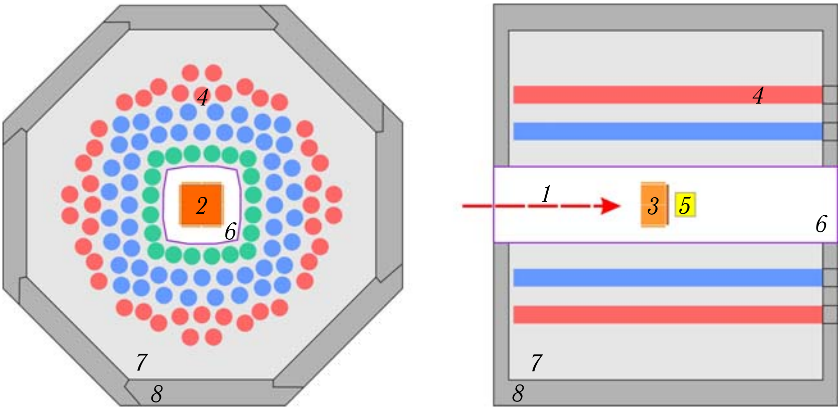

Fig. 2. The SFiNx scheme (front view — left; side view — right): *1* — recoil nuclei; *2* — focal-plane Si detector; *3* — tunnel Si-detector array; *4* — ^{3}He counters; *5* — scintillator detector; *6* — vacuum chamber; *7* — moderator; *8* — shield

The complete fusion reaction of $^{40}Ar^{16+}$ ions from the U-400 cyclotron with ^{208}Pb target nuclei was used to synthesize ^{246}Fm. The PbS target thickness was 450 μg/cm^2 (the ^{208}Pb isotope enrichment was over 99%), and a 1.5 μm thick titanium backing was used. The ion beam energy was

(183 ± 3) MeV and it was chosen close to the maximum of the excitation function of the $2n$-evaporation channel [5, 6, 9]. The total number of beam ions that passed through the target and then stopped in a Faraday cup was about $1.7 \cdot 10^{18}$.

Since ^{206}Pb impurities in the target ($< 1\%$) can lead to the production of the spontaneously fissile ^{244}Fm with $T_{1/2} \sim 3$ ms, fission fragments from ^{246}Fm were searched for in the time interval of 30 ms – 15.4 s following the registration of the implanted reaction products.

A total of 235 ^{246}Fm spontaneous fissions were found during data analysis, and only 2 events (in the interval of 0–30 ms) were assigned to the spontaneous fission of ^{244}Fm. The ^{246}Fm half-life was obtained as $T_{1/2} = 1.50^{+0.08}_{-0.07}$ s from the time distribution shown in Fig. 3. The value is in good agreement with the previously measured values of (1.54 ± 0.04) s [10], (1.3 ± 0.2) s [5] and (1.6 ± 0.2) s [6].

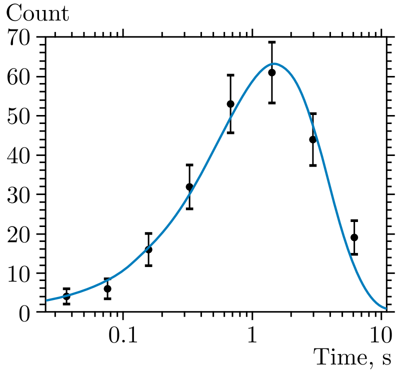

Fig. 3. Distribution of time differences between recoil nuclei and fission fragments registration for ^{246}Fm (dots) and fitting by method [11] (curve)

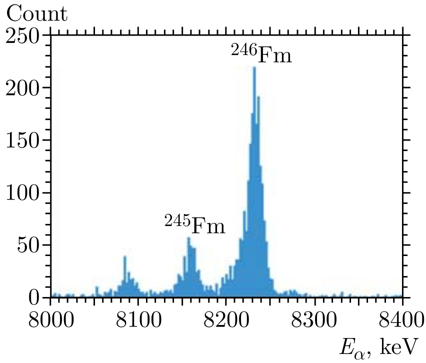

Fig. 4. Spectrum of α particles obtained in the search for correlations with implanted recoil nuclei

To determine the spontaneous fission branching ratio, α particles were searched for in the time interval of 30 ms – 15.4 s from the recoil implantation signal (Fig. 4). After background subtraction, 1809 α decays were found in the energy range of 8190–8260 keV. The spontaneous fission branching ratio was obtained as $b_{\rm SF} = 0.061 \pm 0.005$, which agrees with the previously published values of 0.068 ± 0.006 [10] and 0.05 ± 0.03 [5].

Prompt neutrons emitted in the spontaneous fission of fermium were searched for in the time interval of 0–128 μs from the moment of the fission-fragment registration. The neutron time distribution in the coincidence window is shown in Fig. 5. A total of 488 prompt neutrons from 235 ^{246}Fm spontaneous fission events were registered.

The prompt neutron multiplicity for ^{246}Fm extracted from the data is shown in Fig. 6. Taking into account the detector efficiency, the characteristics

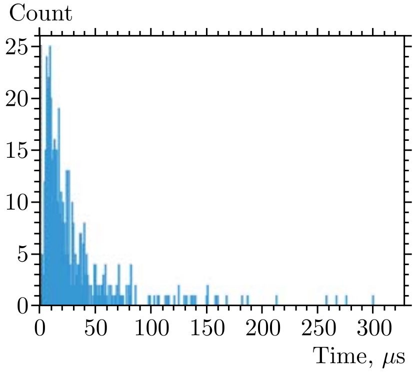

Fig. 5. Distribution of time differences between neutron detection and the spontaneous fission of ^{246}Fm

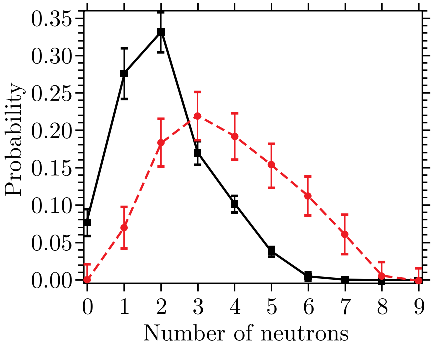

Fig. 6. Multiplicity distributions of the neutrons emitted in the spontaneous fission of ^{246}Fm: detected in the experiment (squares) and reconstructed (circles). The lines connecting points have been added for clarity

of the multiplicity distribution of the emitted neutrons were obtained: the mean as $\overline{\nu} = 3.79 \pm 0.30$ and the variance as $\sigma_\nu^2 = 2.1$. The obtained $\overline{\nu}$ value agrees well with 3.55 ± 0.50 already measured in Ref. [5].

The true form of the neutron multiplicity distribution was restored using Tikhonov's method of statistical regularization [12]. The reconstruction results are shown in Fig. 6 and in Table 1.

For the reconstructed distribution, the average number of neutrons is 3.79 ± 0.20 and the variance is 2.8.

An additional search was made for neutron events in the time interval of 200–328 μs from the ^{246}Fm spontaneous fission registration for background estimation. The interval duration was equal to the duration of the ^{246}Fm prompt neutrons search. The choice of the lower boundary of the search interval was based on the average neutron lifetime in the detector and set to eliminate the detection of fermium prompt neutrons. Only 5 background neutrons were detected (see Fig. 5) for 235 ^{246}Fm spontaneous fission events.

Table 1. **Prompt neutron emission probability for ^{246}Fm. The legend: n — number of neutrons; P_m — measured value; ΔP_m — error of measured value; P_r — reconstructed value; ΔP_r — error of reconstructed value**

n	P_m	ΔP_m	P_r	ΔP_r	n	P_m	ΔP_m	P_r	ΔP_r
0	0.077	0.018	0	0.021	5	0.038	0.006	0.154	0.030
1	0.277	0.034	0.070	0.028	6	0.004	0.002	0.113	0.027
2	0.332	0.027	0.184	0.033	7	0	<0.002	0.061	0.027
3	0.170	0.016	0.220	0.032	8	—	—	0.006	0.018
4	0.102	0.010	0.192	0.031	9	—	—	0	0.016

The influence of the background was insignificant in comparison with the level of statistical errors obtained in the experiment (the background to signal ratio is about 1%, whereas the relative statistical error is about 8%).

2. MODEL

The theoretical calculations of neutron multiplicity distributions were carried out with the improved scission point model [13]. The model assumes that the observed characteristics of the fission process are formed at the scission point, where the fissile nucleus can be represented as a superposition of systems consisting of two adjoining fragments (A_1, Z_1, β_1) and (A_2, Z_2, β_2). Assuming statistical equilibrium, the probability of realization of various binary configurations $G(A_i, Z_i, \beta_i, i = 1, 2)$ is determined by the potential energy as a function of masses A_i, charges Z_i, deformations β_i of the fragments [14] and excitation energy $U^\star$. For a particular scission configuration with fixed charge and mass numbers of the fragments, the potential energy as a function of the deformation parameters β_1 and β_2 can have one or several minima depending on the interplay of the macroscopic liquid-drop energy and microscopic shell effects. Calculations show that for the nuclei considered in this paper, the potential energy minimum occurs at deformations $\beta_i \gtrsim 2$. For such large deformations, the pocket in the interaction potential of the fragments disappears, and the system becomes unstable. Thus, the fissile nucleus reaches strongly deformed configurations with fewer probabilities than it follows from the assumption of statistical equilibrium. To account for this effect, the probabilities $G(A_i, Z_i, \beta_i)$ are multiplied by a factor

$$\prod_{i=1,2} \frac{1}{1 + \exp \dfrac{\beta_i - \beta_0}{s}}. \tag{1}$$

In this work, for all nuclei, the parameter values $\beta_0 = 1.7$ and $s = 0.08$ were used.

For each system with given masses, charges, and deformations of the fragments, the probability of n neutrons being emitted is calculated as

$$P(n) = \sum_{x=1}^{n} \int_{0}^{U^\star} F(U_1) P_1(x, U_1 + U_1^d) P_2(n - x, U_2 + U_2^d) dU_1, \tag{2}$$

where U_i and U_i^d $(i = 1, 2)$ are the excitation energy of the ith fragment available at the scission point and its deformation energy with respect to its ground state, respectively. The quantity $P_i(x, U_i + U_i^d)$ determines the probability that exactly x neutrons will be emitted from the ith fragment [15]. The probability that the excitation energy at the scission point will be distributed between the fragments as U_1 and U_2, $U_1 + U_2 = U^\star$, is given by the function

$$F(U_1) \sim \rho(a_1, U_1)\rho(a_2, U_2 = U^\star - U_1), \tag{3}$$

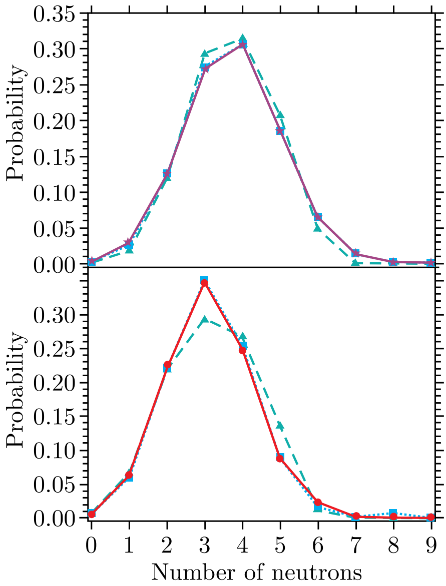

Fig. 7. Prompt neutron multiplicity distributions for ^{252}Cf (top) and ^{248}Cm (bottom). Symbols: triangles — calculations within the scission point model; squares — data from [17]; stars — data from [18]; circles — current work

where the level densities ρ of the fragments are taken in the form of a Fermi gas distribution with the level density parameters $a_i = A_i/12$ [16]. After separation, it is assumed that the deformation energy of the fragment is converted into its excitation energy.

Within the framework of this model, the distributions of prompt neutron multiplicities for ^{252}Cf and ^{248}Cm were calculated (Fig. 7). One can observe a reasonable agreement between the model predictions and experimental data for both the average neutron numbers and the shape of the distributions (Table 2).

In Fig. 8, the calculation results are presented for ^{246}Fm. The calculated average number of neutrons in the spontaneous fission process is 3.77, which agrees very well with the value measured in this experiment

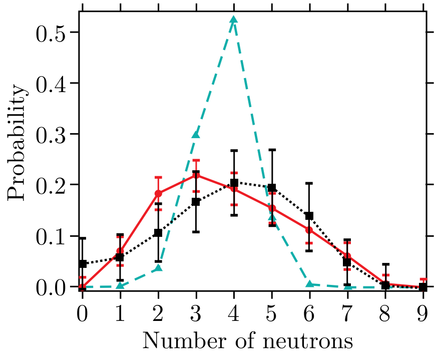

Fig. 8. Reconstructed prompt neutron multiplicity distributions for ^{246}Fm (circles — values obtained in this work; squares — values taken from [12]) and calculation made within the scission point model (triangles). The lines have been added to guide the eye

T a b l e 2. **Comparison of experimental and model prompt neutron multiplicity distributions for ^{248}Cm, ^{252}Cf and ^{246}Fm**

Isotope	Property	Experiment				Model prediction
		[17]	[18]	[12]	This work	
^{248}Cm	$\overline{\nu}$	3.13	3.13	—	3.13	3.21
	σ_ν^2	1.29	1.37	—	1.35	1.42
^{252}Cf	$\overline{\nu}$	3.76	3.76	—	—	3.72
	σ_ν^2	1.59	1.62	—	—	1.26
^{246}Fm	$\overline{\nu}$	—	—	3.9	3.79	3.77
	σ_ν^2	—	—	3.1	2.80	0.56

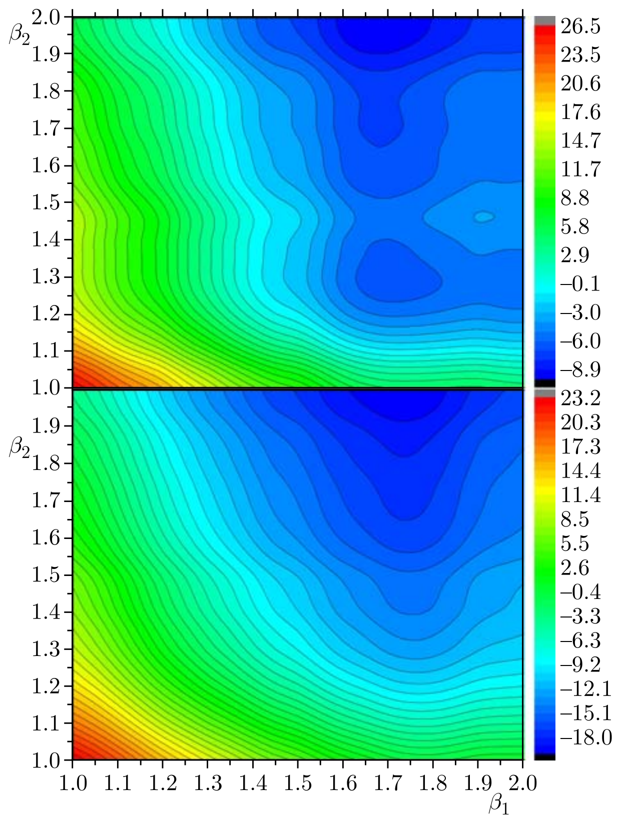

Fig. 9. Potential energy of scission configurations ^{108}Mo$\,+\,^{140}$Xe (top) and ^{102}Mo$\,+$ $+\,^{144}$Ce (bottom) as a function of the fragment deformations. These configurations have the maximal yields in the fission of ^{248}Cm and ^{246}Fm, respectively

3.79 ± 0.30. However, the calculated distribution for ^{246}Fm is narrower than the experimental one (the numerical parameters of the distributions are given in Table 2). The discrepancies between the theoretical and experimental distributions exceed the values of statistical errors.

A comparison of the theoretical distributions for ^{252}Cf, ^{248}Cm, and ^{246}Fm shows that for the latter there is a strong suppression of low multiplicity events. To understand this, a comparative analysis of the corresponding potential energy surfaces has been conducted (Fig. 9). We observed that for the most probable scission configurations of ^{252}Cf and ^{248}Cm, the potential energy as a function of the fragment's deformation has two minima: a more compact one ($\beta_1 \approx 1.7, \beta_2 \approx 1.3$) and a strongly deformed one ($\beta_1 \approx 1.7, \beta_2 \gtrsim 2$). The compact minimum leads to larger total kinetic energy of the fragments and a smaller number of emitted neutrons. In the statistical equilibrium assumption, the probability of realization of different deformations is split into two sizeable parts corresponding to these two minima. The use of the factor from Eq. (1) which accounts for deviations from statistical equilibrium increases the role of the compact minimum.

However, in the case of ^{246}Fm, the compact minimum in the potential energy as a function of deformation is absent. The probability distribution taken in the statistical equilibrium approach is fully concentrated in the region of large deformation. The compact systems related to a small number of emitted neutrons are realized with negligible probabilities. This cannot be significantly corrected by the factor from Eq. (1). Therefore, we can conclude that a more accurate account for the non-equilibrium effects is needed for ^{246}Fm. This work is currently in progress.

CONCLUSIONS

As a result of the experiment, the emission probabilities of ^{246}Fm prompt neutrons of different multiplicities were measured with the best accuracy (Fig. 8). The most accurate value of the average number of neutrons per spontaneous fission decay of ^{246}Fm is obtained: $\overline{\nu} = 3.79 \pm 0.30$.

Since the neutron detection efficiency of the SFiNx [8] is $(54.8 \pm 0.1)\%$ and the collected statistics is small (235 spontaneous fission events with 488 prompt neutrons in total), in order to obtain the true form of the prompt neutron multiplicity distribution, the use of the statistical regularization method is necessary. This procedure was successfully made for ^{246}Fm prompt neutrons data and, as a result, the true shape of the emitted neutron multiplicity distribution was obtained with the best precision.

Today there is no complete model of nuclear fission that could describe all the characteristics of the fission process for each specific nucleus. For the development of theoretical approaches, it is important to obtain new experimental data, especially for short-lived nuclei in the $Z \geqslant 100$ region. In this paper, we have used an improved scission point model [13], which made it possible to predict prompt neutron multiplicity distributions for ^{252}Cf, ^{248}Cm, and ^{246}Fm nuclei. The model perfectly describes the average number of neutrons in the fission processes of all the listed nuclei as well as the shapes of the neutron multiplicity distributions for ^{252}Cf and ^{248}Cm. For ^{246}Fm the width of the calculated distribution is significantly smaller than that of the experimental distribution. The latter indicates that a more sophisticated way to account for non-equilibrium effects is needed. This requires further theoretical developments, which are underway.

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141980, г. Дубна, Московская обл., ул. Жолио-Кюри, 6.
E-mail: publish@jinr.ru
www.jinr.ru/publish/