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SHELL EFFECTS AND THE FISSION
OF SUPERHEAVY NUCLEI AT LOW
EXCITATION ENERGIES

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Оболочечные эффекты и деление сверхтяжелых ядер при низких энергиях возбуждения

Представлены новые результаты исследований процессов деления и квазиделения сверхтяжелых ядер с $Z=102,112,114,118$, образованных в реакциях с ионами ^{48}Ca и ^{86}Kr . С помощью установки CORSET+DEMON были измерены распределения масс и энергий осколков деления, сечения деления компаунд-ядер, множественность пред- и постделительных нейтронов при энергиях вблизи и ниже кулоновского барьера. Впервые показано, что соотношение между сечением деления компаунд-ядра и сечением квазиделения резко изменяется при переходе от реакции $^{238}\text{U}+^{48}\text{Ca}$ к $^{244}\text{Pu}+^{48}\text{Ca}$, вследствие чего сечения деления σ_f для $^{292}114$ и $^{286}112$ примерно одинаковы, в то время как сечение квазиделения для $^{292}114$ в 6—8 раз меньше, чем для $^{286}112$. В случае реакции $^{208}\text{Pb}+^{86}\text{Kr}$, в отличие от реакций с ионами ^{48}Ca , вклад квазиделения доминирует и в области масс осколков, близких к $A/2$.

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Shell Effects and the Fission of Superheavy Nuclei at Low Excitation Energies

The work presents new results of the investigations of the fission and quasi-fission process of the superheavy nuclei with $Z=102,112,114,118$ produced in reactions with ions of ^{48}Ca and ^{86}Kr . Using the CORSET+DEMON set-up, the mass and energy distributions of the fission fragments, the fission cross sections of the compound nuclei, the multiplicity of the pre- and postfission neutrons at energies close to and below the Coulomb barrier have been measured. For the first time it has been shown that the ratio between the compound nucleus fission cross section and quasi-fission cross section changes drastically as one goes from the reaction $^{238}\text{U}+^{48}\text{Ca}$ to the reaction $^{244}\text{Pu}+^{48}\text{Ca}$. Owing to this the fission cross sections σ_f for $^{292}114$ and $^{286}112$ are approximately equal whereas the quasi-fission cross section for $^{292}114$ is 6—8 times smaller than that for $^{286}112$. In the case of the reaction $^{208}\text{Pb}+^{86}\text{Kr}$, as opposed to reactions with ^{48}Ca ions, the contribution of quasi-fission is also dominant in the region of fragment mass close to $A/2$.

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

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1 Introduction

Interest in the study of the fission process of superheavy nuclei in reactions with heavy ions is connected first of all with the possibility of obtaining information, the most important for the problem of synthesis, on the production cross section of compound nuclei at excitation energies of $\approx 15 - 30$ MeV (i.e. when the influence of shell effects on the fusion and characteristics of the decay of the composite system is considerable), which makes possible prediction on its basis of the probability of their survival after evaporating 1, 2 or 3 neutrons, i.e. in "cold" or "warm" fusion reactions. However, for this problem to be solved, there is a need for a much more penetrating insight into the fission mechanism of superheavy nuclei and for a knowledge of such fission characteristics as the fission-quasi-fission cross section ratio in relation to the ion-target entrance channel mass asymmetry and excitation energy, the multiplicity of the pre and postfission neutrons, the kinetic energy of the fragments and the peculiarities of the mass distributions of the fission and quasi-fission fragments etc. Undoubtedly all these points are of great independent interest to nuclear fission physics.

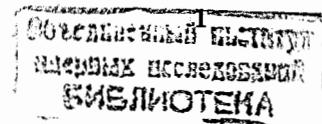
In this connection this work presents the first preliminary results of the experiments on the fission of superheavy nuclei in the reactions $^{208}\text{Pb} + ^{48}\text{Ca} \rightarrow ^{256}\text{No}$, $^{238}\text{U} + ^{48}\text{Ca} \rightarrow ^{286}112$, $^{244}\text{Pu} + ^{48}\text{Ca} \rightarrow ^{292}114$, $^{208}\text{Pb} + ^{86}\text{Kr} \rightarrow ^{294}118$ carried out at FLNR JINR in the last year. The choice of the indicated reactions has undoubtedly been inspired by the results of the recent experiments on producing the nuclides $^{283}112$, $^{287}114$, $^{289}114$ at Dubna^{1,2} and $^{293}118$ at Berkeley³ in the same reactions.

2 Experiment

The experiment was carried out on the extracted beam of ^{48}Ca and ^{86}Kr ions of the FLNR JINR U-400 accelerator, using a set-up that included:

- the two-armed time-of-flight reaction products spectrometer CORSET built with the use of microchannel plates (MChP);
- a 24 detector time-of-flight neutron spectrometer Demon using scintillation modules⁴;
- a 4 detector scintillation γ -quanta multiplicity spectrometer.

The reaction fragments spectrometer allows one to obtain the mass energy and angular distributions of fission fragments and scattered particles. It is based on 2 identical movable time-of-flight arms. The start detectors used MChP amplifiers equipped with an electrostatic mirror for the electrons knocked out by a particle passing through a 25×35 mm thin $120 \mu\text{g}/\text{cm}^2$ gilded mylar foil placed at 30 mm from the target.



As stop detectors, 4 coordinate sensitive MChP modules of 40×60 mm operating zone were used in each arm, the modules being placed at 180 mm from the target. The detectors were installed symmetrically relative to the beam at an angle of about 60° depending on the reaction type.

The spectrometer was calibrated and adjusted with the use of ^{226}Ra α -particle sources, the fission fragments of ^{252}Cf and elastic scattering peaks directly during the experiment. The following values were obtained for its main characteristics:

Time resolution	≤ 150 ps
Measuring range of the angles of divergence of reaction products	
- in the reaction plane Θ	$\pm 20^\circ$
- beyond the Ψ -plane	$\pm 15^\circ$
Angle measuring accuracy	0.1°
Solid angle of one arm	300 mst
Mass resolution	3 amu

The γ -quanta multiplicity spectrometer consisted of four 63×63 mm NaJ(Tl) detectors, which were placed in lead collimators and installed in the lower hemisphere at 35 cm from the target. The γ -quanta registration threshold was 100 keV. To reduce the number of accidental coincidences, the times were measured from the start pulse due to fission fragments to the pulses from the γ -detectors. The time resolution was 7 ns.

The neutron spectrometer detectors (built with the use of a Ne213 liquid scintillator) arranged in groups of three were located at 130 cm from the target at angles of 17° , 0° , -17° beyond the reaction plane and at angles of 45° , 65° , 155° , -65° , -90° , -135° in the reaction plane. Some detectors were located in the vertical plane along the beam axis at angles of 10° , 25° , 74.5° , 90° , 105.5° , 163° . The signals due to neutrons and γ -radiation were separated by analysing the shape of the pulse generated by the photomultiplier.

The discrimination thresholds were set during calibrating the detectors with γ -sources of ^{22}Na , ^{88}Y , ^{60}Co , ^{137}Cs , ^{226}Ra . To ensure the stability of the energy scale, the calibration was repeated several times during the experiment.

As the target, $120 - 170 \mu\text{g}/\text{cm}^2$ ^{208}Pb , ^{238}U and ^{244}Pu spectrometric layers were used deposited on a $40 - 50 \mu\text{g}/\text{cm}^2$ carbon backing.

The ion beam energy was periodically measured with an accuracy of ± 1 MeV with use of a semiconductor detector, using its scattering by a tantalum foil, and constantly monitored with a semiconductor monitor immediately in the reaction chamber, using the scattering of the beam nuclei by the target. The total ion current that had passed through the target was measured with the help of a Faraday cup, taking account of the dead time of the collecting system and its rest time.

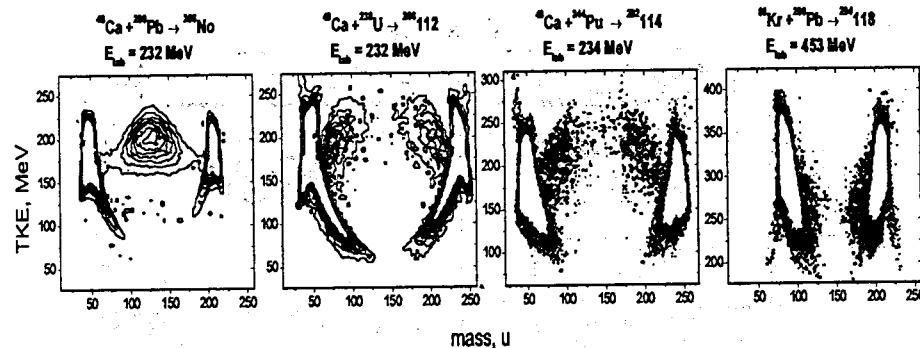


Figure 1: Two-dimensional matrices TKE–Mass of the products of the indicated reactions.

The data processing was carried out with the use of traditional techniques, assuming the process under study to be a two body process. The losses of the energy of the fragments in the target, backings and start detectors were taken account of. Special attention was paid to the angular folding correlations of the fragments both in and beyond the reaction plane. Only those events were selected and then analysed that corresponded to the two body process of complete momentum transfer.

3 Results and discussion

Fig. 1 shows the TKE–M two-dimensional matrices for the studied reactions at the energy of ^{48}Ca ions $E_{\text{lab}} = 233$ MeV and ^{86}Kr ions $E_{\text{lab}} = 453$ MeV, which corresponds to the excitation energy of compound nuclei of ^{256}No , $^{286}112$ and $^{292}114$ $E^* \approx 33$ MeV, and of compound nuclei of $^{294}118$ $E^* \approx 15$ MeV. It is clearly seen that the form of the TKE–M matrix between the elastic scattering peaks changes drastically as one goes from ^{256}No to the superheavy nuclei.

For ^{256}No it is of a triangular form characteristic of compound nucleus fission; and only at its edges the contribution is seen of events that can be considered as quasi-fission. As one goes to the $^{286}112$ nucleus, the quasi-fission process becomes distinctly dominant in the TKE–M matrix, the maximum yield of fragments lying around mass 208 and additional to it. For the $^{292}114$ nucleus the picture again changes, it is seen that the intensity of the quasi-fission peaks in relation to the yield of fragments in the symmetric mass region differs essentially from the similar ratio for the $^{286}112$ nucleus. This tendency is more clearly seen from the lower part of Fig.2, which shows the yields $Y(M)$ for the two reactions $^{48}\text{Ca} + ^{238}\text{U}$ and $^{48}\text{Ca} + ^{244}\text{Pu}$, which correspond to an approximately similar fluence of ^{48}Ca ions at the target. The obtained ratio

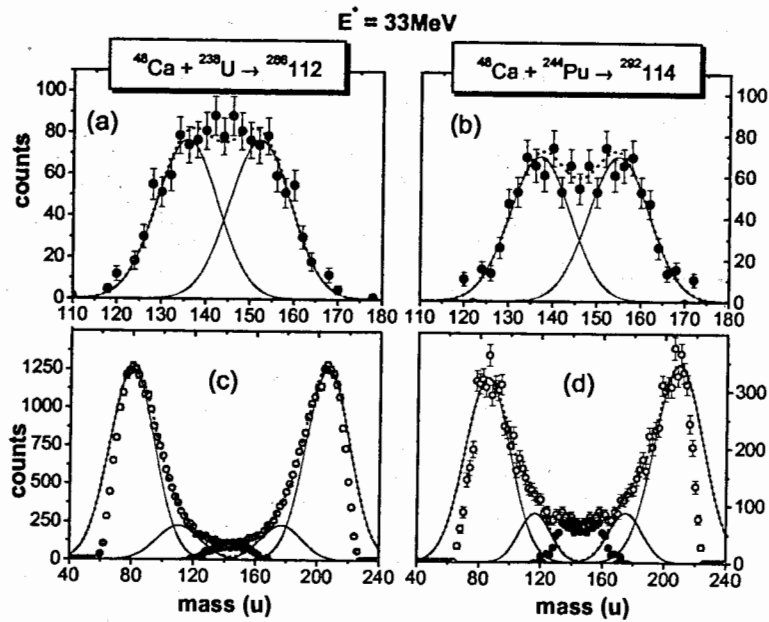


Figure 2: At the bottom: mass distributions of the reaction products. The experimental data are shown by open points; solid points represent the extracted components, corresponding to the compound nucleus (CN) fission. At the top: the extracted components of CN fission and their description by a sum of two Gaussians.

between the intensities of the quasi-fission peaks for these two reactions is $\approx 6 - 8$ in favour of $^{48}\text{Ca} + ^{238}\text{U}$. At the same time, as seen from the upper part of Fig.2, the distributions $Y(M)$ in the symmetric mass region ($A/2 \pm 25$), derived using the same technique and corresponding, in our opinion, to the fission of $^{286}\text{112}$ and $^{292}\text{114}$ compound nuclei, differ little from each other in shape, the intensity of the maximum and the area of the distributions. Hence it follows that the fission cross section for compound nuclei of $^{286}\text{112}$ and $^{292}\text{114}$ (and consequently the cross section for the complete fusion and production of compound nuclei) are approximately equal, whereas the quasi-fission cross section differ by a factor of 6 – 8. This appears to be connected with the fact that, on one hand, an increase in the values of $Z_i \times Z_{\text{targ}}$ on the whole results in the fusion cross section σ_0 being decreased and, on the other hand, great mass asymmetry in the reaction entrance channel is favourable for the cross section of the complete fusion and production of a compound nucleus being increased. Another important consequence of these data is that the asymmetric shape of the mass distribution has been observed. In this case, as opposed to the case

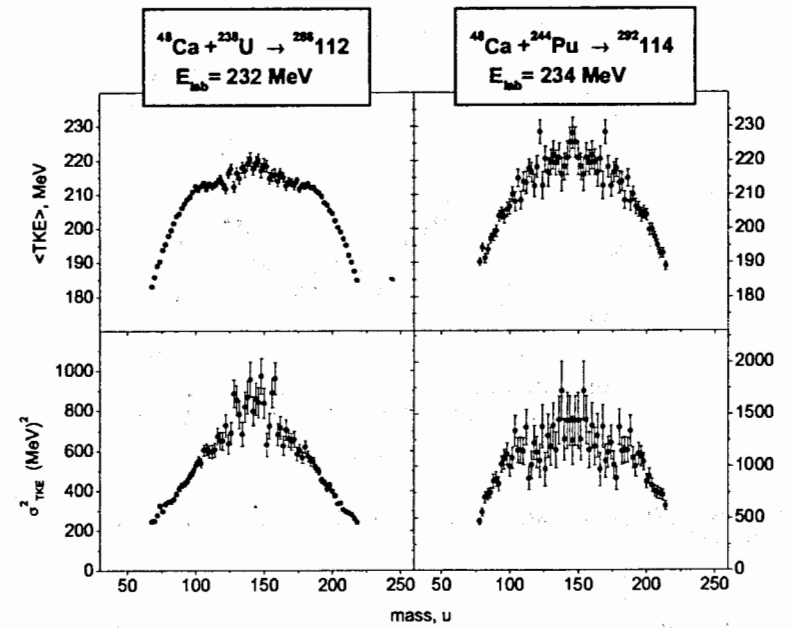


Figure 3: TKE and σ_{TKE}^2 distributions in dependence on the fragment mass for the same reactions as in Fig.2.

of actinide nuclei, the stabilizing role appears to be played by the nearspherical shell of the light fragment of mass 130 – 134, but not by the deformed shell of the heavy fragment of mass ≈ 140 .

Fig. 3 presents the characteristics of the fragment energy distributions TKE(M) and $\sigma_{\text{TKE}}^2(M)$ for $^{286}\text{112}$ and $^{292}\text{114}$. A distinguishing feature of the dependence TKE(M) for both nuclei is that there is a peak in the region of fragment mass $A/2 \pm 8$. This peak seems to be due to the superposition of the contributions from the compound nucleus fission and quasi-fission in this mass region. This interpretation is supported by an analysis of the fragment yield $Y(M)$ for the reaction $^{48}\text{Ca} + ^{244}\text{Pu}$ in relation to the TKE energy range. As our investigations have shown⁵, the absolute values of TKE for the quasi-fission process in the region of $Z > 100$ nuclei are, on the average, higher than the corresponding values for the fission of compound nuclei; and the higher Z of the fissioning nucleus is, the greater the difference in TKE (which, as will be shown below, may be as much as 20 – 40 MeV). Fig. 4 shows the fragment yield $Y(M)$ for the $^{292}\text{114}$ nucleus in the range of the TKE values > 230 MeV ($\langle \text{TKE} \rangle = 220$ MeV). It is clearly seen that in the same region of fragment mass as that shown in Fig.3 there appears a peak in $Y(M)$ whereas in the total yield

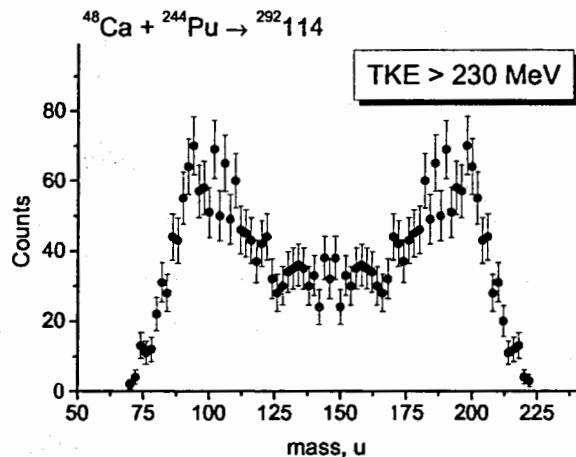


Figure 4: Fission fragment mass distribution for the $^{48}\text{Ca} + ^{244}\text{Pu} \rightarrow ^{292}114$ reaction for the range of $\text{TKE} > 230$ MeV.

$Y(M)$ shown in Fig.2b there is no peak. It is also to be noted that this effect masks the fact that the mass distribution of the fission fragments of compound nuclei presented in Fig.2 is asymmetric, i.e. the peak-valley ratio for the mass distribution seems to be slightly higher than that in Fig.2b.

On the whole, the analysis performed of the mass and energy distributions of the fission fragments of nuclei in reactions with ^{48}Ca ions allowed reasonably reliable values to be found for the quasi-fission and fission cross sections of compound nuclei of ^{256}No , $^{286}112$, and $^{292}114$, which are given in Table 1. These data show reactions with ^{48}Ca ions to be promising for producing not only element 114 but also heavier nuclides of elements 115, 116 and 118.

Let us discuss the reaction $^{86}\text{Kr} + ^{208}\text{Pb} \rightarrow ^{294}118$, for which in Fig.5 the

Table 1: Cross sections and TKE.

Reactions	E_{lab} (MeV)	E^* (MeV)	σ_{fis}	$\sigma_{\text{fis}}/\sigma_{\text{cap}}$ (%)	$\langle \text{TKE} \rangle$ (MeV)
$^{48}\text{Ca} + ^{208}\text{Pb}$	230	33	350 mb	96	193
$^{48}\text{Ca} + ^{238}\text{U}$	232	33	6 mb	3	215
$^{48}\text{Ca} + ^{244}\text{Pu}$	233.5	33	4 mb	9	220
$^{86}\text{Kr} + ^{208}\text{Pb}$	468	28	$\sim 6\mu\text{b}$	$\leq 10^{-3}$	260
	453	15	≤ 500 nb	-	260

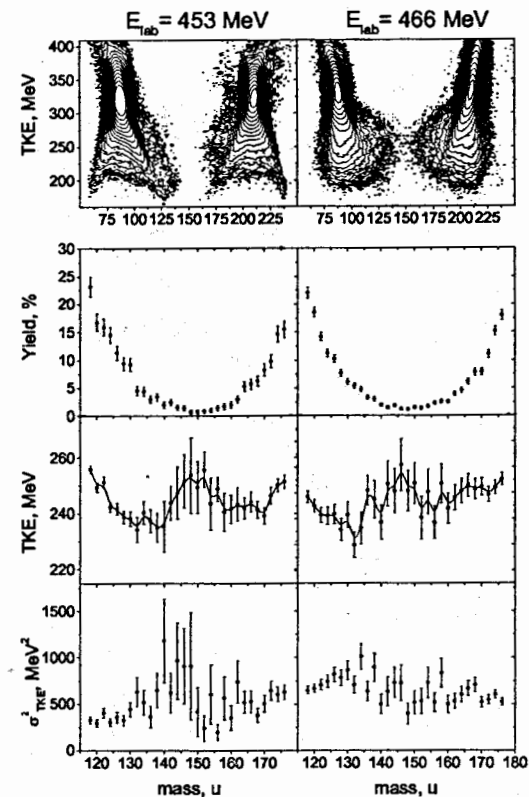


Figure 5: Mass-energy distributions of the $^{86}\text{Kr} + ^{208}\text{Pb}$ reaction products.

TKE-M matrices are presented for two values of excitation energy. Here too the fragment yields in the symmetric mass region $A/2 \pm 30$ amu and the dependence $\text{TKE}(M)$ and $\sigma_{\text{TKE}}^2(M)$ are presented. The data presented differ essentially from the data for the reaction $^{244}\text{Pu} + ^{48}\text{Ca}$. First, the quasi-fission peaks of fragment average mass $A = 208$ and additional to it are not separated from the elastic and quasi-elastic scattering peaks; second, the dependence $\text{TKE}(M)$ differs sharply from that in Fig. 3.

In this case the TKE average value is markedly greater than the value to be expected for compound nucleus fission. To illustrate this point, Fig.6 gives the known TKE systematics in relation to the Coulomb parameter $Z^2/A^{1/3}$. This systematics includes both the data from work ⁶ for $Z > 100$ obtained

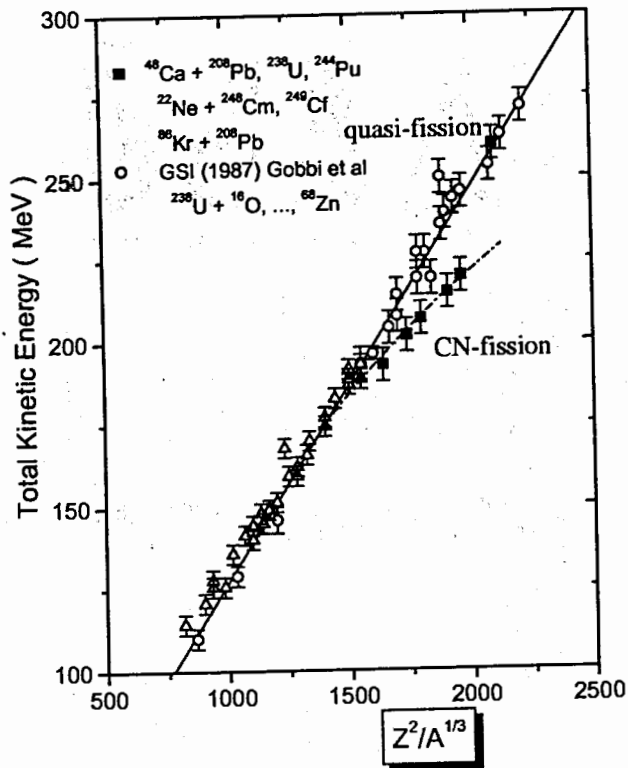


Figure 6: The dependence of TKE on $Z^2/A^{1/3}$ -parameter. The solid squares represent the data from our work, another open signs – the data from work [6].

in a reaction characterized by quasi-fission being the dominant process and our results just for the fission of compound nuclei of ^{256}No , ^{270}Sg ⁷, ^{271}Hs ⁸, $^{286}\text{112}$, $^{292}\text{114}$. It is seen that the dependences of TKE on $Z^2/A^{1/3}$ for fission and quasi-fission differ essentially from each other and the difference in TKE even for $Z = 114$ is as much as 25 MeV. Extrapolation of the dashed line up to $Z = 118$ would give ≈ 230 MeV for the value of TKE for the compound nucleus fission whereas our experimental result was 260 MeV for TKE for this nucleus. The latter value is in excellent agreement with the results of work⁵ for the quasi-fission process. From this only one conclusion can be drawn. In the case of the reaction $^{86}\text{Kr}+^{208}\text{Pb}$, in the symmetric fission region ($A/2 \pm 30$) the quasi-fission process dominates, by which this reaction differs sharply from the reaction $^{48}\text{Ca}+^{238}\text{U}$ and especially from $^{48}\text{Ca}+^{244}\text{Pu}$, in which the contribution of the compound nucleus fission in the same region of fragment

mass is dominant.

An attempt can be made to evaluate the contribution of the fission cross section of the compound nucleus for the reaction discussed, using the dependence of TKE on $Z^2/A^{1/3}$ found by us, according to which the kinetic energy of the fragments will be 230 MeV for the fission of the $^{294}\text{118}$ compound nucleus, and to select the events that belong to the range of 230 ± 20 MeV. Table 1 presents both the results obtained from the studied reactions with ^{48}Ca ions and our estimates for the quasi-fission and fission of the compound nuclei for the reaction $^{86}\text{Kr}+^{208}\text{Pb}$.

From these data it follows that the fission cross section, and consequently the production cross section of the $^{294}\text{118}$ compound nucleus at the excitation energy $E^* = 28$ MeV is more than two orders lower than σ_f for $^{286}\text{112}$ and $^{292}\text{114}$ nuclei ($E^* \approx 33$ MeV). It should be noted that, as the excitation energy changes by 13 MeV (from $E^* = 28$ MeV up to $E^* = 15$ MeV), the cross section σ_f for the $^{292}\text{118}$ drops by no more than one order and it is beyond reason to assume that an increase in energy from $E^* = 28$ to $E^* = 33$ MeV will result in the cross section being changed by more than a factor of 3 – 5. From this the conclusion could be drawn that more symmetric reactions similar to $^{86}\text{Kr}+^{208}\text{Pb}$ are far less promising for producing superheavy nuclei. Nevertheless the recent results of the synthesis of element 118 in the cold fusion reaction $^{86}\text{Kr}+^{208}\text{Pb}$ at Berkeley³ deny such an assumption, that is, if the value $\sigma_{ev} = 2$ pb obtained from this experiment is correct, then the relationship between the fission width and neutron width at excitation energy of ≈ 15 MeV changes drastically in favour of the survival of the compound nucleus as compared with the case of “cold” fusion in the reaction $^{48}\text{Ca}+^{208}\text{Pb}$ at the same excitation energy and the case of “warm” fusion in the reactions $^{48}\text{Ca}+^{238}\text{U}$ and $^{48}\text{Ca}+^{244}\text{Pu}$. Therefore further research in this direction is extremely interesting both to fission physics and to the problem of synthesising superheavy nuclei.

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