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Production of Exotic Nuclei in Low-Energy Multi-Nucleon Transfer Reactions

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Abstract: Multinucleon transfer processes in low-energy heavy ion collisions open a new field of research in nuclear physics, namely, production and studying properties of heavy neutron rich nuclei. This not-yet-explored area of the nuclear map is extremely important for understanding the astrophysical nucleosynthesis and the origin of heavy elements. Beams of very heavy U-like ions are needed to produce new long-living isotopes of transfermium and superheavy elements located very close to the island of stability. The calculated cross sections are high enough to perform the experiments at available accelerators. Beams of medium-mass ions (such as ^{136}Xe, ^{192}Os, ^{198}Pt) can be used for the production of neutron rich nuclei located along the neutron closed shell $N=126$ (the last waiting point) having the largest impact on the astrophysical r-process. The Low-energy multinucleon transfer reactions is a very efficient tool also for the production and spectroscopic study of light exotic nuclei. The corresponding cross sections are 2 or 3 orders of magnitude larger as compared with high energy fragmentation reactions.

Key words: exotic nuclei; multi-nucleon transfer reaction; superheavy

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1 Introduction

Upper part of the present-day nuclear map consists mainly of proton rich nuclei approaching the proton drip line (see Fig. 1). Neutron enriched isotopes of heavy elements were not synthesized and studied so far. This unexplored area of heavy neutron enriched nuclides (also those located along the neutron closed shell $N=126$ to the right-hand side of the stability line) is extremely important for nuclear astrophysics investigations and, in particular, for the understanding of the r process of astrophysical nucleogenesis.

Light and medium-mass neutron rich nuclei are successfully produced in high-energy fragmentation processes and in fission reactions, correspondingly. Great progress here was done lately and dozens of new nuclei have been synthesized and studied, mainly at the laboratories of NSCL MSU[1], RIKEN[2] and GSI[3]. Evidently these reactions cannot be used for the production of heavy neutron enriched nuclei.

There are only three methods for the production of heavy elements: (1) fusion reactions, (2) a sequence of neutron capture and β^- decay processes, and (3) multi-nucleon transfer reactions.

Because of the bending of the stability line toward the neutron axis, in fusion reactions only proton-rich isotopes of heavy elements can be produced. That is the main reason for the impossibility of reaching the island of stability ($Z\sim 110/120$ and $N\sim 184$) in the superheavy mass region by fusion reactions with stable projectiles. The use of neutron rich radioactive projectiles cannot help to solve this problem due to low intensities of such beams and extremely low production cross sections.

Further progress in the synthesis of new elements with $Z>118$ is also not quite evident. Cross sections of the "cold" fusion reactions decrease very fast with increasing charge of the projectile (they become less than 1 pb already for $Z\geqslant 112$[4–5]. For the more asymmetric ^{48}Ca induced fusion reactions rather constant values (of a few picobarns) of the cross sections for the production of superheavy (SH) elements with $Z=112/118$ have been predicted in Refs. [6–7]. This unusual (at first sight) behavior of the cross sections was explained in Ref. [6–7] by the relatively slow decrease of the fusion probability (in contrast to the more symmetric "cold" fusion reactions) and by the increasing survival probability of compound nuclei (CN) ow-

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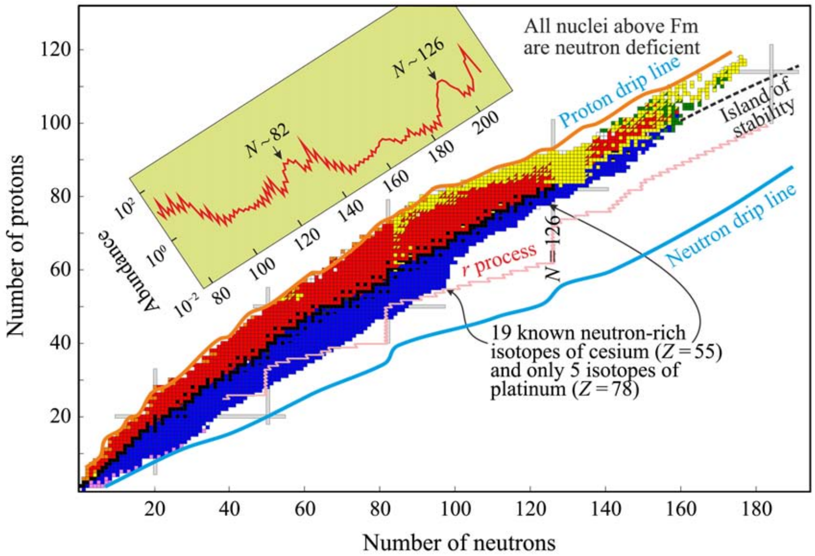

Fig. 1 (color online) Nuclear map as it looks today. Upper-right (north-east) area of the nuclear map (near and to the right of the beta-stability line) is not yet explored whereas on the proton rich side there is almost no space for discovery of new isotopes.

ing to increasing values of their fission barriers caused by the larger shell corrections as the CN approach the neutron and proton closed shells[8–9] in the region of the island of stability. These predictions have been fully confirmed by the experiments performed in Dubna[10] and later in Berkeley[11] and at GSI[12–13].

For the moment Cf ($Z=98$) is the heaviest available target that can be used in experiments. The half-life of the einsteinium isotope, $^{254}_{99}$Es, is 276 days, sufficient to be used as target material. In principle, this isotope might be produced in nuclear reactors, but it is rather difficult to accumulate the required amount of this matter (several milligrams) to prepare a target. The cross section for the production of element 119 in the hypothetical ^{48}Ca+^{254}Es fusion reaction is about 0.3 pb[14] which is more promising as compared with the ^{50}Ti+^{249}Bk fusion reaction[15]. Anyhow, to get SH elements with $Z>118$ in fusion reactions in a more realistic way, one should proceed to heavier than ^{48}Ca projectiles.

Strong dependence of the calculated evaporation residue (EvR) cross sections for the production of SH elements on the mass asymmetry in the entrance channel makes the nearest to ^{48}Ca projectile, ^{50}Ti, most promising for further synthesis of SH nuclei. Our calculations demonstrated that the use of the titanium beam instead of ^{48}Ca decreases the yield of the same SH element due to a worse fusion probability by about factor 20[15]. The elements 119 and 120 can be produced in the fusion reactions of ^{50}Ti with ^{249}Bk and ^{249}Cf targets (or in the ^{54}Cr+^{248}Cm fusion reaction) with the cross sections of about 0.04 pb[15] which are already at the limit of the experimental possibilities. The synthesis of these nuclei may encounter also another important problem. The proton rich isotopes of SH elements produced in these reactions are rather short-living due to large values of Q_α. Their half-lives are very close to the critical value of one microsecond needed for compound nucleus to pass through the separator up to the focal plane detector. The next elements (with $Z>120$) being synthesized in such a way might be already beyond this natural time limit for their detection. Thus, future studies of SH elements are obviously connected with the production of neutron enriched and longer living isotopes of SH nuclei.

The neutron capture process is an alternative (oldest and natural) method for the production of new heavy elements. Strong neutron fluxes might be provided by nuclear reactors and nuclear explosions under laboratory conditions and by supernova explosions in nature. The "fermium gap" consisting of the short-living isotopes $^{258-260}$Fm located on the β–stability line and having very short half-lives for spontaneous fission, impedes the formation of nuclei with $Z>100$ by the weak neutron fluxes realized in existing nuclear reactors. Theoretical models predict also another region of short-living nuclei located at $Z=104/108$ and $A\sim 275$. In nuclear and supernova explosions (fast neutron capture) these gaps may be bypassed if the total neutron fluence is high enough. Note that elements 99 and 100 (einsteinium and fermium) were first discovered in debris of the test thermonuclear explosion "Mike"[16].

The resulting charge number of the synthesized nuclei might be increased by sequential neutron flux exposure if two or several nuclear explosions were generated in close proximity to each other[17]. The same process of multiple neutron exposures might be also realized in pulsed nuclear reactors. Here the pulse duration is much longer than in nuclear explosions (up to several milliseconds). However, the neutron fluence usually does not exceed 10^{16} n/cm^2 in existing nuclear reactors ($n_0 \sim 10^{19}$ n/cm$^2\cdot$s during one millisecond pulse). Thus, the time of neutron capture $\tau_n = (n_0\sigma_{n\gamma})^{-1} \sim 10^5$ s, and only rather long-living neutron rich nuclei (with $T_{1/2}$ longer than several hours) are formed in these reactors. Multi–pulse irradiation here corresponds to the "slow" neutron capture process, in which the isotopes of newly formed elements with larger charge numbers are situated close to the line of stability and finally reach the fermium gap ($Z = 100$) where the process stops. In this case the probability for formation of heavy elements with $Z > 100$ is negligibly small independent of the number of pulses and total time of irradiation.

The situation may change if one could be able to increase somehow the intensity of the pulsed reactor. The neutron fluence of one pulse and frequency of pulses should be high enough to bypass both gaps of short living nuclei on the way to the island of stability. The specifications of the high–intensity pulsed reactors of the next generation depends strongly on properties of heavy neutron rich nuclei located to the right of these gaps. Using theoretical estimations for the decay properties of these nuclei (which are not yet synthesized) we have found that an increase of the neutron fluence in the individual pulse by about three orders of magnitude as compared with existing pulsed reactors could be quite sufficient to bypass both gaps[17].

Thus, for the moment there is only one realistic method for the production of heavy neutron rich nuclei, namely, multinucleon transfer reactions in collisions of heavy ions[18]. Before the practical use of this method the following questions have to be answered. What are the cross sections for production of new neutron rich isotopes of heavy elements? What are the optimal combinations of colliding ions for production of sought for nuclei? What is optimal beam energy? How to separate a given transfer reaction product from other "garbage" produced in such kind of reactions?

2 Multinucleon transfer reactions

Damped collisions of heavy ions and, in particular, multinucleon transfer processes were studied intensively during many years (see, for example, recent review papers[19–20] and appropriate references therein). Transport models (Focker-Planck[21] and master equations[22] for the corresponding distribution functions and the Langevin equations[23]) were proposed many years ago for the description of heavy ion damped collisions. However, those time only qualitative understanding of these reactions was achieved. Quite recently it becomes possible to describe quantitatively all the features of heavy ion deep inelastic scattering and related quasi-fission processes (energy, angular, mass and charge distributions of reaction products)[24].

Damped collision of heavy ions is a very complicated process in which many collective degrees of freedom play an important role while intrinsic (single particle) degrees of freedom effectively damp kinetic energy of relative motion leading to a high viscosity on nuclear matter. The proper choice of the unified degrees of freedom playing most principle role both at approaching stage and at the stage of formation and fission (quasi-fission) of strongly deformed mono-nucleus is essential and rather difficult. The number of the degrees of freedom should not be too large so that one is able to solve numerically the corresponding set of coupled dynamic equations. On the other hand, however, with a restricted number of collective variables it is impossible to describe simultaneously strongly coupled deep inelastic scattering and fusion-fission processes. The second problem here is a calculation of potential energy of the whole nuclear system gradually transforming from the configuration of two approaching nuclei into configuration of deformed mono-nucleus and then to final reaction fragments or to more or less spherical compound nucleus. Finally dynamic equations of motion should be formulated for the chosen degrees of freedom.

Calculations performed within the microscopic time-dependent Schrödinger equations[25] have clearly demonstrated that at low collision energies of heavy ions already at approaching stage the wave functions of valence nucleons occupy the two-center molecular states spreading gradually over volumes of both nuclei, that is the valence nucleons start to move in volumes of both nuclei thus forming a mono-nucleus configuration with a two-center mean field. The same adiabatic low-energy collision dynamics of heavy ions was found also within the TDHF calculations[26–27].

This means the following. (1) Any perturbation model based on a calculation of the sudden overlapping of single-particle wave functions of transferred nucleons (in donor and acceptor nuclei, respectively) is not applicable for description of nucleon transfers in low-energy heavy-ion damped collisions. (2) The so called DNS model assuming independent nucleon motion in two isolated mean fields is absolutely contrary to physics. (3) One-dimensional potential energy $V(R)$ has no meaning at $R \leqslant R_{\rm contact}$ as well as any speculations on the depth of potential pocket of $V(R)$. In low-energy collisions of heavy ions at $R \leqslant R_{\rm contact}$ the *multi-dimensional* potential energy (dependent on the shape parameters of the united nuclear system) must be used. The two center shell model looks most appropriate for calculation of this potential energy.

It was shown in Ref. [24] that the Langevin-type dynamical equations of motion (using this two center adiabatic driving potential) can be successfully applied for simultaneous description of strongly coupled multinucleon transfer, quasi-fission and fusion-fission reaction channels of low-energy heavy ion collisions. The distance between the nuclear centers $\boldsymbol{R}$ (corresponding to the elongation of a mono-nucleus when it is formed), dynamic spheroidal-type surface deformations of both fragments δ_1 and δ_2, their rotation angles, and the neutron and proton asymmetries, η_N and η_Z are the most relevant degrees of freedom for the description of damped collisions of heavy ions jointly with fusion-fission dynamics. For all the variables, with the exception of the neutron and proton transfers, we use the usual Langevin equations of motion with the inertia parameters, μ_R and μ_δ, calculated within the Werner-Wheeler approach

$$\frac{{\rm d}q_i}{{\rm d}t} = \frac{p_i}{\mu_i},\ \frac{{\rm d}p_i}{{\rm d}t} = \frac{\partial V_{\rm eff}}{\partial q_i} - \gamma_i \frac{p_i}{\mu_i} + \sqrt{\gamma_i T}\,\varGamma_i(t)\ .$$

Here q_i is one of the collective variables, p_i is the corresponding conjugate momentum, $V_{\rm eff}$ includes the centrifugal potential, $T = \sqrt{E^*/a}$ is the local nuclear temperature, $E^* = E_{\rm c.m.} - V_{\rm eff}(q_i;t) - E_{\rm kin}$ is the excitation energy, γ_i is the appropriate friction coefficient, and $\varGamma_i(t)$ is the normalized random variable with Gaussian distribution. The quantities γ_i, E^* and T depend on the coordinates and, thus, on time.

Nucleon exchange (nucleon rearrangement) is described by the inertialess Langevin type equations of motion derived from the master equations for the corresponding distribution functions[24]

$$\frac{{\rm d}\eta_N}{{\rm d}t} = \frac{2}{N_{\rm CN}} D_N^{(1)} + \frac{2}{N_{\rm CN}} \sqrt{D_N^{(2)}}\,\varGamma_N(t)\ ,$$
$$\frac{{\rm d}\eta_Z}{{\rm d}t} = \frac{2}{Z_{\rm CN}} D_Z^{(1)} + \frac{2}{Z_{\rm CN}} \sqrt{D_Z^{(2)}}\,\varGamma_Z(t)\ .$$

Here $D^{(1)}$, $D^{(2)}$ are the transport coefficients. We assume that sequential nucleon transfers play a main role in mass rearrangement. Other details as well as discussion on the values of nuclear viscosity and nucleon transfer rate can be found in Ref. [24].

For the moment this approach is the only one which reproduces quite properly all the regularities of heavy ion deep inelastic scattering and quasi-fission processes. As an example, in Fig. 2 experimental and

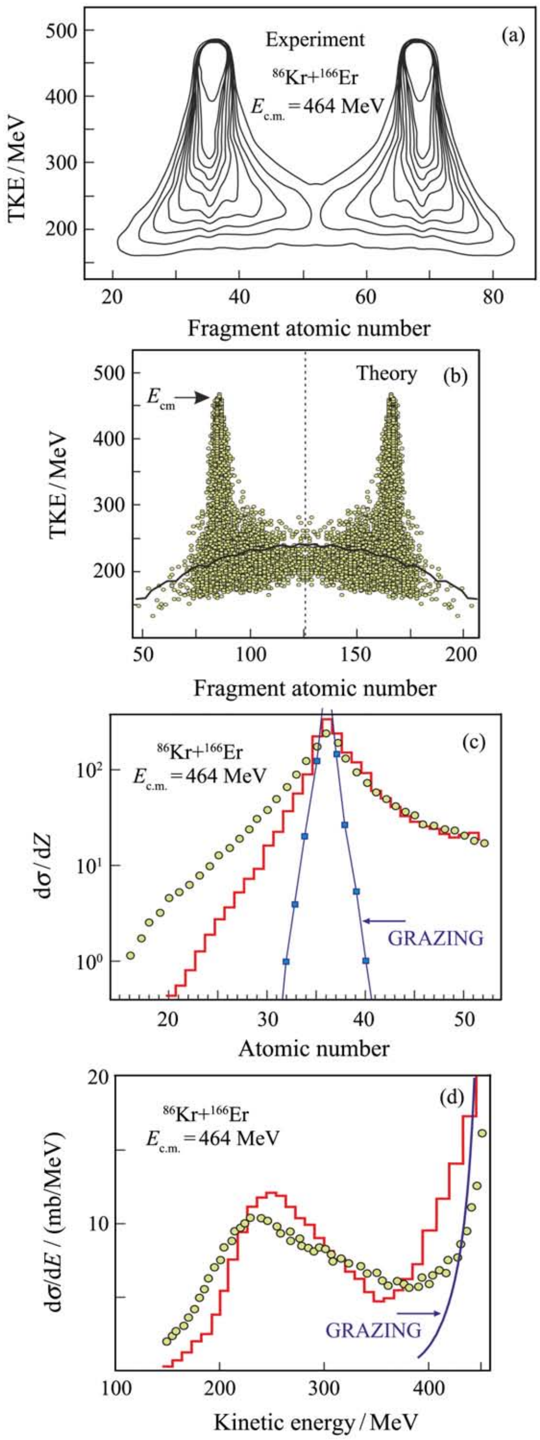

Fig. 2 (color online) Charge, mass and energy distributions of reaction fragments in collisions of ^{86}Kr with ^{166}Er at $E_{\rm c.m.}$=464 MeV[28]. The histograms indicate the calculations performed within the model described above whereas the curves show the GRAZING calculations.

theoretical energy–mass distributions of reaction fragments are shown formed in collisions of ^{86}Kr with ^{166}Er at 464 MeV center-of-mass energy.

Some misunderstanding (or terminology) should be clarified here. The well known GRAZING code[29] (which is used very often for description of nucleon transfers) describes quite well few nucleon transfer reactions in grazing collisions. However, this code is not designed for the description of *multinucleon* transfer processes. It gives too narrow mass distributions of reaction fragments because the damped reaction channels with large kinetic energy loss are not included in the model (see Fig. 2).

3 Production of new nuclei located along the closed neutron shell $N = 126$

Near barrier collisions of ^{136}Xe and ^{192}Os with ^{208}Pb target were predicted to be quite promising for the production of new nuclei with $N \sim 126$[30]. This area of the nuclear map (being the last waiting point) has largest impact on astrophysical r process. Our predictions of the production cross sections are not confirmed yet experimentally (because of difficulty of isotopic separation of heavy transfer reaction products) but all other reaction regularities (mass, energy and angular distributions) were found in good agreement with the predictions (see Fig. 3). Recently (July of 2014) this reaction was studied in Legnaro using the PRISMA separator to measure the cross sections for the production of specific neutron rich target-like nuclei. The data are still under processing.

Low-energy collisions of stable neutron enriched isotopes of elements located below lead (such as ^{192}Os or ^{198}Pt) with available actinide targets look even more favorable for the production and study of new neutron rich nuclei located along the closed neutron shell $N = 126$. Distribution of primary reaction fragments in (Z, N) plane is shown in Fig. 4 for the case of low-energy collisions of ^{198}Pt with ^{238}U at $E_{\rm c.m.}$=700 MeV. As can be seen a lot of new isotopes in the region of the closed neutron shell $N = 126$ can be synthesized in this reaction with cross sections higher than 1 μb.

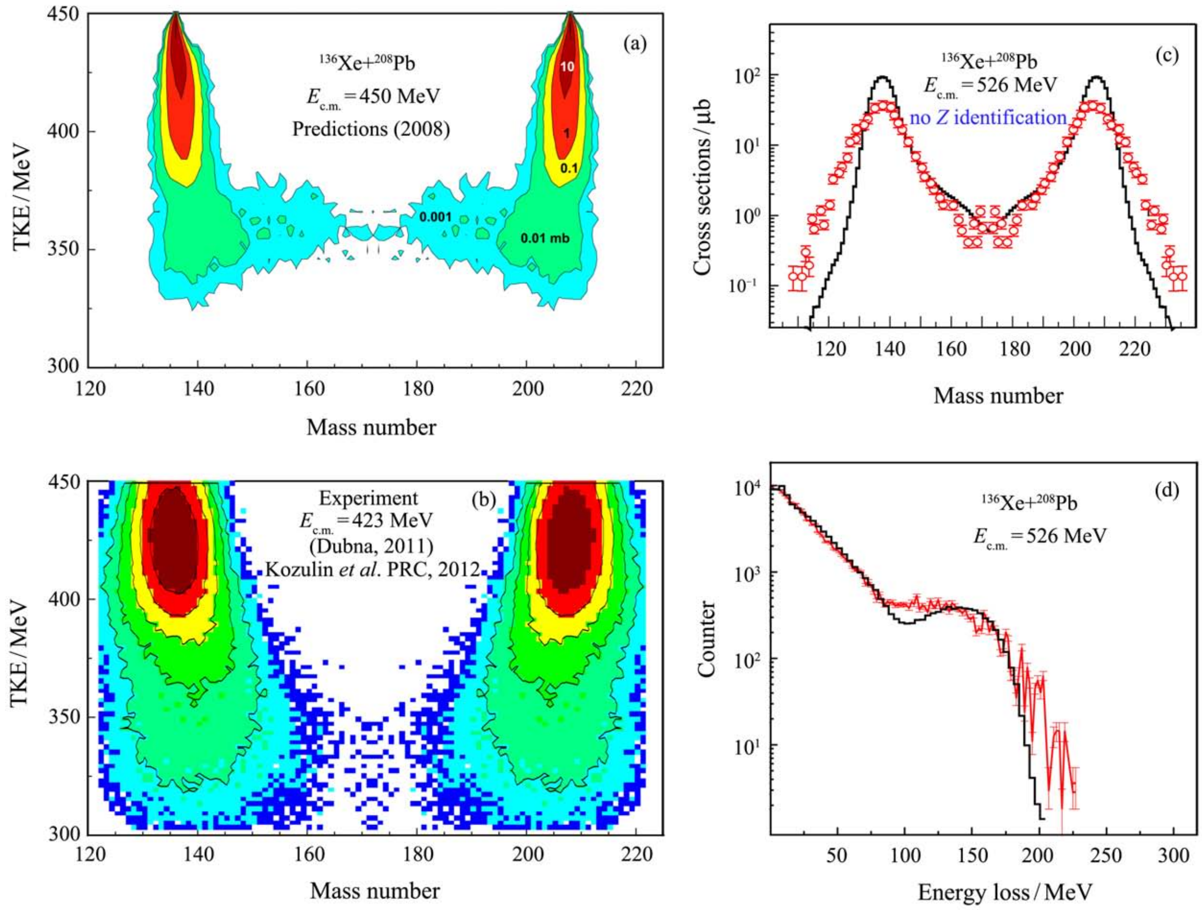

Fig. 3 (color online) Predicted[30] and measured[31] mass and energy distributions of reaction fragments in collisions of ^{136}Xe with ^{208}Pb target at near barrier energy.

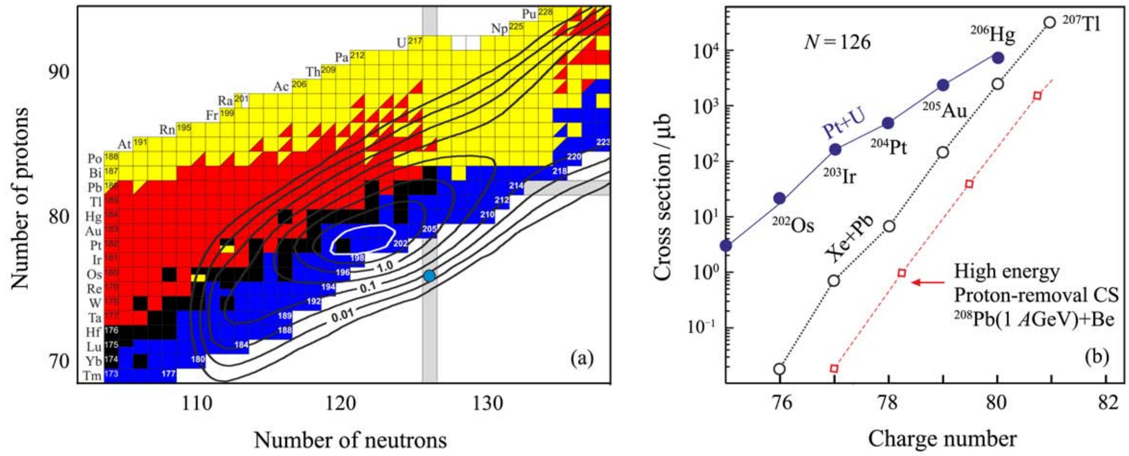

Fig. 4 (color online) (a) Landscape of the cross sections for the production of isotopes around $N = 126$ in collisions of ^{98}Pt with ^{238}U at $E_{\rm c.m.}$=700 MeV. (b) Cross sections for the production of nuclei with $N = 126$ in low-energy collisions of ^{98}Pt+^{238}U and ^{136}Xe+^{208}Pb and in high-energy proton removal reaction[32].

4 Neutron enriched transfermium and superheavy nuclei

Synthesis and studying properties of new neutron rich fermium isotopes with $A > 260$ are extremely interesting by several reasons. First, as mentioned above, all known isotopes of fermium (and of more heavy elements) are located to the left side of the beta-stability line (see Fig. 1). Second, the well known “fermium gap” ($^{258-260}$Fm isotopes with very short half-lives for spontaneous fission) impedes formation of nuclei with $Z > 100$ by the weak neutron fluxes realized in existing nuclear reactors. It is extremely interesting to know what is the first β^--decayed Fermium isotope and how long is its half-life. This is important not only for reactor but also for explosive nucleosynthesis in which this Fermium gap might be bypassed[17]. As can be seen from Fig. 5 neutron rich fermium isotopes can be produced in low-energy transfer reactions with cross sections of about 0.1 μb, that is quite sufficient to be produced at available accelerators.

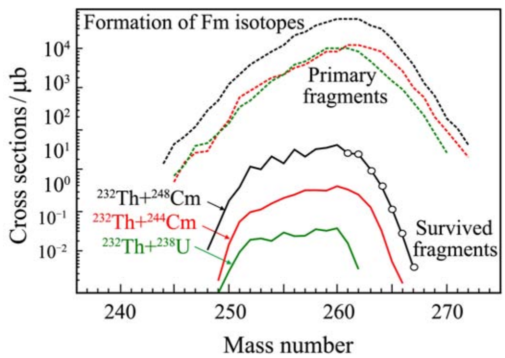

Fig. 5 (color online) Isotopic yields of fermium nuclei formed in collisions of ^{232}Th with ^{238}U, ^{244}Pu and ^{248}Cm at $E_{\rm c.m.}$=715, 730 and 750 MeV, correspondingly. Open circles indicate new isotopes.

The use of actinide beams and actinide targets give us a possibility to produce new neutron enriched isotopes of SH elements located along the stability line and to the right of it, that is in the unexplored area of the nuclear map (see Fig. 1). The yields of neutron enriched long living isotopes of SH elements in transfer reactions might be significantly enhanced owing to the shell effects leading to the so-called “inverse quasi-fission” phenomena[33]. For example, in collisions of ^{238}U with ^{248}Cm target nucleons are predominantly transferred from the lighter partner (here is uranium) to heavy one (*i.e.* U transforms to doubly magic Pb nucleus whereas Cm to 106 element). In this case, instead of conventional mass distribution of reaction fragments (smoothly decreasing with increasing mass transfer, see Figs. 2 and 3) one can expect to see the “lead shoulder” on the “left side” of the mass distribution and enhanced yield of primary SH fragments with masses heavier than target. This phenomena is not discovered yet experimentally because of difficulty to perform such reaction.

However, there is the surrogate reaction, ^{160}Gd +^{186}W, in which the same “inverse quasi-fission” process is also expected[33] owing to the neutron closed shells $N = 82$ and $N = 126$ located from the outside of the colliding partners (to the left from the projectile and to the right from the target on the mass axis). Recently this reaction was studied with the use of radiochemical method[34]. Predicted and observed mass distributions of target-like nuclides formed in this reaction at $E_{\rm c.m.}$=460 MeV are shown in Fig. 6. Unfortunately, experimental technique (catcher foils + off-line radiochemistry) did not allow to measure the yields of stable (as well as short-living) isotopes and to reach the region of expected decrease of the cross sections for the production of trans-lead nuclei (see Fig. 6). On-line measurement of the mass distribution of all the fragments formed in this reaction is planned to be performed in Dubna in 2014. If this "inverse quasi-fission" phenomena will be confirmed it will open a real way for the production of neutron enriched long living isotopes of superheavy elements.

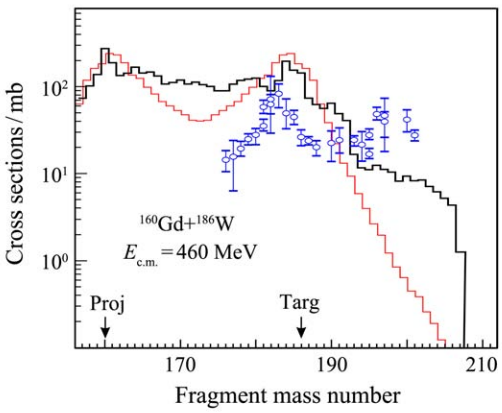

Fig. 6 (color online) (Predicted[33] and observed[34] mass distributions of reaction fragments (with energy loss higher than 15 MeV) formed in collisions of ^{160}Gd with ^{186}W at $E_{\rm c.m.}$=460 MeV. Experimental data are shown only for target-like fragments. Thick and thin histograms show the results of calculations with and without shell corrections in potential energy.

In Fig. 7 the results of our predictions are shown for the formation of survived isotopes of some transfermium elements in reaction ^{238}U+^{248}Cm at 770 MeV center-of-mass energy. The obtained results are rather optimistic. New neutron rich isotopes of transfermium elements with $Z = 100 \sim 104$ (located already at the stability line and beyond it) can be produced with the cross sections of several hundreds of picobarn. The cross sections for the production of new neutron rich isotopes of seaborgim and hassium ($Z = 106, 108$) are also higher than 1 picobarn. Predicted cross sections depend on the values of neutron and proton transfer rates which are not determined yet very accurately (see Refs. [21–22] and [24]).

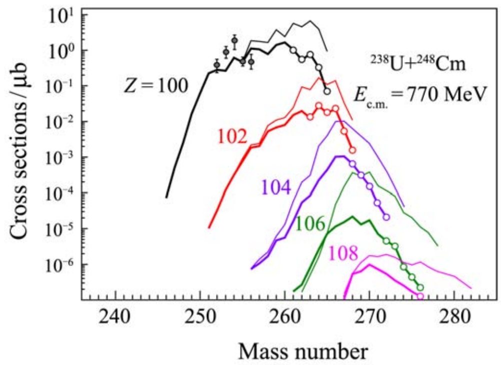

Fig. 7 (color online) Cross sections for the production of neutron rich SH nuclei in collisions of ^{238}U with ^{248}Cm target at $E_{\rm c.m.}$=770 MeV. Thin curves are obtained with the nucleon transfer rate parameter multiplied by factor 2.

5 Low-energy nucleon transfers vs. high-energy fragmentations

Being inspired by good agreement of the model with available experimental data on damped collisions of heavy ions, we studied also the multinucleon transfer reactions in low-energy collisions of light heavy ions with heavy targets[35]. The results of these calculations (shown in Fig. 8) demonstrate that the cross sections for the production of quite exotic light neutron-rich nuclei produced in the low-energy multinucleon

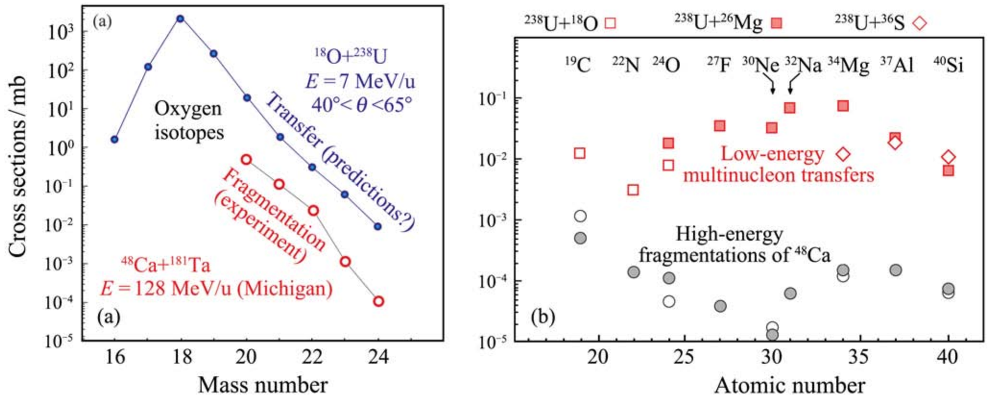

Fig. 8 (color online) (a) Cross sections for the production of neutron rich oxygen isotopes in low-energy collisions of ^{18}O with ^{238}U target and in fragmentation process[36]. (b) Formation of light neutron rich nuclei in low-energy collisions of ^{18}O, ^{26}Mg and ^{36}S with ^{238}U target and in fragmentation of 128 AMeV ^{48}Ca on ^{181}Ta target[36] (filled circles) and 345 AMeV ^{48}Ca on ^{9}Be[2] (open circles).

transfer reactions are higher by about 2 orders of magnitude as compared with high energy fragmentation reactions. Thus the low energy damped collisions of light heavy ions with heavy targets look very promising and quite competitive to the high-energy fragmentation reactions for the production and study of light exotic nuclei. The gain of high-energy beams, namely, the use of thicker targets, is fully compensated by higher intensity of low-energy primary beams of such projectiles as ^{18}O, ^{26}Mg and others, by higher cross sections, and by much cheeper setups used in low-energy experiments. All these factors make low-energy damped collisions of light heavy ions quite attractive for the production and study of light exotic nuclei just at presently available experimental facilities.

6 The problem of separation of heavy reaction products

In contrast with fusion reactions (in which a sought-for residual compound nucleus moves in forward direction with well known velocity) it is more difficult to separate a given product of multinucleon transfer reaction (having rather broad angular and velocity distribution) from all the other reaction fragments. As a result, heavy nuclei with $Z > 70$ formed in the multinucleon transfer reactions cannot be separated and studied at available setups created recently just for studying the products of deep inelastic scattering (such as VAMOS, PRISMA and others). These fragment separators (as well as other setups) cannot distinguish heavy nuclei with $Z > 70$ by their atomic numbers. However, during the last several years a combined method of separation is intensively studied based on stopping nuclei in gas and subsequent resonance laser ionization of them[37]. One of such setups (named GaLS: in Gas cell Laser ionization and Separation) is currently created at Flerov Laboratory (JINR, Dubna, see Fig. 9). Experiments at this setup aimed on the production and studying properties of new neutron rich heavy nuclei are planned to start in the beginning of 2015.

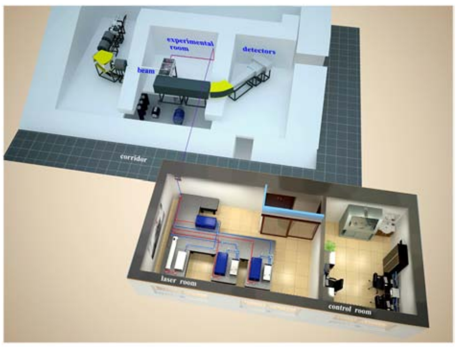

Fig. 9 (color online) 3d view of the setup GaLS (Dubna) for selective laser ionization and separation of heavy transfer reaction products.

7 Summary

The use of multinucleon transfer reactions in low-energy collisions of heavy ions opens a new field of research in nuclear physics: synthesis and studying properties of heavy neutron rich nuclei. These nuclei can be produced neither in fusion reactions nor in fragmentation processes.

^{136}Xe+^{208}Pb, ^{192}Os+^{208}Pb and ^{198}Pt+^{238}U are the most appropriate reactions for the production of neutron rich nuclei located along the closed neutron shell $N = 126$ having the largest impact on the astrophysical r-process. The use of actinide beams and actinide targets allows one to produce new neutron enriched (and longer living) isotopes of transfermium and superheavy elements located along the stability line and to the right of it. Shell effects (inverse quasi-fission mechanism) might significantly enhance the cross sections for the production of trans-target nuclei. The low energy damped collisions of light heavy ions with heavy targets look also very promising and quite competitive to the fragmentation reactions for the production and study of light exotic nuclei. Separators of new type based on selective laser ionization of heavy reaction products have to be designed and installed for this new field of research.

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