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Breakdown of the compound-nucleus model in the fusion-evaporation process for ^{110}Pd + ^{110}Pd*

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The fusion of the massive systems ^{110}Pd + ^{104}Ru and ^{110}Pd + ^{110}Pd was uniquely identified by observing the α decay of the evaporation residues. The observed distribution of the fusion cross section on the different evaporation-residue channels is in clear contradiction to calculations based on the compound-nucleus model. As a possible explanation the precompound evaporation of α particles is proposed.

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During the last years we studied the fusion of heavy symmetric systems like ^{124}Sn + ^{96}Zr [1] and ^{100}Mo + ^{110}Pd [2]. The present work extends these studies to even heavier symmetric systems. In such systems, the ratio between the repelling Coulomb force and the attractive nuclear force at the contact point of the two colliding nuclei, which decides whether the system is able to fuse or not, is as large as in the fusion reactions used to synthesize the heaviest elements, e.g. ^{58}Fe + ^{209}Bi leading to element 109 [3]. However, compared to the reactions which synthesized the heaviest elements the symmetric systems show a much higher evaporation-residue cross section thus allowing a systematic investigation of the fusion process.

In contrast to the light and medium-heavy systems more massive systems slightly heavier than ^{90}Zr + ^{90}Zr [4] exhibit a considerable deficit of fusion above the expected fusion barrier [1, 2, 4, 5]. This deficit is conventionally parametrized by a shift of the fusion barrier called extra-push, which is predicted by macroscopic models. These models consider the development of the combined system in a multidimensional space including potential and inertial forces [6] as well as one-body dissipation [7, 8]. According to the ideas of Swiatecki, the extra-push is mainly determined by the location of the fission barrier relative to the fusion barrier in a two-dimensional landscape. The two dimensions are the deformation or a distance parameter between the centers of the two nuclei, and a parameter representing the neck diameter of the amalgamating nuclei. For light systems, the distance parameter at the fission barrier is larger than that at the fusion barrier which approximately corresponds to the contact point. Therefore fusion may be achieved as soon as the fusion barrier is passed. On the other hand, for heavy systems, the situation will be reversed: The fission barrier corresponds to a distance parameter smaller than the fusion barrier. After contact, i.e. at the fusion barrier, when the system is forming a neck, it may evolve towards reseparation instead of passing the fission saddle point and forming a compound nucleus. An extra-push is necessary in order to drive the system to a deformation smaller than that of the fission saddle. In the case the system ^{110}Pd + ^{110}Pd, an extra-push energy of 60 MeV is predicted by the macroscopic model of Blocki et al. [8].

In this work we extended the investigated reactions to the most massive symmetric systems for which fusion has been measured up to now. As a clear sign of fusion in symmetric systems we studied the evaporation-residue cross sections in the reactions of a ^{110}Pd beam with ^{100}Mo, ^{104}Ru and ^{110}Pd targets at the heavy-ion accelerator UNILAC at GSI. In order to be able to measure the expected small cross sections we made use of the techniques developed for the production of superheavy elements. We used isotopically enriched material for the ion sources (94.6% ^{110}Pd) to increase the beam intensity. The targets were mounted on a rotating wheel to reduce thermal stress. The evaporation residues were separated from the projectiles by the velocity filter SHIP [9] and implanted into an array of semiconductor detectors, where their α decay was observed. The raw α spectra were transformed to production rates taking the decay energies and branching ratios (see [1]) of the known nuclei into account. Absolute cross sections were determined with the help of the measured rates of elastically

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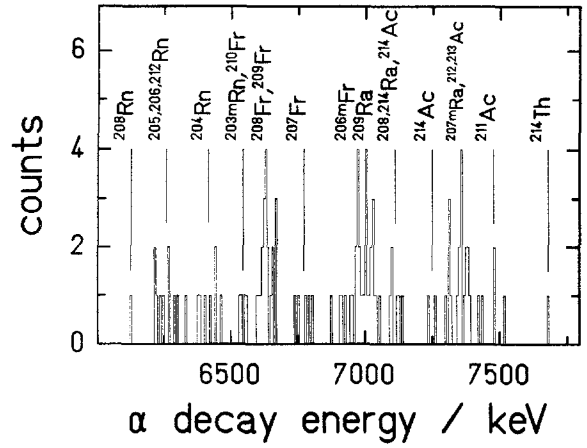

Fig. 1. α decay spectrum recorded in the reaction ^{110}Pd + ^{110}Pd at a center-of-mass energy of 267 MeV

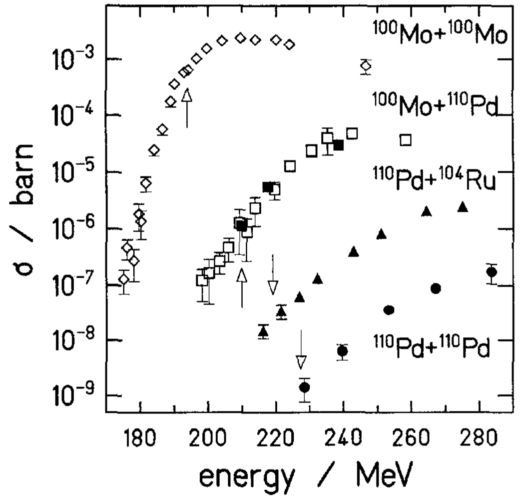

Fig. 2. Total evaporation-residue cross sections for the systems measured in this work (full symbols) and from [2] (open symbols) as function of center-of-mass energy. The arrows indicate the Bass barriers of the systems

scattered projectiles and a calculation for the transmission of the velocity filter SHIP.

In the case of ^{110}Pd + ^{110}Pd the observed α lines correspond to $yp-xn$-($y=2, \dots 5$; $x=1, \dots 5$), $1\alpha-yp-xn$-($y=0$, 1, 3; $x=0, \dots 5$) and $2\alpha-yp-xn$-channels ($y=0$, 1; $x=0, \dots 5$), while the xn-channels were not observed. The number of evaporated particles corresponding to the observed evaporation residues fits to the regular dependence of the number of evaporated particles on the excitation energy of the compound nucleus as observed for lighter systems. At the lowest center-of-mass energy of 288 MeV corresponding to an excitation energy of 28 MeV for the compound nucleus ^{220}U, a total number of 6 events corresponding to the $2p-1n$-channel were identified, thus providing clear evidence that fusion took place. An example of an α decay spectrum with an energy resolution of 60 keV FWHM is shown in Fig. 1. The results for the total evaporation-residue cross sections are shown in Fig. 2 together with some other systems. In spite of the predicted extra-push of 60 MeV [8] the system fused even at the one-dimensional barrier as calculated with the nuclear Bass potential [10]. In Fig. 2 the systems ^{100}Mo + ^{100}Mo, ^{110}Pd measured previously [2] with a ^{100}Mo beam are shown, too. The system ^{100}Mo + ^{110}Pd was also measured in this work with a ^{110}Pd beam for three energies and the cross sections were found to be in agreement with the results of Quint et al. [2]. When comparing these systems we see two main features arising with increasing mass of the systems. The slope of the excitation functions at the low-energy side is decreasing, and at the same time the distance in energy between the position of the maximum of the cross section and the calculated fusion barrier as calculated from the Bass potential is increasing.

In the conventional interpretation of fusion cross sections it is assumed that the formation of the composite system and the subsequent deexcitation process are decoupled by the intermediate stage of a compound nucleus [11]. The cross sections for different reactions and energies can then be calculated by using evaporation codes based on the statistical model. These calculations were applied to lighter systems e.g. ^{90}Zr + ^{89}Y, ^{90}Zr [4, 12] in order to compare the experimental data in detail with the statistical model predictions. The excitation functions for all evaporation channels, including the radiative-fusion channel [4], as well as the spectra of the γ's [13], α's and protons [12] emitted from the compound nucleus could be explained by the statistical model. The only deviation is a small surplus of low-energy protons in the measurements, which could not be explained by the calculation. It may be concluded that the compound-nucleus model is able to describe these medium-heavy systems and can serve as a basis for our heavy systems. In the framework of this model, the decreasing slope of the excitation functions and the increasing shift of the maximum cross-section relative to the Bass barrier with increasing mass may be attributed to an increased fusion-barrier fluctuation and fusion hindrance. In the following it will be discussed whether the compound nucleus model is applicable to the present new, heavy systems, too.

To test the compound-nucleus theory we use our detailed information about the population of the different evaporation channels. In Fig. 3 relative element distributions measured at an excitation energy $E^* \approx 40$ MeV are compared with an evaporation calculation performed with the evaporation code HIVAP [14]. As can be seen, the calculation fits the data points in the system ^{110}Pd + ^{100}Mo, whereas in ^{110}Pd + ^{104}Ru the xn-channels are strongly overestimated by the calculation. In the ^{110}Pd + ^{110}Pd system, the measured element distribution is shifted to nuclei with lower Z suggesting that more charged particles are evaporated. The same effect has been observed in the two heaviest systems also for all other measured energy points ranging from excitation energies of 28 to 84 MeV. Similar deviations were also found previously in Ar- [15] and Sn-induced [1] reac-

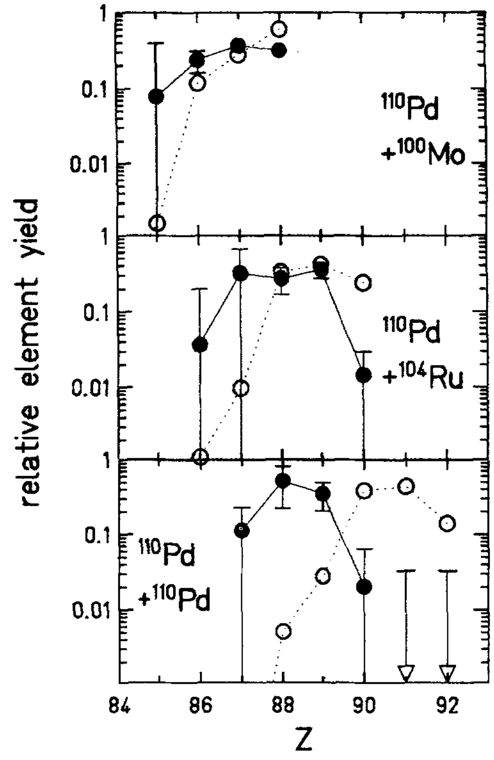

Fig. 3. Relative element distributions from experiment (full points) and HIVAP calculation (open points and dotted lines) for the indicated systems around 40 MeV excitation energy. The arrows indicate upper limits. The element yields are normalized to the sum over all measurement elements

tions leading to the compound nucleus Th. In order to draw reliable conclusions from these observations we investigated the dependence of the calculations from the parameters used in the evaporation code and from the code itself.

For the ^{90}Zr induced reactions mentioned above the spin distribution of the compound nucleus can be fitted to reproduce the fusion excitation functions [4, 12]. For our heavy systems exhibiting a strong extra-push the influence of the fusion and the deexcitation process on the very small evaporation-residue cross sections cannot be disentangled unambiguously. However, due to the high fission probability in these heavy systems the angular momentum ℓ contributing to the evaporation-residue cross section is limited to almost central collisions ($\ell \leqq 15$). The relative population of the different evaporation residue channels as calculated with the statistical model shows no dependence on the initial spin distribution of the compound nucleus. The distribution is determined by the survival probability which depends on the fission barriers, the transmission coefficients of the charged particles as well as the level densities for the different nuclei. It is not possible to reproduce the measured data by changing these parameters and to reproduce the energy dependence of the different channels at the same time. The deviations are also seen with the Monte-Carlo evaporation code CODEX, which was used successfully in the discussion of the cross sections and charged particle spectra of the system ^{89}Y$+^{90}$Zr [12]. Based on this evidence it must be concluded that heavy-ion fusion reactions leading to systems with $Z \geqq 90$ are incompatible with the compound-nucleus model.

We now want to introduce a process which may be able to account for the measured deviations. For the compound nucleus ^{220}U at $E^* = 84$ MeV corresponding to a center-of-mass energy of 284 MeV or 56 MeV above the barrier according to the Bass potential the compound nucleus lifetime may be calculated in the statistical model to be 4×10^{-20} s. In comparison, the transition time for the system ^{110}Pd$+^{110}$Pd from the contact point to the fission saddle point may be estimated from a macroscopic model to be of the same order of magnitude [8]. Thus, the clear separation into a formation and a deexcitation stage is no longer possible, and light particle like neutrons, protons and α particles may be evaporated already during the transition from the contact to the saddle point. In contrast to the preequilibrium emission of high-energetic light particles which was observed at high relative velocities of the reaction partners [16], the precompound emission we propose here is based on full thermalization of the available deformation-dependent intrinsic excitation energy. This precompound emission presumably occurs for all fusing systems at sufficiently high bombarding energies, although the impact on the evaporation-residue cross section is usually very weak. For the massive symmetric systems considered here, however, the hindrance of fusion as predicted by the extra-push model [8] acts as a filter which suppresses specific evaporation channels and leads to the observed drastic effects. In the framework of the extra-push model [8] with a predicted extra-push of 60 MeV it may be assumed that our system evolves towards reseparation after contact. Its only chance to survive is to evaporate an α particle before the saddle point is passed towards reseparation because then the new system has a higher fission barrier and the saddle point moves more outwards towards the contact point, which enables the system to pass towards the spherical compound nucleus. The expected even more abundant precompound emission of the neutrons and protons does not have the impact on the height and the location of the fission barrier which is necessary for the system to proceed towards amalgamation. The precompound α emission immediately explains the fact that no xn- and pxn-channels have been observed experimentally.

A simple calculation was performed for the highest measured energy point at $E^* = 84$ MeV in order to check whether this model is also able to predict the magnitude of the observed cross section. We calculated the α-decay width for different deformations of the system between the contact and saddle point using the average excitation energy of the system on the trajectory between these two points. The transition time for this trajectory was taken from a macroscopic model [8]. The probability p for the evaporation of an α particle from the ^{220}U nucleus on its way to the saddle point was thus found to be $p \approx 10^{-3}$. After evaporation of this α particle we assume the formation of a ^{216}Th compound nucleus. The probability to form evaporation residues from ^{216}Th was taken from the measured cross section in an unhindered Ar-induced reaction to be $p=10^{-2}$ [15]. The product of these probabilities with the fusion cross section of $^{110}Pd+^{110}Pd$ calculated from the geometrical cross section in a sharp cut-off approximation ($\ell_{\mathrm{krit}}=20$) [1] gives 2×10^{-7} barn. This is in good agreement with the cross section of $1.5\pm0.6\times10^{-7}$ barn measured for this energy.

The idea of a precompound emission of light particles as proposed here is equivalent to a generalized view of the diffusion model of Grangé et al. [17] who pointed out that the dissipation during the transition from the compound nucleus to the fission-saddle configuration prevents the fission probability to be treated by the statistical model. In an extended model description, the whole dynamic evolution of the nuclear system from the contact point towards amalgamation as well as fission during the deexcitation process has to be treated as a diffusion process which is in competition with particle evaporation.

Our simple model cannot replace a complete theory for the fusion process which is not available up to now, and it does not explain all features. It shows, however, that the precompound evaporation of α particles may be an important mechanism in heavy-ion fusion which contributes strongly to the evaporation cross section.

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