Mass distributions in the fission of medium-heavy and light nuclei

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The 600 MeV proton-induced fission process of Ce and Yb has been investigated with the double-kinetic-energy method. Mass distributions of these and other medium-heavy and light nuclei are compared to predictions of macroscopic models. No sign of the Businaro-Gallone point, below which the saddle point becomes unstable towards mass asymmetric deformations, is observed. For Ce a stable asymmetric mass distribution is found which may be explained in terms of shell structure effects.

NUCLEAR REACTIONS, FISSION Ce(p, f), Yb(p, f), E = 600 MeV; measured angular correlations, fragment-fragment coin.; deduced TKE, mass distribution.

Fission characteristics of medium-heavy and light nuclei may be calculated with macroscopic models such as the liquid drop model.¹ A common feature of these models²⁻⁵ is the prediction of the existence of a limit below which the symmetric saddle point becomes unstable towards mass asymmetric distortions. The location of this limit, the Businaro-Gallone point, is in all models close to x = 0.4, where x is the fissility parameter.¹ Due to differences in the evaluation of x in the various models, the mass number $A_{\rm BG}$ corresponding to x_{BG} differs; thus, A_{BG} is expected to appear somewhere in the mass region from 100 to 140. Apart from this difference, other model predictions, such as the isospin dependence and the heights of fission barriers, also differ. The models are frequently used in the actinide and superheavy element regions in connection with the macroscopic-microscopic method.¹ In view of the widespread use of the macroscopic models, it is of importance to obtain experimental data for testing various predictions of the models. In this article we present experimental results relevant to the question of the location of the Businaro-Gallone point. Furthermore, we report experimental evidence for a new region of stable massasymmetric deformation similar to that found in the actinide region.

The experiment was performed at the CERN synchrocyclotron. Thin samples of Ce and Yb, both of natural composition, were irradiated with 600 MeV protons with beam intensities of 20-50 nA and a duty factor of about 0.5. Sample thicknesses, around $100 \ \mu g/cm^2$, and impurities were determined with the PIXE method.⁶ Two detector arms were used to register coincidences; one

arm was kept fixed at 90° with respect to the direction of the proton beam, the other moved in plane to the desired position. Each arm contained a Si surface barrier detector of the heavy ion type. The detector on the fixed arm was preceded by a start transmission detector consisting of a 20 $\mu g/cm^2$ carbon foil and two channel electron multiplier plates.⁷ On the movable arm, the Si detector was placed inside a 9.4 cm gas ionization counter⁸ with 90% Ar-10% CH_4 gas at a pressure of 20 Torr. The detectors were calibrated with fission fragments and alpha particles from a thin ²⁵²Cf source. The coincidences, defined by signals in both Si detectors above 5 MeV within 100 ns. were stored event by event on magnetic tape. Each event contains two energy signals (Si detectors). one energy loss signal from the ionization chamber, and four time-of-flight signals. The latter signals were measures of the time differences between the start detector and the two Si detectors, between the two Si detectors, and, finally, between the gas ionization counter and the Si detector inside the chamber. Under beam conditions the time resolution for the first three signals was around 2 ns, which, with our time-of-flight distances of 10-20 cm, prevented any accurate velocity measurements. The time information was merely used to reduce the coincidence window and the experiment is thus to be considered as a double-kinetic-energy measurement. The kinetic energies of the coincident fission products were determined from the Si detector and gas ionization detector amplitudes using, for the Si detectors, the calibration procedure given by Kaufman et al.9

The total fission cross section for mediumheavy and light nuclei is of the order of 1 mb.

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FIG. 1. Coincidence rate as a function of the angle φ_{12} between the detectors for 600 MeV proton induced fission of Yb (lower part). Also shown are the extracted excitation energy (central part) and the measured total kinetic energy release (upper part) as a function of the angle between the detectors.

resulting in fission probabilities in the range of 0.0005 to 0.005. It was nevertheless possible to completely reject background events employing the time-of-flight information and the energy loss measurements.

Some characteristic features of the 600 MeV proton-induced fission process in medium-heavy and light nuclei are illustrated in Fig. 1 with data obtained for Yb. The reaction process is considered to consist of two steps. The fast intranuclear cascade, leaving residuals with broad distributions in mass and charge and in imparted excitation energy, linear and angular momenta, is followed by the slow evaporation step in which fission may compete. The in-plane angular correlation curve (lower part of Fig. 1), which proves the binary character of the process studied, reflects the influence of experimental conditions, reaction steps prior to fission, and the fission process. The shift in the position of the center of gravity from collinearity (180°) is caused by linear momentum deposition in reaction steps prior to fission. An analysis of the contributions to the width of the correlation curve indicates that the width is mainly determined by experimen-

tal conditions and the distribution of linear momentum imparted in the cascade.¹⁰ From the close connection between imparted linear momentum and excitation energy it is possible to extract the mean excitation energy $\langle E_c^* \rangle$ of cascade residuals leading to fission at different angle settings between the detectors. The procedure is somewhat model dependent, as discussed in detail in Ref. 10. The values obtained for Yb are shown in the central part of Fig. 1. The excitation energy increases from around 200 MeV at collinearity to a maximum value of some 400 MeV. This increase is accompanied by a decrease in the cascade residual mass, whereas the charge is almost constant.¹¹ The upper part of Fig. 1 shows the corresponding average values of the total kinetic energy release. The decrease is caused by the combined effects of decreasing average mass and charge and increasing excitation energy $\langle E_F^* \rangle$ of the fissioning nuclei. (Note that $\langle E_F^* \rangle$ is around 50-100 MeV lower than $\langle E^* \rangle$.)

For Ce an angular correlation measurement was prevented by limited irradiation time. The data reported for Ce was taken with $\varphi_{12} = 165^{\circ}$ between the detectors, a value which corresponds to the maximum coincidence rate for the neighboring element La.¹¹

Masses were determined from the single product kinetic energies E_1 and E_2 using the approximate relation $U = M_1/A_F = E_2/(E_1 + E_2)$, where A_F is the



FIG. 2. The distribution of relative masses obtained in the 600 MeV proton-induced fission of Yb (lower part) and Ce (upper part).

average mass number of the fissioning nucleus. The resolution in the relative mass is about 0.07 for Ce and 0.06 for Yb, as estimated from the variances observed for E_1 and E_2 .¹² The distribution of relative masses for Yb and Ce are shown in Fig. 2. For Yb we obtain a symmetric, Gaussian shaped distribution, in agreement with the prediction of the liquid drop model calculations of Nix.² For Ce, the observed mass distribution is wide, non-Gaussian shaped, with a clear indication of a stable asymmetric deformation. We have earlier observed a similar distribution for the element La.¹³ The Ce distribution is somewhat cut off for small values of U, an effect of the corresponding cut off in the E_2 distribution caused by losses of low energy products in the gas ionization counter. Below, we will first discuss the variation of the widths, [full width at half maximum (FWHM) of the distributions of relative masses U and then the distributions for Ce and La.

Experimental values of the mass distribution widths are compared to calculated values in Fig. 3. The liquid drop model calculations of Nix² are employed; however, the other macroscopic models are expected to show the same general trend, i.e., a sharp increase in the calculated width in the vicinity of the critical Businaro-Gallone limit. For Z^2/A values below 30 the experimental values are rather constant and the three points below $Z^2/A = 22$ show widths much smaller than those calculated. The experimental data thus seems to exclude the existence of the Businaro-Gallone point, at least in the mass region investigated.



FIG. 3. Experimental values of the relative widths of mass distributions in the fission induced by 600 MeV protons (Ref. 13 and present work, circles) and 1 GeV protons (Ref. 14, squares). The solid curve is calculated with the liquid drop model (Ref. 2) in which the critical Businaro-Gallone point is located at Z^2/A around 20.

In an analysis of heavy-ion fusion-fission data a similar conclusion is drawn. With ³²S ions Oeschler *et al.*¹⁵ found, for decreasing mass of the composite system, a gradual transformation from symmetric to asymmetric mass division. This is, however, not related to the Businaro-Gallone limit. Instead, the transformation signifies the increasing importance of deep inelastic processes with varying degrees of thermalization.

The mass distributions of all elements displayed in Fig. 3, apart from La and Ce, are symmetric, in agreement with calculations² for nuclei above the Businaro-Gallone limit. For La and Ce we observe stable asymmetric mass distributions which, however, does not indicate that the critical limit is close to these elements, since the Businaro-Gallone limit is connected with an unstable asymmetry with increasing yield for increasing mass asymmetry. Stable asymmetric mass distributions may in the actinide region be correlated to the occurrence of special levels, e.g., in a Nilsson level diagram.^{16, 17} These levels, of opposite parity obeying certain selection rules, show a periodic shell effect and it has been suggested that the mass asymmetry might show a similar periodicity.¹⁶ Thus Johansson¹⁸ has pointed out that neutron numbers around N = 78may favor mass asymmetric divisions. Also, more complete calculations¹⁹ have recently shown that for certain deformation values (ϵ_2 around 0.9) at N = 80 the energy gain for asymmetric fission is about 10 MeV compared to symmetric fission. These neutron numbers around N = 78 - 80are close to those expected in the fission of La and Ce, i.e., with the region where we observe the effect experimentally. Fission-spallation competition calculations¹¹ thus yield average values of N = 55, 74, and 88 for the fissioning nuclei in the cases of Ag, La, and Tb, respectively. The apparent agreement between theory and experiment may be fortuitous, however, as the deformation parameters of the saddle point are not well known, and also, the excitation energies of the fissioning nuclei, around 200 MeV,¹¹ seem too high to allow shell effects to survive.

This work has been supported by the Swedish Natural Science Research Council and in part by the Norwegian Research Council for Science and the Humanities. We are indebted to S. A. E. Johansson, I. Ragnarsson, and S. Åberg for helpful discussions, to several members of the ISOLDE collaboration at CERN for help on numerous occasions, and to the synchrocyclotron staff for the excellent beam quality achieved. We thank B. Sundqvist for providing the thin plastic windows used in the gas ionization chamber.

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