Fission of ²³⁸U induced by inelastic scattering of 120 MeV α particles

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The fission decay of ²³⁸U has been measured as a function of excitation energy in inelastic scattering of 120 MeV α particles. Total kinetic energies and masses of fission fragments were measured by the double energy method. It is observed that the total kinetic energy E_{κ} decreases and that the valley in the mass distribution is reduced when the excitation energy of the system is increased. No indication of anomalous total kinetic energy release in the region of the giant quadrupole resonance has been found. A qualitative interpretation of the data is given on the basis of a static scission point model.

NUCLEAR REACTIONS, FISSION ²³⁸U($\alpha, \alpha' f$), E = 120 MeV, $\theta_{\alpha} = 16^{\circ}$; measured total kinetic energy and fission fragment mass distributions as a function of excitation energy.

I. INTRODUCTION

One of the long standing questions in fission research is concerned with the descent from the saddle point to scission. Whether the available energy is transferred into internal degrees of freedom or into relative motion of the two nascent fragments prior to scission is still largely unknown, since measurements of final parameters of the reaction do not directly address the question of the dynamical evolution of the fission decay.

However, some success in explaining many of the features of fission has been obtained with a static model¹ of the scission point configuration, which takes into account the shell structure in the nascent fragment. Most prominently, it appears that the deformed shell for A = 138 is responsible for the strongly asymmetric mass distributions in fission of most actinide elements. A strong spherical shell closure at Z = 50, N = 82 is believed to be associated with the decrease of the total kinetic energy with excitation energy as observed in several light ion and neutron induced reactions on uranium and plutonium targets.²⁻⁶

To further study this latter correlation, we have measured the fission decay of 238 U induced by the (α, α') reaction. We find that the total kinetic energy decreases with excitation energy at a rate of $dE_K/dE^* = -0.38 \pm 0.07$ and that the effect is concentrated in the heavy fragment mass region of M_H = 125-135.

II. EXPERIMENTAL PROCEDURE AND DATA ANALYSIS

A beam of 120 MeV α particles from the 88" cyclotron at Lawrence Berkeley Laboratory was used to bombard a 530 μ g/cm² thick, self-supporting metallic ²³⁸U target. Inelastically scattered α particles were identified in a triple telescope of solid state detectors located at $\theta = 16^{\circ}$ out of the horizontal plane. Two fission detectors were placed at $\theta = 90^{\circ}$ and $\theta = -90^{\circ}$ in the horizontal plane, each subtending a solid angle of $\Omega_f \simeq 440$ msr. Coincident events between the telescope and the two fission detectors were recorded on magnetic tape for subsequent analysis.

The elements of the detector telescope were gain matched by introducing a calibrated charge pulse on the detector side of the preamplifiers. The energy calibration was obtained from the elastic peak in the α spectrum. The fission detectors were energy calibrated with the fission fragments from a thin ²⁵²Cf source.⁷

The data were analyzed off line by an event-byevent reconstruction of the kinematics. Identification of α particles in the telescope was obtained by generating a particle identification spectrum from the measured pulse heights in each element in the detector telescope and selecting events in the peak corresponding to α particles. The primary energies and masses of the fission fragments were calculated by an iterative procedure, taking

into account the emission of neutrons from the fragments and the mass dependent pulse height defect of the detector response. The mass dependent neutron emission function measured for ²³⁵U(n,f) was used.⁸ The excitation energy dependence of the neutron emission was ignored in the present analysis. The effects of this omission on the final results are discussed in a following paragraph. The main contributions to the mass resolution of the experiment come from the intrinsic detector resolution and the large solid angle of the fission detectors, which combined give an estimated mass resolution of $\Delta M = 5$. The experimental mass spectra have been unfolded assuming this mass resolution. The data have been corrected for accidental coincidences, which amount to $\sim 10\%$ in the energy region above the fission threshold. Because a metallic self-supporting target was used, we are not faced with the problem of strong accidental peaks from scattering off target impurities. Due to the out-of-plane geometry used in the present experiment, there is no contribution from coincidences with recoil nuclei from light target contaminants.

III. RESULTS AND DISCUSSION

The total kinetic energy release E_K , as a function of excitation energy in the fissioning nucleus, and the spectrum of inelastically scattered α particles in coincidence with fission are shown in Fig. 1. We observe a strong increase in the fission coincidence rate up to the threshold for the neutron emission B_n , beyond which the competition



FIG. 1. The average total kinetic energy [(a)] and the fission coincidence spectrum [(b)] are shown as a function of the excitation energy. The neutron binding energy and thresholds for second and third chance fission are indicated by arrows.

from neutron emission introduces a sharp decrease. The increase in the fission yield due to second and third chance fission is clearly visible in the data.

In the energy region from 5–12 MeV, where only first chance fission is allowed, a strong decrease in the average total kinetic energy \overline{E}_K is observed. Rather sharp increases in \overline{E}_K are also observed above the thresholds for second and even third chance fission. These increases in \overline{E}_K most likely reflect the fact that a large fraction of the fission events above these thresholds come from near barrier fission of ²³⁷U and ²³⁶U, respectively, which have high total kinetic energies. At excitation energies above ~25 MeV a weak increase in the total kinetic energy with excitation energy is seen.

The best fit to the total kinetic energy in the excitation energy region $E^* = 5 - 12$ MeV is represented by a solid line with a slope of dE_K/dE^* $= -0.46 \pm 0.07$ in Fig. 1. When corrected for the increase in neutron evaporation with excitation energy, this result is reduced to $dE_K/dE^* = -0.38$ \pm 0.07. This value is significantly lower than the result of David *et al.*⁵ who find a slope of dE_K/dE^* $= -0.8 \pm 0.3$. The origin of this discrepancy is not known at present. Decreasing total kinetic energies have been observed in several other uranium⁵ and plutonium^{2^{-4}} nuclei, although there is a sizable spread in the values of dE_K/dE^* derived from various experiments. Negative slopes dE_{κ}/dE^* have also been found in several Ac isotopes,⁶ however, only for the asymmetric component of the distribution.

A rather strong excitation probability of the giant quadrupole resonance has been observed in the singles ${}^{238}U(\alpha, \alpha')$ reaction⁹ under experimental conditions almost identical to the ones of the present study. Although there is still some question about the fission probability of the giant quadrupole resonance, 9^{-12} it appears that it is comparable to that of the underlying background (which is $P_f \sim 0.2$), which indicates that a substantial fraction of fission events in the energy region $E^* \sim 8-12$ MeV can be associated with the giant quadrupole resonance. However, this does not appear to have any observable effect on the total energy release, which has a dependence on excitation energy very similar to what is observed in other reactions where the giant quadrupole resonance is not excited, e.g., $^{239}Pu(n,f)$ (Ref. 4) and 239 Pu(d, pf).³

The slope dE_K/dE^* over the excitation energy region $E^*=5-12$ MeV is plotted as a function of the mass of the heavy fission fragments in Fig. 2(a). A clear mass dependence of the effect is apparent, most of the effect being correlated with



FIG. 2. Fragment mass dependence of the slope dE_K/dE^* [(a)] and the average total kinetic energy \overline{E}_K [(b)] in the excitation energy region from 5-12 MeV. The mass spectrum of heavy fragments is also shown [(c)].

masses near $M_H = 128 - 130$. The total kinetic energy averaged over the same excitation energy region also displays a strong dependence on the fission fragment mass, with the maximum occurring at $M_H \sim 130$ as shown in Fig. 2(b). However, the most abundant masses of heavy fission fragments are found at $M_H \simeq 138$ [see Fig. 2(c)], which is distinctly different from the peaks in the dE_K/dE^* and \overline{E}_K data.

The dashed curve in Fig. 2(b) represents a calculation of the Coulomb repulsion energy between two coaxial ellipsoids, each deformed to a semiaxis ratio of b/a = 0.6 and separated by d = 2 fm, in order to simulate a neck formation between the two fragments. This comparison was performed only to show the expected dependence on fragment mass for a fixed shape of the nascent fragments. The parameters used are chosen only to give an approximately correct magnitude of the total kinetic energy and should not be considered as measured quantities.

Some qualitative insight into these shell effects may be obtained from a simple static model of the scission point configurations, which includes the effects of nuclear shells in the nascent fragments as proposed by Wilkins *et al.*¹ It appears that the large values of \overline{E}_{K} and $d\overline{E}_{K}/dE^{*}$ found in the region $M_H = 130$ are correlated with the doubly magic shell for spherical shapes at Z = 50 and N = 82. Spherical fragment shapes at scission lead to large kinetic energies due to increased Coulomb repulsion. Although this minimum in the shell energy is quite deep, it is not the main factor in determining the fission mass distribution, which peaks around $M_{H} \simeq 138$. As is the case for most actinide nuclei in the region from Th to Cf, the mass distribution can be explained at least qualitatively by the occurrence of a strong deformed neutron shell at N = 88. The strong Coulomb repulsion between the two nascent fission fragments at scission clearly favors the somewhat weaker minimum in the shell energy surface at large deformation over the strong spherical minimum for the heavy fragment. Studying the \overline{E}_{K} dependence on the excitation energy E^* , we see that the A = 138 shell is very stable, whereas the A = 132spherical shell rapidly disappears with excitation energy. A possible explanation of this phenomenon is that as the excitation energy is raised above the fission barrier, breaking of pairs occurs, which increases the viscous heating during the descent towards scission. Because the spherical minimum in the shell energy occurring at N = 82 is superimposed on a steep slope of the liquid drop potential energy surface, this minimum is rapidly destroyed with excitation energy. Contrasting this, the deformed shell at N = 88 has a substantial stability against excitation energy because it coincides in deformation with the minimum in the liquid drop potential energy surface (see Ref. 1).

This stability of the A = 138 deformed shell is also apparent in Fig. 3, where the fission fragment mass distributions are shown for several excitation energy bins. The strongly asymmetric character is preserved at least up to apparent excitation energies of 50 MeV. However, the excitation energy may not be as high as the energy of the outgoing α particle would indicate, because of (1) prefission neutron emission and (2) the contribution from the $(\alpha, {}^{5}\text{He})$ reaction, which is identified as an (α, α) reaction, due to breakup of the



FIG. 3. Fission fragment mass distributions are shown for different excitation energy bins.

⁵He ejectile.¹³ This interpretation is supported by the comparison with mass distribution data obtained in the ²³⁸U(α , f) reaction,¹⁴ as illustrated in Fig. 4. For the ²³⁸U(α , α f) reaction we observe an



FIG. 4. Peak-to-valley ratios for the measured fission fragment distributions (closed circles) are compared to results obtained in the $^{238}U(\alpha, f)$ reaction (Ref. 14) (open circles).

increasing abundance of symmetric mass splits with increasing excitation energy to $E^* \sim 30$ MeV, beyond which point the peak-to-valley ratio remains almost constant. This saturation effect is not observed in fission of ²⁴²Pu induced by the ²³⁸U(α , f) reaction,¹⁴ where the fusion-fission process clearly dominates.

The standard deviations σ_{E_K} of the total kinetic energy distributions are shown in Fig. 5 as a function of mass for different excitation energy inter-



FIG. 5. The standard deviations of the total kinetic energy distributions are shown as a function of heavy fragment mass for different excitation energy bins.

vals. A strong decrease in σ_{E_K} is observed for increasing mass of the heavy fragment, especially in the lower excitation energy bins. The large width of the E_K distribution at $M_H = 120-125$ is correlated with the coexistence of three competing neutron shells for $N \sim 80$ with very different deformations of $\beta = 0.1$, 0.5, and 0.85, respectively.¹ In the region of heavy fragment masses from M_H = 138-160 a strong neutron shell at N = 88 with a deformation of $\beta = 0.65$ is dominating the scission point shapes and resulting in narrower total kinetic energy distributions, which is also observed in the data. Again we observe that these shell effects are somewhat reduced with excitation energy.

A static description of the scission point configuration as implied in the above discussion is certainly an oversimplification. However, even a dynamical description of the descent from saddle to scission must include the shell effects in the nascent fragments, and we therefore expect that such a calculation would lead to conclusions qualitatively similar to the ones presented here.

IV. CONCLUSIONS

From the present study of fission of ²³⁸U induced by inelastic scattering of α particles we conclude that the effect of nuclear shells in the nascent fragments at the scission point are of major importance for both the kinetic energy release and the mass division in fission. Although a strong deformed neutron shell at N = 88 forces the nucleus to split preferentially into fragments of mass A = 138 and A = 100, we observe that the disappearance of a somewhat weaker (when imposed on the repulsive Coulomb potential) spherical shell at A = 132 leads to a strong decrease of the total kinetic energy, when the nucleus is excited above the fission threshold.

Although it is known that a substantial fraction of the inelastic cross section at $E^* \simeq 10$ MeV goes into excitation of the giant quadrupole resonance,¹² we have no evidence of anomalous kinetic energy release in this region. On the contrary, the observed slope of $d\overline{E}_K/dE^*$ is in very close agreement with measurements of neighboring nuclei using (d, p) reactions at lower bombarding energies, where the giant quadrupole is not excited to any degree.

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