## Three fission modes of <sup>220</sup>Ra

I. V. Pokrovsky,<sup>1</sup> L. Calabretta,<sup>2</sup> M. G. Itkis,<sup>1</sup> N. A. Kondratiev,<sup>1</sup> E. M. Kozulin,<sup>1</sup> C. Maiolino,<sup>2</sup> E. V. Prokhorova,<sup>1</sup>

A. Ya. Rusanov,<sup>3</sup> and S. P. Tretyakova<sup>1</sup>

<sup>1</sup>Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research, RU-141980 Dubna, Moscow region, Russia

<sup>2</sup>Laboratorio Nazionale del Sud, Istituto Nazionale di Fisica Nucleare, I-95123 Catania, Italy

<sup>3</sup>Institute of Nuclear Physics of the National Nuclear Center of Kazakhstan, 480082 Alma-Ata, Kazakhstan

(Received 29 March 1999; published 8 September 1999)

Mass-energy distributions of the <sup>220</sup>Ra fission fragments produced in the <sup>12</sup>C+<sup>208</sup>Pb reaction at projectile energies higher and deep below the Coulomb barrier have been measured using a two-arm time-of-flight spectrometer CORSET. It has been shown that an asymmetric component appears at the sides of the prevailing symmetric fission with a cooling of the compound nucleus. The component consists of two standard modes with average masses of  $\approx$ 132 and  $\approx$ 139. A three-component structure has also been observed in the total kinetic energy distributions in the range of asymmetric fission fragment masses. [S0556-2813(99)50909-6]

PACS number(s): 21.10.-k, 25.85.-w, 27.90.+b

It is well known from numerous experimental studies that actinides with masses of up to  $A \approx 256$  mainly undergo asymmetric fission, i.e., there are two-humped mass-energy distributions (MED) of fission fragments with a minimal yield for fission into two equal parts [1,2]. In contrast, nuclei in the Pb region mainly undergo symmetric fission but the asymmetric component is also present and its contribution does not exceed 0.5%. It decreases with a decrease in A of a fissile nucleus [3]. The fission by the two modes simultaneously is typical for the region of <sup>225</sup>Ra-<sup>228</sup>Ac at medium excitation energies, with a predominance of the asymmetric fission mode in the case of energies near the barrier [4]. As for the fission modes, the domain of nuclei with A =214-224, which conventionally can be called intermediate, has been practically unstudied until recent. Only in Ref. [5] an attempt was made to investigate the MED in the reactions <sup>7</sup>Li+<sup>209</sup>Bi and <sup>12</sup>C+<sup>208</sup>Pb at medium excitation energies. However, no evidence concerning the asymmetric fission was obtained, though in the energy distributions, namely, in the total kinetic energy distributions and its dispersion some irregularities were observed connected with a possible manifestation of the asymmetric mode.

Several years ago, experiments on the investigation of different fission properties for the intermediate region of nuclei were carried out first in Dubna, then continued in Catania, Grenoble, and Strasbourg. The MED of fission fragments of the compound <sup>219</sup>Ac and <sup>220,224,226</sup>Th nuclei formed in reactions with <sup>16,18</sup>O ions were studied at energies near and below the Coulomb barrier [6–9]. The multiplicities of pre- and post-neutrons and gamma quanta from the fission fragments produced in the fission of <sup>226</sup>Th [10,11] were also studied.

Approximately at the same time, at GSI (Darmstadt) experiments started aiming at the study of fission fragment charge distributions of secondary radioactive beams of nuclei from <sup>214</sup>Ra to <sup>234</sup>U occurring via the giant dipole resonance as a result of electromagnetic interaction between the ion beam and lead target nuclei.

The experiments [6-14] showed that it is in this region of

nuclei that a transition from the predominantly symmetric to predominantly asymmetric fission takes place with an increase in A of the fissile nucleus at low excitation energies.

For the intermediate region of nuclei, theoretical calculations of the potential energy surface using a method of shell corrections as a function of the mass-asymmetric deformation were made in Refs. [8,9,15-17]. The calculations of [8,9,17] showed that, first of all, for the isotopes  $^{220-232}$ Th, there are symmetric and asymmetric valleys that are clearly divided by the high potential hump. The last-mentioned vallev consists in its turn of two components, although the barrier between them is not so distinct. Second, the saddle points for the symmetric and asymmetric fission, determining the population of the valleys, are different in height as well as in deformation. Third, with increasing the Th nuclear masses from 220 to 232, the sign of the difference in the saddle point heights changes. For light isotopes it is  $E_f^a$  $-E_f^s > 0$ , whereas for heavy ions the picture is the reverse, which agrees with the experimental results from [6-14].

The present study continues a series of our studies devoted to the investigation of the fission process in the intermediate region of nuclei, and here we present some experimental results on the MED of the <sup>220</sup>Ra fission fragments formed in the reaction <sup>12</sup>C+<sup>208</sup>Pb at <sup>12</sup>C projectile energies of near and below the Coulomb barrier.

The experiment was carried out at a tandem accelerator of the INFN, Catania (Italy) at the <sup>12</sup>C ion beam energies  $E_{\rm lab}$ =57, 59, 62, 65, 73, and 90 MeV. The velocities and coordinates of pair fission fragments were measured using a method of kinematic coincidence and with the use of the two-arm time-of-flight position-sensitive spectrometer COR-SET [18], whose arms accepted a solid angle of 360 msr. The two arms were positioned at the angles  $\Theta_1 = 64^\circ$  and  $\Theta_2 = 102^\circ$  of the laboratory coordinate system. The mass resolution of the spectrometer was 3–4 u. and the position resolution was  $\pm 0.1^\circ$ . The characteristics of the spectrometer, the method of measurement and results of data procession are presented in more detail in [18,19]. The target was a



FIG. 1. From top to bottom: fission fragment mass yields, TKE(M), and  $\sigma_{\text{TKE}}^2(M)$  distributions for the three indicated energies  $E_{\text{lab}}$ . The symmetric component of the mass yields is shown by the solid curve. In the TKE(M) and  $\sigma_{\text{TKE}}^2(M)$  distributions the solid curves are drawn along the experimental points.

<sup>208</sup>Pb layer, 220  $\mu$ g/cm<sup>2</sup> in thickness, delivered to a carbon backing, 50  $\mu$ g/cm<sup>2</sup> in thickness.

It was established that for the <sup>12</sup>C ion energy  $E_{\rm lab}$ = 57 MeV the fission cross section decreased by ~10<sup>5</sup> times compared with the case of the ion energy  $E_{\rm lab}$ = 90 MeV. Due to this fact, for the energy of 90 MeV, 3 ×10<sup>5</sup> fission pair events were registered, whereas for the energies of 59 and 57 MeV—2×10<sup>4</sup> and 1.5×10<sup>3</sup> events were registered, correspondingly.

Figure 1 shows the fission fragment mass yields *Y*, normalized to 200%, as well as dependences of the fission fragment total kinetic energy TKE and its dispersion  $\sigma_{\text{TKE}}^2$  on the fission fragment masses for three projectile energies:  $E_{\text{lab}} = 57$ , 59 and 90 MeV, which correspond to the initial excitation energies of the <sup>220</sup>Ra compound nucleus  $E^* = 21.9$ , 23.8, and 53.1 MeV (without taking into account the emission of prescission neutrons  $\nu_{\text{pre}}$ ). The fission barrier for this nucleus is  $E_f = 12.5$  MeV [20], then the excitation energies above the barrier will be  $E_{sp}^* = 9.4$ , 11.3, and 40.6 MeV, respectively.

It is seen from Fig. 1 that in the fission fragment mass distributions for the two lowest energies at the sides of the prevailing symmetric fission, which was approximated by the Gaussian function in the region of masses from 110 to 125 and the complementary masses, the asymmetric component (fission mode) is distinctly seen in the form of "shoulders." In the TKE distributions and in the dispersion  $\sigma_{TKE}^2$ for the fission fragment range of masses M > 125, an increase in their values is also observed, which is characteristic of the asymmetric fission mode. For the energy  $E_{lab} = 90$  MeV, the fission fragment MEDs are close by their properties to the predictions of the liquid drop model (LDM) [21,22], i.e., to their high temperature limit, when Y(M) is the Gaussian function and TKE(M) is a parabola. However, in the dependence  $\sigma_{\text{TKE}}^2(M)$ , the characteristics sensitive to the presence of different fission modes, there is a peak corresponding to the fission fragment mass 135, which, no doubt, testifies to a presence of the asymmetric mode for that energy also. The presence of the asymmetric mode can only be explained by

PHYSICAL REVIEW C 60 041304



FIG. 2. The extracted asymmetric component  $Y_a$  for two lower energies  $E_{\text{lab}}$  and its description by a sum of two Gaussians.

the fact that real excitation of the fissile nucleus is much less than the initial one due to the emission of a certain number of prescission particles. Unfortunately, a "chance" structure of the fission probability is not yet known for that nucleus, and there are no direct experimental data concerning the prescission neutron multiplicities. However, in accordance with the systematics  $\langle v_{\rm pre} \rangle$  [23,24], <sup>220</sup>Ra nuclei before the scission point must emit on the average 2.0 neutrons  $\langle \nu_{pre} \rangle$  at the initial excitation energy  $E^* = 53.1$  MeV; 0.3 neutrons  $\langle v_{\rm pre} \rangle$  at  $E^* = 23.8$  MeV; and 0.2 neutrons  $\langle v_{\rm pre} \rangle$  at  $E^*$ = 21.9 MeV. The average effective excitation energy after the emission  $\langle v_{\rm pre} \rangle$  can be found proceeding from the correlation:  $E_{\text{eff}}^* = E^* - \langle \nu_{\text{pre}} \rangle \langle E_{\text{pre}} \rangle$ , where  $\langle E_{\text{pre}} \rangle = \langle B_n \rangle + \langle E_n \rangle$ ,  $\langle B_n \rangle$  is the average neutron binding energy for the chain of fissile nuclei, and  $\langle E_n \rangle = 2T_n$  is the average kinetic energy carried away by the neutrons and calculated for the nuclear chain,  $T_n = \sqrt{E^*/a}$  is the temperature of nuclei for which the averaging  $B_n$  is done, a = 0.093A is the parameter of level densities [25]. Thus at  $E_{lab}=90$  MeV, it is not the  $^{220}$ Ra nucleus that undergoes fission but the <sup>218</sup>Ra nucleus with an effective excitation energy of the ground state  $E_{\rm eff}^*$  $\approx$  35 MeV, assuming that all the neutrons  $\langle v_{\rm pre} \rangle$  have been emitted before the saddle point. For the lowest energies at  $E_{\rm lab}$  = 57 and 59 MeV, it is necessary to use not the averaged value  $\langle B_n \rangle$  but the  $B_n$  value of the first neutron, which is quite large for this nucleus and is equal to 7.2 MeV. In that case, the emission of a single neutron with the average energy of  $2T_n$  will lead the nucleus to excitation energies  $E_{\text{eff}}^*$ , which are close to the threshold energies, and the fission probability may fall down sharply. That is why for these energies in fact only the first fission "chance" will be realized, i.e., the initial nucleus <sup>220</sup>Ra will undergo fission, and the MED of the fission fragments will not be distorted by the "chances."

Figure 2 shows the yield of the extracted asymmetric component  $Y_a$  for the two lower excitation energies, ob-



FIG. 3. (a) The dependence of the yield probability ratio  $Y_s/Y_a$  for the nuclei from <sup>204</sup>Pb to <sup>234</sup>U on mass  $A_{CN}$  of the fissile nucleus at excitation energies above the fission barrier  $E_{sp}^*=9-10$  MeV. Open circles represent the data from work [3], the filled circle represent results of the present study; (b) experimental difference of the symmetric and asymmetric barriers  $E_f^a - E_f^s$  for <sup>210</sup>Po and <sup>213</sup>At, heavy isotopes of Ra and Ac [3], and Pa and Np [27].

tained as a result of the difference between the experimental yield  $Y_{exp}$  and the Gaussian yield  $Y_G$ , shown in Fig. 1:  $Y_a$  $= Y_{exp} - \dot{Y}_G$ . It is clearly seen that  $Y_a$  has the complex structure and consists of two asymmetric modes. Such structure is similar to the observed one in the fission of pre-actinide nuclei [3] and <sup>220-226</sup>Th nuclei [6-9]. That is why we described the distribution  $Y_a(M)$  as a sum of two Gaussians using a method of the root mean square:  $Y_a = Y_{a0} + Y_{a1}$ , and did not impose any conditions on the description parameters. The average masses of the components turned out to be  $\langle M_{a1} \rangle \simeq 132$  and  $\langle M_{a0} \rangle \simeq 139$ . In the terminology of Brosa et al. [26], these are the fission modes called standard-I and standard-II, correspondingly. Note that the shape of the fissioning nucleus, corresponding to the fission mode  $a_1$ , is a very compact one, since the fission fragment  $\langle M \rangle = 132$  (Z = 50 and N=82) and the light fission fragment  $\langle M \rangle$ =88 (Z=38 and N=50) are spherical. As will be shown below, it leads to a sharp increase in the TKE for this mode.

Thus, the common properties for the mass distributions of the <sup>220</sup>Ra fission fragments and other more light as well as more heavy nuclei have been established unambiguously.

Figure 3(a) presents the dependence of the ratio between the probabilities of the symmetric and asymmetric fission modes  $Y_s/Y_a$  as a function of the mass number  $A_{CN}$ , for the nuclei from <sup>204</sup>Pb to <sup>234</sup>U at  $E_{sp}^*=9-10$  MeV [3]. The results for <sup>220</sup>Ra of the present study are also shown in the figure. They fit well into the unified dependence. Figure 3(b) shows the experimentally found differences between the asymmetric and symmetric barriers  $E_f^a - E_f^s$  for <sup>210</sup>Po, <sup>213</sup>At, nuclei in the region of heavy isotopes <sup>226</sup>Ra-<sup>228</sup>Ac from work [3], and <sup>233</sup>Pa-<sup>237</sup>Np from [27]. Thus in Figs. 3(a) and 3(b) we can observe a clear correlation between the ratio  $Y_s/Y_a$  and the difference of the barrier height values and we expect that for <sup>220</sup>Ra  $E_f^a > E_f^s$  and the difference will be 0.6–1.3 MeV, which is indicated in Fig. 3(b).



FIG. 4. Distributions of the total kinetic energy N(TKE) at  $E_{\text{lab}}=59$  MeV ( $E^*=23.8$  MeV) for the indicated fission fragment mass ranges and their description by one or three Gaussians, on condition that the areas under each peak are equal to the sum of yields of each component from Fig. 1 and Fig. 2 for the same fission fragment mass range.

Figure 4 presents the TKE distributions at the excitation energy  $E^* = 23.8$  MeV for different ranges of fission fragment masses: near the symmetric peak, where there is no contribution from  $Y_a$ , and for the range of masses from 125 to 135, where all three components are present. It is seen quite clearly that in the last-mentioned case the structure of the TKE distribution is complex, and both asymmetric components are seen in the form of "shoulders" at the right side. This distribution can be very well described by a sum of three Gaussians; the area below each peak is precisely equal to the yield of a corresponding component for this mass range as seen from Fig. 1 and Fig. 2. The modes can be identified with the TKE: for the symmetric mode it is  $\langle TKE_s \rangle \simeq 157$  MeV, for the asymmetric one- $\langle TKE_{a0} \rangle$  $\simeq 168$  MeV, and for the other asymmetric mode—  $\langle TKE_{a1} \rangle \simeq 178$  MeV. Summing up, one can state with certainty that the three fission modes were experimentally observed in the fission of <sup>220</sup>Ra. They are distinctly traced in the mass and energy distributions of fission fragments. Their properties agree well with the earlier known behavior regularities of the fission modes.

This work was supported by the Russian Foundation for Basic Research under Grant No. 99-02-17981 and by INTAS under Grant No. 11929.

- R. Vandenbosch and J. Huizenga, *Nuclear Fission* (Academic Press, New York, 1973).
- [2] F. Gönnenwein, in *Nuclear Fission Process*, edited by C. Wagemans (CRC Press, Boca Raton, FL, 1991), p. 287.
- M. G. Itkis, V. N. Okolovich, A. Ya. Rusanov, and G. N. Smirenkin, Z. Phys. A **320**, 433 (1985); Nucl. Phys. A**502**, 243c (1989); Fiz. Elem. Chastits At. Yadra **19**, 701 (1988) [Sov. J. Part. Nucl. **19**, 301 (1988)].
- [4] H. J. Specht, Rev. Mod. Phys. 46, 733 (1974); Nukleonika 20, 717 (1975).
- [5] B. D. Wilkins, B. B. Back, H.-G. Clerc, J. E. Gindler, B. G. Glagola, and L. E. Glendnin, Lect. Notes Phys. 158, 150 (1982).
- [6] M. G. Itkis, Yu. Ts. Oganessian, G. G. Chubarian, V. V. Pashkevich, V. S. Salamatin, A. Ya. Rusanov, V. N. Okolovich, and G. N. Smirenkin, in *Proceedings of the Workshop on Nuclear Fission and Fission-product Spectroscopy*, Seyssins, France, 1994, edited by H. Faust and G. Fioni (ILL Grenoble, 1994), p. 77.
- [7] M. G. Itkis, Yu. Ts. Oganessian, G. G. Chubarian, V. S. Salamatin, A. Ya. Rusanov, and V. N. Okolovich, in *Proceedings* of the XV EPS Conference on Low Energy Nuclear Dynamics (LEND-95), St. Petersburg, Russia, 1995, edited by Yu. Ts. Oganessian et al. (World Scientific, Singapore, 1995), p. 177.
- [8] M. G. Itkis *et al.*, in *Tours Symposium on Nuclear Physics III*, Tours, France, 1997, edited by M. Arnould *et al.*, AIP Conf. Proc. No. 425 (AIP, New York, 1988), p. 189.
- [9] M. G. Itkis *et al.*, Proceedings of the International Conference on Nuclear Physics (INPC '98), Paris, France, 1998 [Nucl. Phys. A (in press)].
- [10] A. Kelic et al., Phys. Rev. C (to be published).
- [11] G. G. Chubarian *et al.*, JINR Rapid Communication No. 4 [90]-98, 1998; in *Proceedings of the 2nd International Conference on Exotic Nuclei and Atomic Masses (ENAM'98)*, Shanty Creek Resort, Bellaire, Michigan, 1998, edited by B. M. Sherril *et al.*, AIP Conf. Proc. No. 455 (AIP, New York, 1998).
- [12] K.-H. Schmidt et al., Phys. Lett. B 325, 313 (1994).

## PHYSICAL REVIEW C 60 041304

- [13] S. Steinhäuzer et al., in Proceedings of the 3rd International Conference on Dynamical Aspects of Nuclear Fission, Časta-Papiernička, Slovak Republic, 1996, edited by J. Kliman and B. Pustylnik (JINR, Dubna, 1996), p. 151.
- [14] K.-H. Schmidt et al., Nucl. Phys. A630, 208c (1998).
- [15] P. Möller, Nucl. Phys. A192, 529 (1972).
- [16] V. V. Pashkevich, Proceedings of the International School-Seminar on Heavy Ion Physics, Alushta, USSR, 1983 (JINR, Dubna, 1983), p. 405.
- [17] V. V. Pashkevich, in Proceedings of the XVEPS Conference on Low Energy Nuclear Dynamics (LEND-95) [7], p. 161.
- [18] E. M. Kozulin, N. A. Kondratjev, and I. V. Pokrovski, in Heavy Ion Physics, Scientific Report 1995-1996, JINR, FLNR, Dubna, 1997, p. 215; N. A. Kondratiev, E. M. Kozulin, I. V. Pokrovski, and E. V. Prokhorova, in Fourth International Conference on Dynamical Aspects of Nuclear Fission (DANF'98), Časta-Papiernička, Slovak Republic, 1998 (World Scientific, Singapore, in press).
- [19] M. G. Itkis et al., Phys. Rev. C 59, 3172 (1999).
- [20] V. V. Pashkevich (private communication).
- [21] J. R. Nix and W. J. Swiatecki, Nucl. Phys. 71, 1 (1965).
- [22] G. D. Adeev, I. I. Gonchar, V. V. Pashkevich, N. I. Pischasov, and O. I. Serdyuk, Fiz. Elem. Chastits At. Yadra 19, 1229 (1988) [Sov. J. Part. Nucl. 19, 529 (1988)]; Nucl. Phys. A502, 405c (1989).
- [23] E. M. Kozulin, A. Ya. Rusanov, and G. N. Smirenkin, Phys. At. Nucl. 56, 166 (1993).
- [24] M. G. Itkis and A. Ya. Rusanov, Phys. Part. Nuclei 29, 160 (1998).
- [25] A. V. Ignatyuk, K. K. Istekov, V. N. Okolovich, and G. N. Smirenkin, *Proceedings of the 4th International Symposium on Physics and Chemical Fission*, Julich, Germany, 1979 (Vienna, IAEA, 1980), v. 1, p. 421.
- [26] U. Brosa, S. Grossmann, and A. Möller, Phys. Rep. 197, 167 (1990).
- [27] T. Ohtsuki, H. Nakahara, and Y. Nagame, Phys. Rev. C 48, 1667 (1993).