THE COLD FRAGMENTATION OF ^{234}U in ^{233}U (n_{th},f)

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Abstract

The transformation of the uranium nucleus into a separated two fragment configuration has been found to be possible without appreciably heating the fragments via intrinsic excitation or deformation. A small part of the fragmentations in thermal neutron induced fission proceeds with a release of mutual kinetic energy of the fission fragments, which is not far from the total Q-value of each fragmentation[1,2]. 1 permille of all fragmentations occur with an excitation energy smaller than 7 MeV, an excitation energy of the fragments not larger than the excitation energy of the fissioning nucleus after thermal neutron capture.

Experimental Method and Results

This is a report on an experimental investigation of the cold fragmentation of ^{234}U. ^{233}U targets of 40 µg/cm^2 thickness were exposed to a thermal neutron flux of $\sim 7 \cdot 10^{14}$ neutrons/cm$^2 \cdot$s at the High Flux Reactor of the ILL, Grenoble. The fission fragments of the reaction ^{233}U (n_{th},f) were mass and energy separated within microseconds, using the fission product separator "LOHENGRIN" of the ILL[3]. The separation occurs as a function of mass over ionic charge A/q and of the velocity v of the fragments. The mass yields were measured with an ionization chamber[4].

Since "LOHENGRIN" provides mass separated beams at constant kinetic energy E, it is possible to use an absorber at the exit slit of "LOHENGRIN" in which a mass separated fragment loses a certain amount of energy $\Delta E(Z)$ according to its nuclear charge Z. We used a parylene-absorber and measured with the ionization chamber the rest energy $E_R = E - \Delta E$[4]. With this degrader-rest energy technique, which is complementary to a ΔE measurement, it was possible to determine the nuclear charges in the light group of fission products at all masses and energies present. The large sensitive area and the good nuclear charge and energy resolution of the detector system allowed measurements of isotopic yields down to 10^{-4} and of mass yields down to 10^{-6} of the absolute fission yield.

Fig. 1a shows mass distributions of light fragments selected at kinetic energies of 114.1 MeV, 116.1 MeV, and 118.1 MeV. Measurements in steps of 1 MeV for light fragments in the energy region between (112-119) MeV have been made. Fig. 1b gives the maximum possible kinetic energies of light fission fragments using the mass formula of Møller and Nix[5]. Measurements of isotopic yields are available up to E = 112 MeV, for selected chains up to E = 114 MeV. The yields are absolute fission yields and given as $dY/dA \cdot dZ \cdot dE$. Fig. 2 gives as examples the energy dependence of the chains A = 90 and A = 100. We fitted second order polinomials to the logarithmic yields. The finite energy resolution of ±0.5 MeV has been unfolded in the yields evaluated. It is an important but still minor correction even for the lowest yields measured. The energy resolution is limited mainly by the target thickness and energy dispersion of the spectrometer over the entrance slit of the ionization chamber. At an absolute level of 10^{-5} all masses in the mass range A = 79-104 were detected, about 25% of the chains were even detected having yields less than 10^{-6}.

If the excitation energy of the two fragments is smaller than the neutron binding energy (~ 7 MeV), primary yields are measured. As mass and atomic number are conserved the determination of the mass and the atomic number of one fragment allows the identificaiton of the pair of fragments ($A_H = 234-A_L$; $Z_H = 92-Z_L$). In addition from momentum conservation in the binary breakup a calculation of the total kinetic energy E_{TK} of the fragment pair from the kinetic energy E_L of one fragment can be done $E_{TK} = E_L (1 + A_L/A_H)$. Fig. 3 compares the experimentally determined E_{TK} values at the 10^{-5} chain yield level with a calculated Q-value. $\bar{Q}$ is the mean of the Q values for a given mass split into different nuclear charges as determined by the new mass tables of Møller and Nix[5], weighted with the independent yields as measured at kinetic energy $E_K \leq 112$ MeV and extrapolated into the range of high kinetic energies $E_K > 112$ MeV, $\bar{Q} = \Sigma_i Q_i \cdot Y(Z_i)$. The difference $\bar{Q}-E_{TK} = E^*$, which is the excitation energy of the reaction is given in Fig. 4 as a function of the mass split. In almost all cases even mass splits are more excited
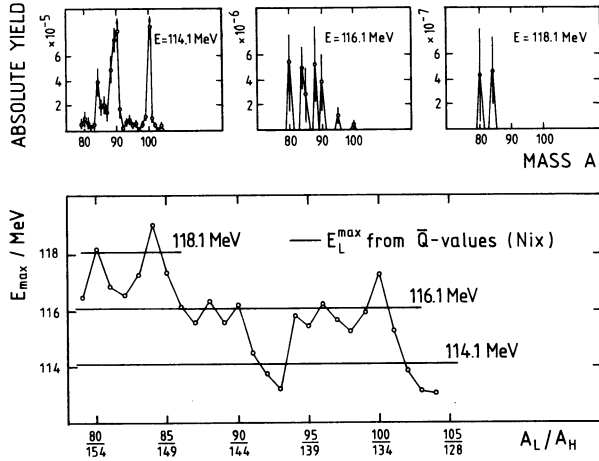
Fig. 1a: Mass yields for cold fragmentation of ^{234}U at 3 energies of the light fission fragments.

Fig. 1b: Maximum kinetic energies for light fragments calculated from $\bar{Q}$-values[5] together with the corresponding cuts chosen for the mass yields of Fig. 1a.

Fig. 3: Total kinetic energy E_{TK} at $Y(A) = \int_{Q}^{TK} dY/dE \cdot dE = 10^{-5}$ in comparison to the $\bar{Q}$-values[5] as a function of the mass ratio. The isotopes of even elements with independent yields larger than 90% and the odd-odd nuclei with yields larger than 50% are indicated.
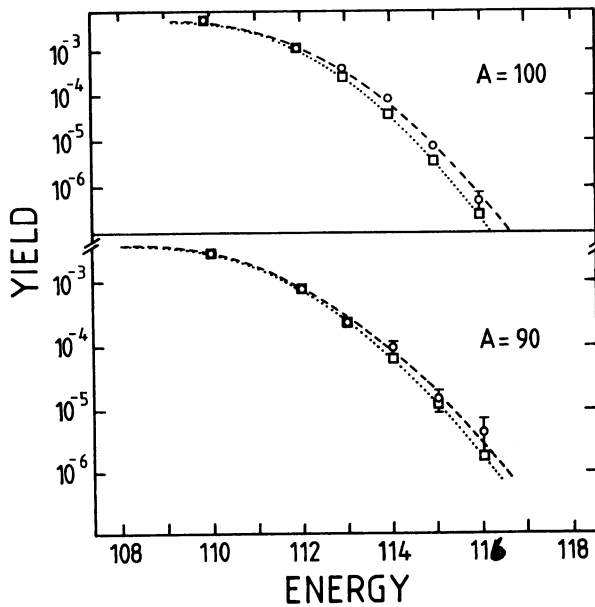

Fig. 2: The mass yields at A = 90 and A = 100 as a function of the kinetic energy of the light fragment (see the logarithmic scale).

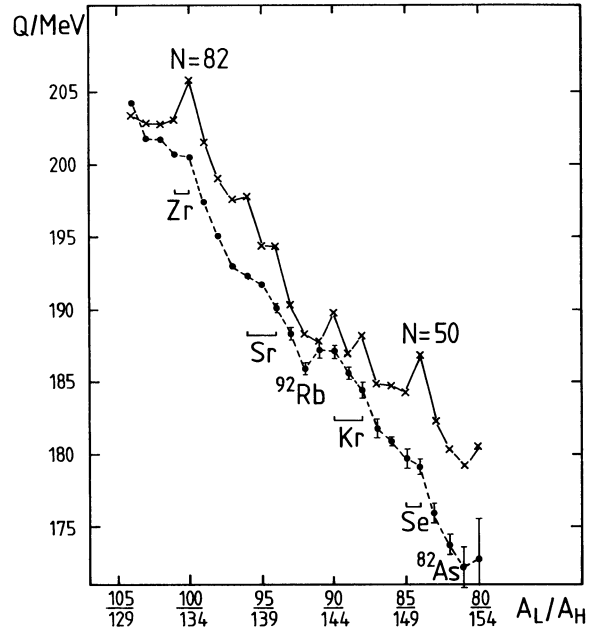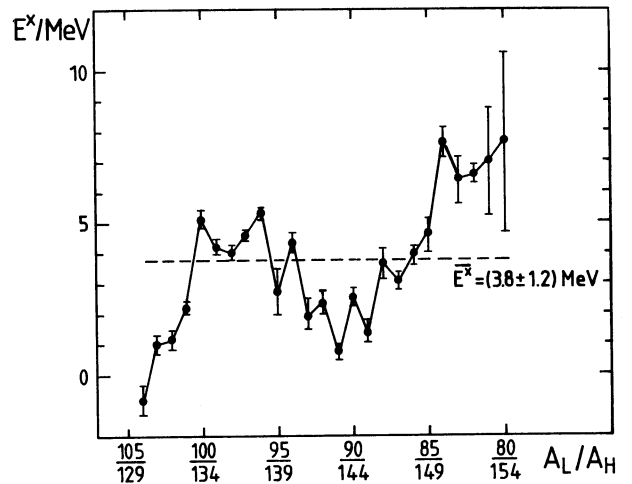----- measured

..... resolution corrected

Fig. 4: Excitation energy $E^* = \bar{Q} - E_{TK}$ at $Y(A) = \int_{Q}^{TK} dY/dE \cdot dE = 10^{-5}$ as a function of the mass ratio.

than odd ones. Low values for E* are observed within the range of A $\sim$ 92 where the highest mass yields in the light fission product group are known to exist. Unless there is a failure of the mass formula not taking into account properly the onset of deformation in the mass range A > 100 or the shell effect at N = 82 in the complementary fragment, the coldest fragmentations are found for mass splits with a broken N = 82 shell.

The maximum possible kinetic energies of light fission fragments can also be deduced from the calculated Q values, using momentum conservation (Fig. 1b). They are given to show the cuts connected to the light fragment energies chosen in the mass yield measurements presented in Fig. 1a.

The independent yield measurement at 112 MeV in the region of cold fragmentation allows a first direct determination of the odd-even effect $(Y_g-Y_u)/(Y_g+Y_u)$ in the neutron yields. A value of 11% is found for neutrons as an average over the mass range, which compares with 46% as measured for protons. The extrapolation of independent yields from measurements at high fission yields $(E_L < 108$ MeV$)$[6] into the range of cold fragmentation gives values of 10% and 40%, respectively. In the following we take mean values from the two determinations. We interpret (following Ref. 9) that about 11% of the fragmentations are unaffected by neutron pair breaking, whereas 43% of the fragmentation are unaffected by proton pair breaking. Even at low excitation energies fragments with broken pairs prevail.

Discussion of Results

The fact of reduced pairing is seen directly from Fig. 4. The excitation energy of even-mass chains is larger than of odd-mass chains. For the difference an average value of (1.0 ± 0.2) MeV is found for the mass range considered. Its origin is the contribution of the pairing energy to the Q-values, which cannot be realized in the case of broken pairs. Taking an odd-even term in the Q-values, e.g. as in the tabulation of Myers[8], we get for the mass range of fission products Δ = 2.1 MeV for even-even and odd-odd pairs of fragments, respectively. The pair breaking leads to an increase in the observed excitation energy in even chains, which is given by $\Delta E^* = 2\Delta\, Y_{uu}$. With the experimental value $Y_{uu} = 0.24 \pm 0.02$, that is 24% of the yield in even chains is found in odd-odd nuclei, we get $\Delta E^* = (1.0 \pm 0.1)$ MeV as non-realized pairing energy in good agreement with the difference in the excitation energies of odd and even mass chains.

The persisting pairing can be seen directly from Fig. 3. It manifests itself in sequences of nuclei, as $^{84-86}$Se, $^{88-91}$Kr, and $^{94-96}$Sr, which carry nearly all the yields of the chains. No comparable sequence is found for even neutron numbers. The small probability of neutron pairing is demonstrated by the high yield of ^{95}Sr and ^{91}Kr. An odd mass isotope successfully competes with its even neighbours, which are unable to take advantage of the neutron

pairing energy.

The charge distribution of fission products is determined by the minimum potential energy of the scission configuration[9,10]. The optimum charge values for most of the chains would be noninteger values. The energy necessary to change from the optimum charge values Z_{opt} on the minimum of the potential energy surface to the actual integer charge value is ΔE^*/MeV = 1.6 $(Z-Z_{opt})^2$[1,10]. In favourable cases, as in the chain A = 82, the energy available for even-even break-ups is reduced by 1.6 MeV, thus leading to a higher yield of odd-odd ^{82}As than its even-even neighbouring isobars. On the average in the mass range considered, the excitation energy bound by non-optimum charge values is 0.4 MeV. Pair breaking and grained charge values averaged over all chains contribute to the excitation on the average $(0.4 + 1.0/2)$ MeV = 0.9 MeV.

The gross features of the strong yield variations demonstrated in Fig. 1 may be understood from the Q-value systematics. The peaks in the 114.1 MeV yield distribution are due to nuclear structure effects. Shell effects are clearly seen. The peak at A = 100 belongs to a fragmentation into the nearly doubly magic ^{134}Te and the soft nucleus ^{100}Zr, which may profit from the N = 60 deformed shell[9]. The peak at A = 90 may be understood from the deformed shell at N $\sim$ 88 [9], and the set-in of deformation at N = 88 for ^{144}Ba [11]. The peak at A = 84 is caused by the N = 50 shell in ^{84}Se.

The peak structures demonstrate that the system is able to take advantage of a few MeV in the reaction Q-value. At low intrinsic excitation energies finally the gain of Q-values by shell effects determines the production yields. Systems stabilized by shell effects are found as reaction products at this level of intrinsic energy predominantly. The predominance is lost for E* > 10 MeV.

However, as Fig. 4 demonstrates, the phenomenon of cold fragmentation is a general one found for all masses in the range A = 80 - 104. Having subtracted from the average experimental excitation energy (3.8 ± 1.2) MeV the energy bound by pair breaking and the integer charge values not meeting the minimum potential energy condition, an excitation energy of (2.9 ± 1) MeV is found in the two fragments at the 10^{-5} chain yield level. This value includes the excitation energy $(B_n - B_f) = 1.1$ MeV of the system at the saddle point. The excitation energy generated by the necking-in process may be estimated to range in the mass range considered between $(0 - 5)$ MeV with an average value of about (1.8 ± 1) MeV.

Converting the measured E_{TK}-values into effective separation distances, we obtain values between $(15 - 16)$ fm. These distances are 4 fm larger than the distance of two touching spheres calculated with $r_0 = 1.17$ fm. From the elongation of the configuration at the second saddle point[12] the corresponding separation distance between the charge centers may be estimated to be 14 fm. The

breaking of the nucleus in cold fission is a necking-in process with a very small increase in the separation distance.

How a necking-in may be accomplished with low energy dissipation is a completely open question. Possibly collective modes such as the giant quadrupole resonance of the configuration at the second saddle point could be involved in initiating the nearly cold transition to a two fragment system. A fast collective necking-in mode with $\Gamma \leq 2$ MeV corresponding to times larger than 3×10^{-22} s would meet the energy rates and allow for pair breaking as deduced from the experiment.

Conclusions

(1) About 10^{-3} of all fission events have excitation energies smaller than 7 MeV. At a yield level of 5×10^{-6} fragmentations with excitation energies less than 1 MeV are found.

(2) The structure in the mass yields for cold fragmentation is determined from the Q-value systematics and the potential energy surface of the scission configuration. For excitation energies smaller than 10 MeV closed shell nuclei dominate the yield.

(3) In the range of cold fragmentation, variations as small as 1 MeV in the intrinsic excitation energy drastically change the mass yields.

(4) The lowest excitation energies are observed for the fission products within the range A $\sim$ 92, which are the most abundant masses of the light fission products. At the lowest excitation energy, nuclei with odd atomic numbers prevail.

(5) The flow of nuclear matter past the second saddle point is predominantly transverse. The necking-in process proceeds with a creation of only a few MeV of intrinsic energy. In nearly all fragmentations broken neutron pairs are found, whereas in about 50% of all cases proton pairs remain unbroken. The matter flow may be predominantly carried by neutrons, and it destroys at least partially the superfluidity of the system.

Acknowledgement

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DISCUSSION

J.B. Wilhelmy: You have shown a picture of touching fission fragments for the cold fragmentation. Is this consistent with the Coulomb energy required with this configuration? Do we need some distortion energy in the fragments to obtain an energy balance?

P. Armbruster: The fragmentation ^{100}Zr/^{134}Te needs, if you convert the Q-value into a separation distance of the charge centres, a deformation of the fragments. Assuming ^{134}Te spherical ^{100}Zr should be deformed with an axis ratio of 2:1. The deformation energy needed for this deformation is assumed to be small, as the nuclear ^{100}Zr is assumed to be a soft one.

D.C. Hoffman: Can you really say for certain that the highest total kinetic energy is for Z = 52, N = 82 (^{134}Te) and Z = 40, N = 60 (^{100}Zr) rather than for $^{132}_{50}$Sn (doubly magic) and $^{102}_{42}$Mo?

P. Armbruster: We have determined the isotopic composition of this A = 100 as a function of the kinetic energy of the light fragment. At high values of E_{TK} the yield of ^{100}Zr becomes 100%, that is the partner must be ^{134}Te.

K. Bleuler: Is it still right to maintain that the two fission products have the same temperature?

P. Armbruster: This assumption is still made, but can be proved experimentally only indirectly. The measured excitation energies are the sum of deformation energy + collective and intrinsic nodes of excitation. How to calculate the temperature from the excitation energy measured is not known. Moreover, at the small energies in cold fission the temperature may not be defined at all. The energy is distributed across the fragments according to the levels available in the fragments.

J.R. Nix: Your larger even-odd effect for protons relative to that for neutrons presumably arises because the small neutron skin and Coulomb polarization freezes in the proton distribution slightly sooner than the neutron distribution. At this point the temperature-dependent pairing gap $\Delta(T)$, upon which the even-odd effect depends exponentially, is larger for protons than for neutrons, as illustrated in this figure:
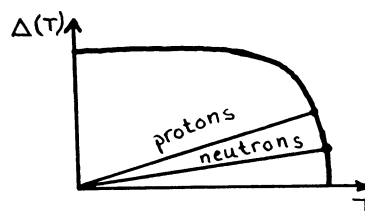
Furthermore, the presence of an even-odd effect means that the excitation energy at scission is very low, which rules out high-dissipation theories for describing thermal-neutron-induced fission.

J. Theobald: Our question is, if fission fragments can be in their ground states in the scission configuration. If the peak region between the nascent fragments is neutron rich, one has to describe neutrons by complicated wave functions. The neutrons remain hot with many pairs broken, while the protons get frozen in the nascent fragments conserving partially the proton pairs. This picture depends of course on the adiabaticity of the fission process.

F. Tondeur: I would like to suggest a schematic explanation of the larger amount of neutron pair breaking in cold fission. Consider the pairing strength G usually taken $\sim A^{-1}$. Protons, already separated in the two fragments, as well as most of the neutrons, should feel the larger pairing strength corresponding to the lower A, whereas the part of the neutrons still belonging to the whole fissioning nucleus should feel a smaller G, so their pairs would be more easy to break. The same conclusion is obtained by considering $\Delta \sim A^{-1/2}$: nucleons in the neck -- probably mainly neutrons -- are expected to be less paired than those who already belong to one of the fragments.