Energy dependence of nuclear charge distribution in neutron induced fission of *Z*-even nuclei

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For the first time the distribution of nuclear charge of fission products with mass numbers 87, 88, 89, 91, 93, 94, 95, 137, 138, 139, and 140 and their complementary products have been studied for neutron induced fission of ^{235}U and ^{239}Pu in the energy range from thermal up to 1.2 MeV. The energy dependences of the cumulative yields of ^{87}Br, ^{88}Br, ^{89}Br, ^{91}Br, ^{93}Kr, ^{94}Rb, ^{95}Rb, ^{137}I, ^{138}I, ^{139}I, and ^{140}I have been obtained by delayed neutron measurements. The most probable charge $Z_P(A)$in the appropriate isobaric β-decay chains was estimated. The results were analyzed in terms of the deviation $\Delta Z_P(A')$ of the most probable charge of isobaric β-decay chains from the unchanged charge distribution before prompt neutron emission (nuclear charge polarization) and they are compared with experimental data of other authors and with predictions from Nethaway's Z_P-formula and Wahl's Z_P-model. We show that the nuclear charge polarization of primary fission fragments $\langle\Delta Z_P(A')\rangle$ before prompt neutron evaporation decreases as the excitation energy of the compound nucleus increases. This decrease is more pronounced for fission of ^{235}U. The energy dependencies of $\Delta Z_P(A')$ and $\Delta Z_P(Z_P)$ obtained in the present work show an attenuation of the odd-even effects in the charge distribution as the excitation energy of the compound nucleus increases.

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I. INTRODUCTION

Distribution of nuclear charge in the process of formation of fission fragments is one of the least investigated aspects of nuclear fission. Most data on nuclear charge distribution in fission reactions at higher energy has been obtained by radiochemical methods [1]. Low energy data concern thermal neutron induced fission of ^{233}U, ^{235}U, ^{239}Pu, and spontaneous fission of ^{252}Cf [2]. Recently, due to developments of new experimental technique data on the charge distribution in thermal neutron induced fission of ^{229}Th, ^{238}Np, ^{232}U, ^{249}Cf have been obtained [3–5]. Isobaric charge distributions of fission fragments are given by its first [the most probable charges $Z_P(A)$] and second (the variances σ^2) moments. Odd-even effects in charge yields are given as a function of the total kinetic energy of fission fragments [6–9]. Few data are known on the behavior of fission fragment charge distribution as function of excitation energy of the compound nucleus, and the existing data are too scarce for deriving a systematics behavior. Recent progress in the development of an appropriate database is described in Ref. [10]. Reviews of theoretical models can be found in Refs. [11–13]. For thermal neutron induced fission of the main uranium and plutonium isotopes information comes through some empirical procedures (Z_P- and A'_P-empirical models by Wahl [2]). Being of one of the fundamental aspects of nuclear fission, the knowledge of the nuclear charge distribution in fission reactions is also closely related to the development of the fission product yields data base which is of importance in reactor design and operation, burnup determination, decay heat calculations and other related applications.

The objective of the present work is to investigate the influence of the excitation energy of the fissioning nucleus on the first moment of the isobaric charge distribution of fission fragments originated in neutron induced fission of *Z*-even nuclides ^{235}U and ^{239}Pu. The cumulative yields of delayed neutron precursors ^{87}Br, ^{88}Br, ^{89}Br, ^{91}Br, ^{93}Kr, ^{94}Rb, ^{95}Rb, ^{137}I, ^{138}I, ^{139}I, and ^{140}I are used to determine the most probable charge in their respective isobaric β-decay chains and the complementary fragments for neutron induced fission in the energy range from thermal up to 1.2 MeV.

II. METHOD FOR DETERMINATION OF THE MOST PROBABLE CHARGE OF FISSION FRAGMENTS

The main features of the experimental method employed in the present work are described in Ref. [14]. The determination of the most probable charge of fission fragments departs from assumption that the primary distribution of fission fragments $Y_{FI}(A, Z)$ in a given isobaric chain A can be described by a Gaussian characterized by the most probable charge Z_P, and the dispersion σ^2 [1]

$$Y_{FI} = \frac{1}{\sqrt{2\cdot\pi\cdot\sigma^2}}\cdot\exp\left[-\frac{1}{2\cdot\sigma^2}\cdot(Z-Z_P)^2\right]. \quad (1)$$

The cumulative yield of an individual fission product in a given isobaric β-decay chain A is uniquely determined by the primary charge distribution. Knowing the fractional cumulative yield of individual member of the isobaric chain $Y_{FC}(A, Z)$ and the charge distribution width σ, one can calculate the most probable charge of fission fragments in this isobaric chain. It was shown by Wahl [2] that the charge dispersion σ^2 for asymmetric fission events in low energy fission of actinides (^{233}U, ^{235}U, ^{239}Pu) is approximately independent of the isobaric mass chain and has a negligible dependence on the incident neutron energy [15]. A cumulative form of the Gaussian distribution gives the fractional cumulative yield of the fission product A with charge Z [1]

$$Y_{FC} = \sum_{0}^{Z} (Y_{FI})_n = \frac{1}{\sigma \cdot \sqrt{2 \cdot \pi}} \cdot \int_{-\infty}^{(Z+1/2)} \exp\left[\frac{-(n - Z_P)^2}{2 \cdot \sigma^2}\right] dn = \frac{Y_m}{2} \cdot \left\{1 + f\left[\left(Z - Z_P + \frac{1}{2}\right) \Big/ \sigma\right]\right\}, \quad (2)$$

where $f[X] = \frac{1}{\sqrt{2 \cdot \pi}} \cdot \int_{-X}^{X} \exp[\frac{-\alpha^2}{2}] d\alpha$. Moreover the fractional cumulative yield $Y_{FC}(A, Z)$ of the fission product (A, Z) after prompt neutron emission can be obtained by the expression [16]

$$Y_{FC}(A, Z) = \frac{Y_d(A, Z)}{P_n(A, Z) Y(A)} \cdot \nu_d, \quad (3)$$

where $Y(A)$ is the chain yield, $Y_d(A, Z)$ and $P_n(A, Z)$ are the relative abundances of delayed neutrons, and the delayed neutron emission probability corresponding to precursor (A, Z), respectively; ν_d is the total delayed neutron yield per one fission. A comprehensive data set on $Y(A, Z)$ and $P_n(A, Z)$ has been gathered in Refs. [10,17]. The measurements of the energy dependence of the relative abundances of delayed neutron precursors $Y_d(A, Z)$ and the total yield of delayed neutrons $\nu_d(E_n)$ from neutron induced fission together with the data base on $Y(A)$ and $P_n(A, Z)$ allow to obtain information on the most probable charge of fission fragments and its behavior as a function of primary neutron energy by solving Eq. (2) relatively to Z_P.

III. MEASUREMENTS AND PROCESSING OF EXPERIMENTAL DATA

The experimental method employed for the determination of $Y_d(A, Z)$ is based on a cyclic irradiation of a fissionable sample by neutrons generated in the $^3H(p, n)^3He$ reaction at CG-2.5 accelerator of IPPE, followed by transfer of the sample to a 4π-neutron detector and measurements of the composite decay of the gross neutron activity [18].

The ion current on tritium target was 50 μA that allowed to produce a neutron flux of about 10^8 neutrons·cm^{-2}·s^{-1} at the sample. The energy spread of the primary neutron flux was determined by a Monte Carlo method on the basis of the double-differential cross section of the $^3H(p, n)^3He$ reaction and accounting for both the slowing down process of protons in the tritium target and neutron multiple-scattering effects in the materials of the setup. In Fig. 1 are shown the energy distributions of primary neutrons $\varphi(E_n)$ which correspond to proton energies of 1.318, 1.550, 1.777, and 1.974 MeV averaged over the volume of the ^{235}U sample.

The ^{235}U and ^{239}Pu sample were enclosed in stainless steel capsules with 0.3 mm thick walls. The capsules were protected against shocks by enclosing them in thin titanium cans. Each can contains four capsules of uranium or plutonium with total weights of up to 2.5 g and 3.6 g, respectively. The isotopic content of uranium sample was 99% of ^{235}U and 1% of ^{238}U; and for the plutonium sample: 95.96% of ^{239}Pu, 3.9% of ^{240}Pu, and 0.14% of ^{241}Pu.

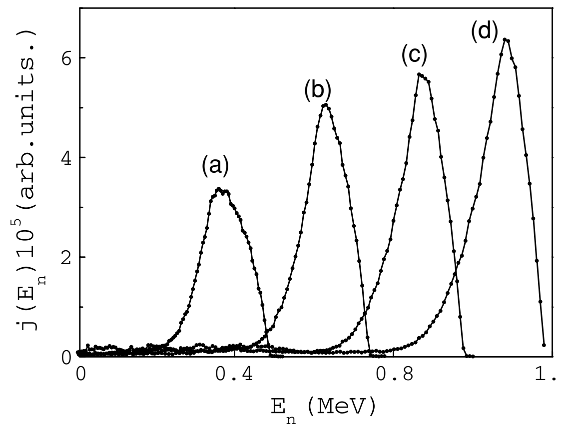

FIG. 1. The energy distributions of primary neutrons $\varphi(E_n)$ averaged over the volume of the ^{235}U sample. (a),(b), (c), (d) denote the energy spectra of $^3H(p, n)^3He$-neutrons generated by protons with energy of 1.318, 1.550, 1.777, and 1.974 MeV, respectively.

Measurements with different irradiation time intervals were done to enhance the contribution of certain delayed neutron groups in the composite delayed neutron decay curve. In the present experiment the irradiation time was 180.06 and 300.06 s. The delayed neutron counting intervals were 424.5 and 724.5 s with the different time sequences after the end of sample irradiation: 0.01 s, 0.02 s, 0.1 s, 1 s, and 10 s. The sample delivery time was 150 ms which is short enough to get information on the relative abundance of delayed neutrons related to the shortest precursors groups.

The energy dependences of the total delayed neutron yield $\nu_d(E_n)$ were obtained in separate experiments based on the method described in Refs. [19,20] and using the relation $\ln[\nu_d(E_n)] = a + b \cdot \ln[\langle T(E_n)\rangle]$ [21,22], where a and b are constants being equal for all fissioning isotopes of one element (see Table I), $\langle T(E_n)\rangle$ is the average half-life of delayed neutron precursors for the nuclide under investigation.

The average half-life values $\langle T(E_n)\rangle$ are calculated from relative abundances Y_i and decay constants λ_i from least squares fitting of delayed neutron decay curves according to $\langle T\rangle = \sum_i Y_i \cdot T_i$, where $T_i = \ln 2/\lambda_i$ [23]. Until recently the total delayed neutron yields ν_d was correlated with parameter $(A_c - 3 \cdot Z_c)$ in the form of exponential dependence [24],

TABLE I. Values of constants a and b for the equation $\ln[\nu_d(E_n)] = a + b \cdot \ln[\langle T(E_n)\rangle]$ determining the relation between the total delayed neutron yield and the average half-life of delayed neutron precursors for fast neutron induced fission of isotopes of thorium, uranium, plutonium, and americium.

Element	a	b
Th	4.06 ± 0.24	-1.26 ± 0.11
U	5.16 ± 0.15	-2.17 ± 0.07
Pu	7.25 ± 0.80	-3.29 ± 0.35
Am	6.30 ± 1.08	-2.93 ± 0.46

where Z_c and A_c are the atomic and mass number of fissioning nucleus, respectively. Later this relation was altered to the form of $(A_c - 3 \cdot Z_c) \cdot (A_c/Z_c)$ [25]. An attempt to justify the dependence $(A_c - 3 \cdot Z_c)$ was undertaken by Pai [26]. Recently, it was found that the average half-life of delayed neutron precursors $\langle T \rangle$ also follow the exponential law in relation to $(A_c - 3 \cdot Z_c) \cdot (A_c/Z_c)$ for isotopes of one elements [22]. As consequence one can suppose that the total delayed neutron yields ν_d and the average half-life values $\langle T \rangle$ for isotopes of one element are related to each other by a power law. Moreover, it is shown that this law is valid at least up to 3 MeV [21]. The exponential dependence of the ν_d value on $(A_c - 3 \cdot Z_c) \cdot (A_c/Z_c)$, used for deriving the recommended values of unmeasured total delayed neutron yields [27], has essentially an isotopic character [21], meaning that isotopes of each fissionable element have their own dependence of ν_d on $(A_c - 3 \cdot Z_c) \cdot (A_c/Z_c)$.

The general equation for determination of the delayed neutron characteristics (Y_{di}, λ_i) on the basis of the measured decay curves $N(t_k)$, summed up over all cycles of irradiation, can be represented by the expression

$$N(t_k) = A \cdot \sum_{i=1}^{m} T_i \cdot \frac{Y_{di}}{\lambda_i} \cdot (1 - e^{-\lambda_i \cdot \Delta t_k}) \cdot e^{-\lambda_i \cdot t_k} + B \cdot \Delta t_k,$$
$$T_i = (1 - e^{-\lambda_i \cdot t_{\text{irr}}}) \cdot \left(\frac{n}{1 - e^{-\lambda_i \cdot T}} - e^{-\lambda_i \cdot T} \cdot \left(\frac{1 - e^{-n \cdot \lambda_i \cdot T}}{((1 - e^{-\lambda_i \cdot T})^2} \right) \right), \quad (4)$$

where $N(t_k)$ is the number of counts registered by 4π-neutron detector in the time-channel t_k with time-channel width Δt_k; A is the saturation activity of a sample; $B$is the intensity of neutron background; λ_i and Y_{di} is the decay constant and relative abundance of delayed neutrons related to i-th group of delayed neutron precursors, respectively; n is the number of cycles; m is the number of delayed neutron groups; T is the duration of one cycle of measurements, which includes the irradiation and the delayed neutron counting time; $t_{\text{irr}}$is the irradiation time.

In processing the experimental data $N(t_k)$ two 12-group models of the time distribution of the delayed neutron precursors based on the known half-lives of 17 precursors were used [14]. The first model was employed to obtain information on the relative abundances Y_{di} of delayed neutrons related to precursors ^{87}Br, ^{88}Br, ^{89}Br, ^{91}Br, ^{93}Kr, ^{94}Rb, ^{95}Rb and the second one for obtaining the relative abundances of delayed neutrons related to precursors ^{137}I, ^{138}I, ^{139}I, and ^{140}I. The group periods were chosen in a way to properly allocate the appropriate delayed neutron precursors, placing each of them in a separate group. The remained groups are composite, comprising of several delayed neutron precursors with the effective periods obtained by an averaging procedure. The analysis of the delayed neutron decay curves with the purpose of obtaining the relative abundances of delayed neutrons related to the delayed neutron precursors has been carried out by the iterative least squares procedure described in Ref. [18]. The data on the emission probability $P_n(A, Z)$ for the investigated delayed neutron precursors were taken from the evaluation by Rudstam *et al.* [17]. The results on the energy dependence of the cumulative yields from neutron induced fission of ^{235}U and ^{239}Pu obtained with Eq. (3) are presented in Table II, along with the appropriate evaluated data from ENDF/B-VI [28]. The uncertainties of the cumulative yields were estimated with

$$\Delta Y_C = \sqrt{(\Delta Y_d)^2 + (\Delta \nu_d)^2 + (\Delta P_n)^2}.$$

IV. RESULTS

A. Energy dependence of the most probable charge

The values of the most probable charge $Z_P(A)$ in the isobaric chains with mass numbers $A = 87, 88, 89, 91, 93, 94, 95, 137, 138, 139, 140$ was estimated with the help of Eq. (2) on the basis of the cumulative yields of delayed neutron precursors ^{87}Br, ^{88}Br, ^{89}Br, ^{91}Br, ^{93}Kr, ^{94}Rb, ^{95}Rb,^{137}I, ^{138}I, ^{139}I, and ^{140}I presented in Table II. As an example in Table III are shown the numerical data on the most probable charges and their uncertainties obtained from 0.5 MeV-neutron induced fission of ^{235}U. As it can be seen from this table the uncertainties of the Z_P values obtained with the above procedure have the same order of magnitude as the uncertainties of corresponding radiochemical data [2].

The most probable charge of isobaric chains with the mass numbers of complementary fission fragments was obtained using the charge conservation law $Z_L + Z_H = Z_C$, where Z_C, Z_L and Z_H are the atomic numbers of the compound nucleus, light and heavy fission fragments, respectively, and the expression $A_L + A_H + \nu_p(E_n) = A_C$ connecting the mass numbers of the light (A_L) and heavy (A_H) fission products after emission of ν_p prompt neutrons with the mass number of the compound nucleus A_C. The results on the $Z_P(A)$ dependence for thermal-, 0.5- and 1.2 MeV-neutron induced fission of ^{235}U and ^{239}Pu are shown in Fig. 2. For clarity, the 0.75 and 1 MeV data are omitted. Along with the present data we show experimental data from Wahl's compilation [2], data calculated in frame of the Z_P-model [2], and the data obtained from Waldo's empirical Z_P-equations [29]

$$Z_P(A) = 0.04153 \cdot A - 1.19 + 0.167 \cdot (236 - 92 \cdot A_c/Z_c), \quad A < 116 \quad (5)$$

$$Z_P(A) = 0.04153 \cdot A - 3.43 + 0.243 \cdot (236 - 92 \cdot A_c/Z_c), \quad A > 116. \quad (6)$$

It is seen from this figure that the present data are in a good overall agreement with the experimental data and the data calculated with the Z_P-model by Wahl [2]. Waldo's Z_P-equations do not reproduce well the experimental $Z_P(A)$ data in the region of heavy fission products, especially in the case of ^{235}U.

The most prominent feature of the present $Z_P(A)$ data is that the most probable charge of light fission products has a tendency to decrease when the excitation energy of the compound nucleus increases. The values of the slope $\langle dZ_P(A)/dE_n \rangle$ averaged over light fission products are – 0.030 ± 0.008 and – 0.029 ± 0.010 ch.u./MeV for fission of ^{235}U and ^{239}Pu, respectively. This effect is clearly seen in Fig. 3 where the present data are compared with two empirical

TABLE II. The cumulative yields of delayed neutron precursors from neutron induced fission of ^{235}U and ^{239}Pu in the energy range from thermal to 1.2 MeV.

Nuclide	Present work			ENDF/B-VI	
	Thermal	0.5 MeV	1.2 MeV	Thermal	0.5 MeV
^{235}U					
^{87}Br	2.20 ± 0.08	2.19 ± 0.12	2.26 ± 0.14	2.04 ± 0.04	2.11 ± 0.13
^{88}Br	2.42 ± 0.09	2.43 ± 0.13	2.35 ± 0.15	1.78 ± 0.05	2.12 ± 0.13
^{89}Br	1.54 ± 0.07	1.63 ± 0.10	1.76 ± 0.12	1.09 ± 0.03	1.44 ± 0.33
^{91}Br	0.22 ± 0.03	0.20 ± 0.03	0.21 ± 0.03	0.22 ± 0.03	0.14 ± 0.09
^{93}Kr	0.29 ± 0.02	0.29 ± 0.03	0.30 ± 0.03	0.49 ± 0.02	0.35 ± 0.16
^{94}Rb	2.01 ± 0.09	2.16 ± 0.13	2.28 ± 0.15	1.65 ± 0.05	1.95 ± 0.12
^{95}Rb	1.29 ± 0.07	1.16 ± 0.08	1.15 ± 0.08	0.77 ± 0.03	0.92 ± 0.41
^{137}I	3.39 ± 0.13	3.44 ± 0.19	3.59 ± 0.22	3.07 ± 0.09	2.57 ± 0.15
^{138}I	1.71 ± 0.09	1.68 ± 0.11	1.74 ± 0.13	1.49 ± 0.04	1.35 ± 0.05
^{139}I	0.67 ± 0.04	0.66 ± 0.05	0.67 ± 0.05	0.78 ± 0.06	0.47 ± 0.04
^{140}I	0.15 ± 0.02	0.15 ± 0.02	0.15 ± 0.02	0.15 ± 0.04	0.11 ± 0.07
^{239}Pu					
Nuclide	Present work			ENDF/B-VI	
	Thermal	0.5 MeV	1.2 MeV	Thermal	0.5 MeV
^{87}Br	0.75 ± 0.05	0.76 ± 0.05	0.83 ± 0.05	0.69 ± 0.04	0.81 ± 0.06
^{88}Br	0.75 ± 0.05	0.71 ± 0.05	0.74 ± 0.05	0.51 ± 0.03	0.55 ± 0.25
^{89}Br	0.34 ± 0.03	0.35 ± 0.03	0.38 ± 0.03	0.35 ± 0.01	0.33 ± 0.21
^{91}Br	0.04 ± 0.01	0.04 ± 0.01	0.04 ± 0.01	0.02 ± 0.01	0.03 ± 0.02
^{93}Kr	0.15 ± 0.01	0.15 ± 0.01	0.16 ± 0.01	0.07 ± 0.01	0.14 ± 0.09
^{94}Rb	0.78 ± 0.06	0.79 ± 0.06	0.86 ± 0.06	0.73 ± 0.08	0.67 ± 0.30
^{95}Rb	0.36 ± 0.03	0.41 ± 0.03	0.38 ± 0.03	0.43 ± 0.07	0.29 ± 0.19
^{137}I	2.19 ± 0.14	2.17 ± 0.15	2.11 ± 0.14	2.43 ± 0.09	2.00 ± 0.22
^{138}I	0.99 ± 0.07	1.01 ± 0.07	1.02 ± 0.07	1.28 ± 0.08	1.05 ± 0.12
^{139}I	0.19 ± 0.01	0.19 ± 0.02	0.20 ± 0.02	0.32 ± 0.07	0.19 ± 0.12
^{140}I	0.027 ± 0.004	0.027 ± 0.004	0.030 ± 0.004	0.06 ± 0.02	0.03 ± 0.02

Z_P-formulas which, in contrast to Waldo's equations (4),(5), have energy dependent terms. The first formula is by Nethaway *et al.* [30], the second one is from Wahl [2] for the modified Z_P-model used for the estimation of the fractional independent yields of fission fragments.

TABLE III. The most probable charge and its uncertainty in separate isobaric chains from 0.5 MeV-neutron induced fission of ^{235}U.

Present work			Radiochemical data [1]		
A	$Z_P(A)$	$\pm\Delta Z_P$	A	$Z_P(A)$	$\pm\Delta Z_P$
87	34.84	0.17	86	33.91	0.25
88	35.21	0.07	89	35.42	0.12
89	35.64	0.04	93	37.39	0.1
91	36.46	0.04	94	37.94	0.15
93	37.38	0.02	136	53.53	0.13
94	37.71	0.03	138	53.45	0.1
95	38.01	0.02	144	56.40	0.24
137	53.43	0.05			
138	53.85	0.03			
139	54.17	0.02			
140	54.54	0.03			

Nethaway's Z_P-formula, which most frequently has been used in practice, is based on the Z_P data obtained for thermal-neutron induced fission of ^{235}U

$$Z_P(Z_c, A_c, E_x) = Z_P(92; 236; 6.545) + a \cdot (Z_c - 92) + b \cdot (A_c - 236) + c \cdot (E_x - 6.545), \quad (7)$$

where E_x is the excitation energy of the compound nucleus (A_c, Z_c). The values of the coefficients a and b are different for light and heavy fission products. The last term in Eq. (7) was introduced to account for an influence of the excitation energy of the fissioning nucleus on the most probable charge in an isobaric chain. The value of the coefficient c is the same for all light products, $c_L = 0.0174$ (ch.u./MeV). For the region of heavy products the value of the coefficient c depends on the fragment mass number A_H: $c_H = 0.051 - 0.0023(A_H\text{-}130)$ (ch.u./MeV). A similar $Z_P(A)$-formula was developed by Wang Dao *et al.* [10], which is of the same form as Nethaway's one including the same coefficients a and b, but which has an additional term connected with influence of ternary fission on the Z_P values. Its coefficient c is slightly different from

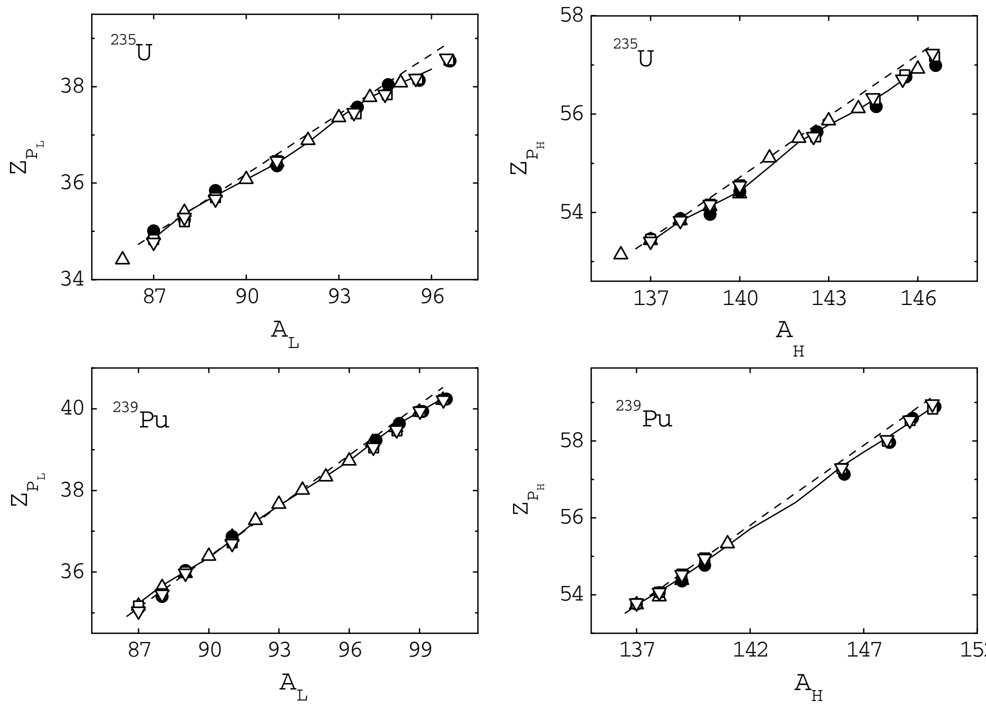

FIG. 2. The most probable charge $Z_P(A)$ as a function of mass number of the fission products after prompt neutron emission from fission of ^{235}U and ^{239}Pu by thermal, 0.5 and 1.2 MeV neutrons. Solid circles, open squares and open down triangles present data related to thermal, 0.5 and 1.2 MeV neutron induced fission, respectively. Open up triangles are the radiochemical data from the compilation by Wahl [2], the dashed curve is Waldo's $Z_P(A)$-formula, the solid curve is the results of calculations made in frame of the Z_P -model by Wahl [2]. Subscripts L, H refer to light and heavy fission products.

Nethaway's formulation. The coefficient c in both equations is positive because the shift of charge distribution in a given isobaric chain is connected only with prompt neutron emission, which shifts fragments closer to the valley of β-stability. The results is shortening the β-decay chain lengths of fission products and consequently an increase of the most probable charge of isobaric chains, see Fig. 3.

The second reason for a shift of the most probable charge of the fission products is connected with a decrease of the charge polarization in primary fission fragments. This polarization effect was observed in thermal-neutron induced fission of heavy nuclides, and amounts to about 0.5 ch.u. [2]. On the average a decrease of charge polarization leads to a decrease of charge density of light fragments, and an increase of charge density of heavy fragments. As the excitation energy of the compound nucleus increases the primary heavy fragments therefore are approaching the line of β-stability and primary light fragments are moving off it. This effect decreases the most probable charge of light fission products and, increases it for the heavy products. Therefore it is likely that the final distribution of nuclear charge of fission products is determined by the two-stage process. The first one is related to the process of forming the fission fragments before prompt neutron evaporation, whereas the second one is connected with prompt neutron emission from excited fragments. We therefore consider the present results on the shift of the most probable charge due to an increase in the excitation energy of the compound nucleus as the cumulative effect of the above mentioned two mechanisms. Taking into account that an increase of the average number of prompt neutrons affects mainly the heavy fission fragments [6,31,32] the decrease of the most probable charge of light fission products observed in the present work can be entirely attributed to a decrease of charge polarization in primary fragments.

This feature was introduced in the empirical Z_P-formula by Wahl [10] which is presented in the form of the nuclear charge deviation from the unchanged charge distribution (UCD)

$$Z(A_L) = A'_L \cdot \left[\frac{Z_C}{A_C}\right] - \Delta Z(A'_{H_C}),\quad A'_{H_C} = A_C - A'_L, \tag{8}$$

$$Z(A_H) = A'_H \cdot \left[\frac{Z_C}{A_C}\right] + \Delta Z(A'_H), \tag{9}$$

$$\Delta Z(A'_H) = \Delta Z(140) + \frac{\partial \Delta Z}{\partial A'} \cdot [A'_H - 140],\quad A'_L = A_L + \nu_{pL},\quad A'_H = A_H + \nu_{pH}, \tag{10}$$

where subscripts L, H, and C denote the light and heavy fission fragments, and the compound nucleus, respectively; A'_{H_c} is the atomic number of the heavy fragment which is complementary to light fragment A'_L . After substituting numerical data in the $\Delta Z(140)$ term one can obtain the final expression for the deviation of the most probable charge Z_P from UCD, $Z_{\rm ucd} = (Z_c/A_c) \cdot (A')$ as a function of the

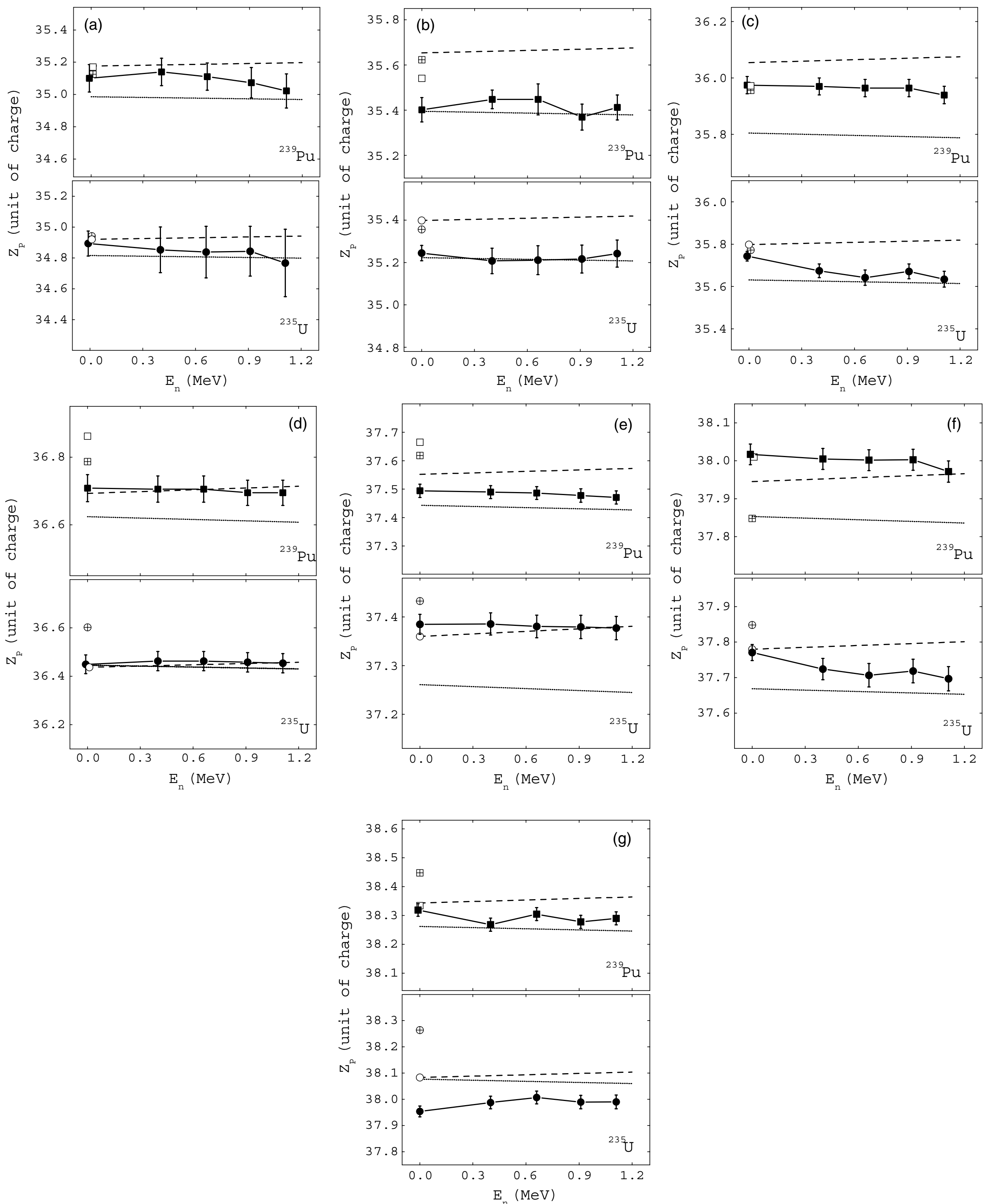

FIG. 3. The energy dependence of the most probable charge in separate isobaric chains after prompt neutron emission from neutron induced fission of ^{235}U and ^{239}Pu. (a), (b), (c), (d), (e), (f), and (g) denote graphs related to the isobaric chains with mass number $A = 87, 88, 89, 91, 93, 94, 95$, respectively. Solid circles and squares show the present data; open circles and squares show the experimental data from compilation [2]; crossed open circles and squares show the data calculated with help of Waldo's Z_P-equation; dashed and dotted lines show the data calculated with the help of Nethaway's and Wahl's Z_P-equations, respectively.

excitation energy E_x of any fissioning nucleus A_c

$$\Delta Z(A'_H) = (-0.495 \pm 0.013) + (0.0034 \pm 0.029) \cdot (A_C - 236) + (0.0137 \pm 0.0054) \cdot (E_x - 6.551) - 0.006 \cdot (A'_H - 140). \quad (11)$$

It is seen from Fig. 3 that the above Z_P-formula reproduce the effect of the linear decrease of Z_P in the mass region of light fission products as the excitation energy of the compound nucleus increases. However, the absolute Z_P values, being based on a linear approximation of experimental data, do not reproduce the odd-even effects in the charge distribution. As a consequence in Fig. 3 can be seen the discrepancy between the present data and the Z_P data calculated from above equation for some isobaric chains, which are of order of the amplitude of the odd-even effect (0.1-0.2 ch.u.). At the same time Fig. 3 shows good agreement between the present thermal-neutron $Z_P$values and the corresponding experimental data [2] for all isobaric chains, except chains with $A = 88$ and 95 for fission of ^{235}U. In the case of ^{239}Pu agreement with the present data is obtained in isobaric chains $A = 87, 89, 94, 95$. In the remaining chains $A = 88, 91, 93$ one can see a discrepancy between the data which amounts up to 0.1-0.2 ch.u. Waldo's formula agrees with the present data only in the case of isobaric chains $A = 87$ and 89 for both ^{235}U and ^{239}Pu fission.

B. Energy dependence of the most probable charge of primary fission fragments and value of the odd-even effect

Until now we considered the most probable nuclear charge of fission products after prompt neutron emission. A different approach in a discussion of the charge distribution in fission is to analyze the behavior of the most probable charge in terms of its deviation from the unchanged charge distribution (UCD) $\Delta Z_P = Z_P\text{-}Z_{\text{ucd}}$, as a function of the mass number of primary fission fragments A' or the most probable charge Z_P [15]. The primary fragment mass number A' was calculated by

$$A'_L = A_L + \nu_{pL}, \quad A'_H = A_H + \nu_{pH}, \quad (12)$$

where ν_{pL} and ν_{pH} is the average number of prompt neutrons emitted by light (L) and heavy fission fragments (H). The average number of prompt neutrons induced by energetic neutrons (0.5, 0.75, 1, 1.2 MeV) was estimated from Terrell [33]

$$\nu_{pL} = 0.531 \cdot \nu_p + 0.062 \cdot (A_L + 143 - A_C), \quad (13)$$
$$\nu_{pH} = 0.531 \cdot \nu_p + 0.062 \cdot (A_H + 143). \quad (14)$$

The $\nu_p(A)$ values, weighted by the chain yield $Y(A)$, were summed and normalized to the experimental average number of prompt neutrons per fission $\nu_p(E_n)$ [34]. Experiment shows that an increase in the excitation energy of the fissioning system leads to an increase of the average number of prompt neutrons $\nu_p(A)$ mainly for heavy fragments [6,31,32].

Results for the $\Delta Z_P(A')$ and $\Delta Z_P(Z_P)$ dependencies for thermal, 0.5 and 1.2 MeV neutron induced fission of ^{235}U, ^{239}Pu are shown in Figs. 4 and 5. The deviations of the average nuclear charge from UCD $\Delta Z_P(A')$are compared with $\Delta\bar{Z}(A')$ derived from the evaluated experimental fractional independent yields [2] and $\Delta\bar{Z}(A')$ obtained from the fractional independent yields by Wahl's Z_P-model [2], which accounts for even-odd-proton and -neutron effects. ΔZ_P is compared with the $\Delta\bar{Z}(\bar{Z})$data obtained in Ref. [15] and Wahl's Z_P-model, both related to fission by thermal neutrons. The values of the most probable charge Z_P and the average nuclear charge $\bar{Z}$ of charge distribution in isobaric chain agree closely with each other.

Figures 4 and 5 show the results for the deviation of the most probable nuclear charge from UCD as function of Z_P and A' for fission of ^{235}U, which are in good agreement with the evaluated experimental data [2] and the results from Z_P- model [2], as well as with data from Ref. [15]. The present results on the $\Delta Z_P(A')$ dependence for ^{235}U are in a better agreement with values from [2] than with the evaluated experimental data, especially in the heavy fragment region. In the case of ^{239}Pu, taking into account the systematic bias of about 2 a.m.u. in the present $\Delta Z_P(A')$ results in relation to the other data, good agreement with both the evaluated data and results from the Z_P- model is seen. The present $\Delta Z_P(Z_P)$ data for ^{239}Pu and the appropriate values obtained by the physical method are also displaced in relation to each other by one ch.u. At present there is no theory on where the maxima in $\Delta Z_P(A')$ should be. Therefore it is difficult to judge about the possible cause of the observed overall shift of the $\Delta Z_P(A')$ dependences. As regards the $\Delta Z_P(Z_P)$ dependences, the maxima most likely should be closer to even numbers of Z_P since these give the maximum in the independent fission fragment yields which are used as a weight function for obtaining the values of the average nuclear charge $\bar{Z}(A')$. This correlation is observed in the present evaluation.

In general our results show the well established features for the $\Delta Z_P(A')$ and $\Delta Z_P(Z_P)$ dependences [15]. For asymmetric fission the values of ΔZ_P are negative for heavy fission fragments and positive for light fission fragments. $|\Delta Z_P|$ values are within limits of 0.5-0.6 ch.u.; a large oscillation with

TABLE IV. The average charge polarization $\langle \Delta Z_P \rangle$ of light fission fragments before prompt neutron evaporation. The letters a, b, c and d in the second column denote results of the present work, the experimental radiochemical data [2], and the results obtained from the Z_P -model by Wahl [10] and Nethaway [30], respectively. The numbers in parentheses are the difference Δ between the charge polarization values derived from 1.2 MeV- and thermal-neutron induced fission.

Target nuclide		$\langle \Delta Z_P \rangle$,ch.u. (Light fission fragments)	
		Thermal	1.2 MeV
^{235}U	a	0.56 ± 0.06	0.53 ± 0.06 ($\Delta = -0.035$)
	b	0.57 ± 0.05	–
	c	0.51	0.49 ($\Delta = -0.017$)
	d	0.57	0.59 ($\Delta = 0.02$)
^{239}Pu	a	0.61 ± 0.06	0.59 ± 0.06 ($\Delta = -0.016$)
	b	0.58 ± 0.05	–
	c	0.52	0.50 ($\Delta = -0.016$)
	d	0.64	0.66 ($\Delta = 0.022$)

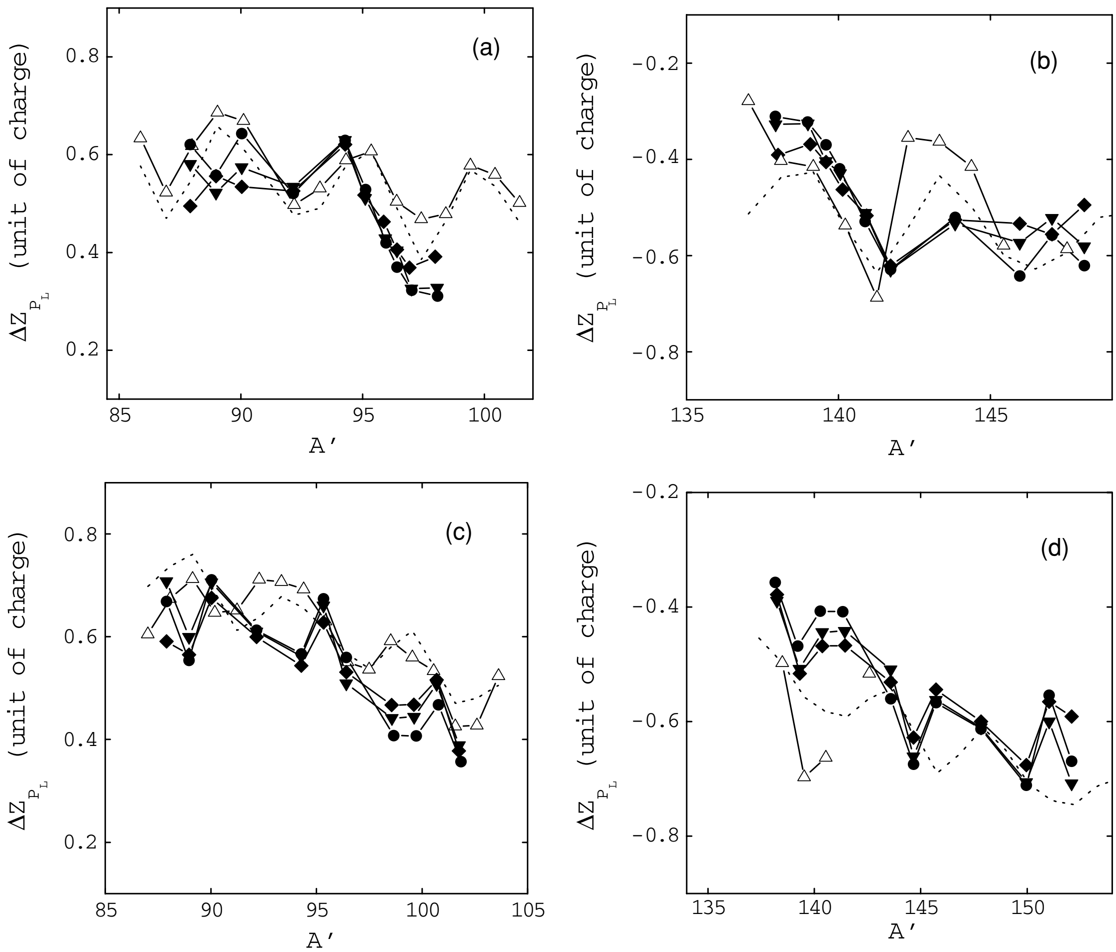

FIG. 4. Deviation of the most probable charge from the unchanged charge distribution $\Delta Z_P(A')$ as a function of the mass number of primary fission fragments A'· (a), (b) - ^{235}U; (c), (d) - ^{239}Pu. Solid circles, solid diamonds and solid up triangles show the present data corresponding to thermal, 0.5 and 1.2 MeV neutron induced fission, respectively. Open up triangles show the experimental data for thermal neutron induced fission taken from the compilation by Wahl [2]. Dashed curve shows results from the Z_P-model by Wahl [2]. Subscripts L and H denote the light and heavy fission fragments. For clarity of display, the points of each data set are connected by different types of line, and 0.75 and 1 MeV data are omitted.

a spacing of two charge units for Z-even compound nucleus as function of the most probable charge Z_P, and spacing of five mass units as function of $A'$are observed.

To make a quantitative estimate of the influence of the excitation energy ΔE_x on the value of the charge polarization the ΔZ_P data were averaged over the mass regions of the light fission fragments, Eq. (5). The range of averaging is large enough for elimination of a possible influence of the odd-even effect. Results are presented in Table IV together with radiochemical data [2] for thermal neutron induced fission. In Table IV are presented also the data on $\langle \Delta Z_P \rangle$ at incident neutron energy of 1.2 MeV calculated from Wahl's and Nethaway's Z_P-formulas.

Uncertainties in the present data are mainly systematic, resulting from uncertainties in chain yields $Y(A)$, emission probabilities of delayed neutrons P_n , and total delayed neutron yields $\nu_d(E_n)$. The present data on the most probable charge for all fissioning systems and incident neutron energies were obtained by the same method, experimental installation and data processing procedure. Therefore the trends in a shift of the most probable charge caused by a change of the excitation energy of the compound nuclei should not be affected by systematic errors, even though a value of this shift is the same order of magnitude as the quoted uncertainties.

It is seen from Table IV that the average deviations of the most probable charge from UCD for thermal neutron induced fission of ^{235}U, ^{239}Pu are in good agreement with the charge polarization calculated from the radiochemical Z_P-data [2]. The average charge polarization corresponding to thermal-neutron induced fission of ^{235}U, ^{239}Pu calculated from the Z_P-formula [10] show relatively large deviation from the radiochemical data and the present work. The $\langle \Delta Z_P \rangle$ values for 1.2 MeV neutron induced fission of ^{235}U derived from this equation are in a good agreement with the present data, whereas in the case of ^{239}Pu they disagree by 0.1 ch.u. As the excitation energy of both fissioning nuclides increases, one can see the decrease of the charge polarization $|\Delta Z_P|$. On the average over the light fragments the decrease is 0.035

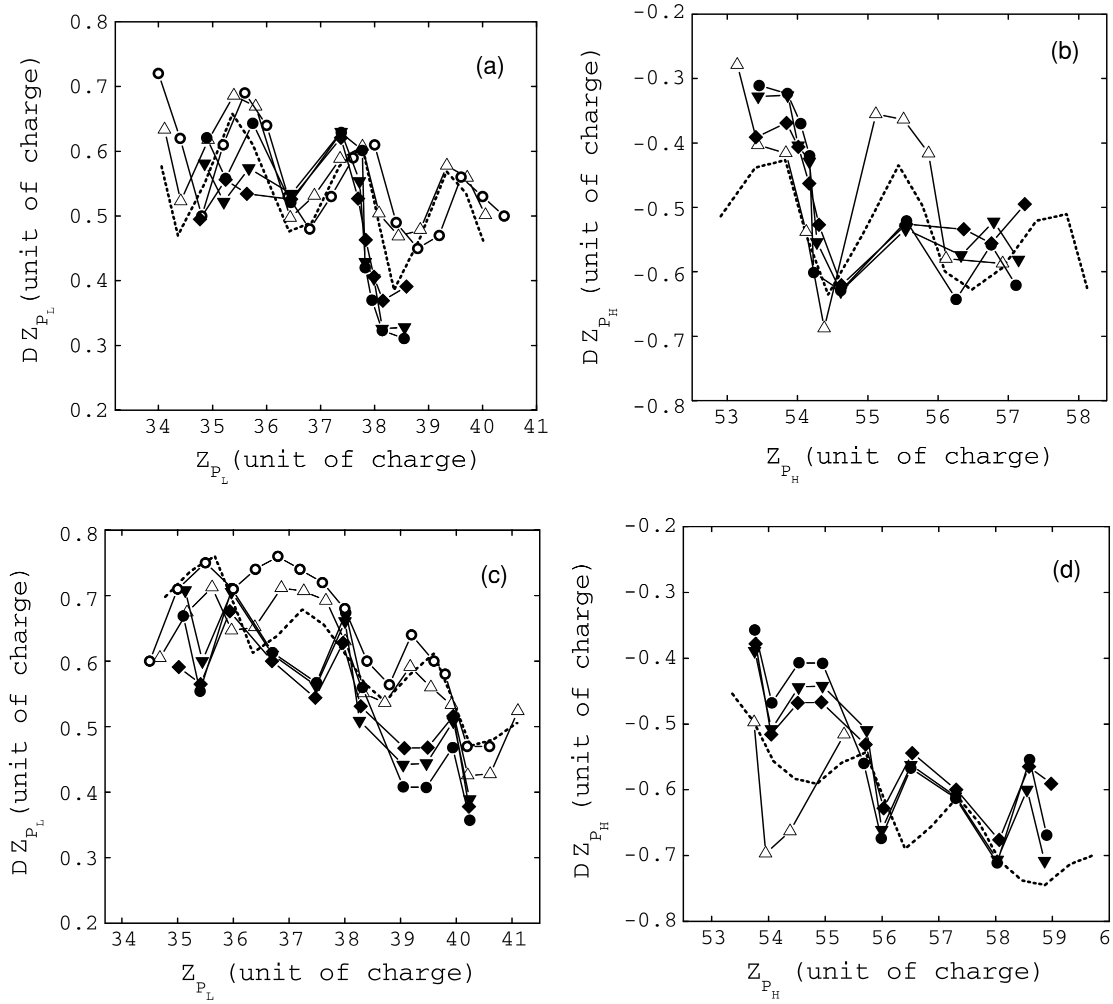

FIG. 5. Deviation of the most probable charge from unchanged charge distribution (UCD) as a function of the most probable charge of isobaric chain $\Delta Z_P(Z_P)$. (a), (b) - ^{235}U; (c), (d) - ^{239}Pu. Solid circles, open squares and open down triangles show the present data related to thermal, 0.5 and 1.2 MeV neutron induced fission, respectively. Open circles and up triangles show the data on the deviation of the average nuclear charge from UCD as a function of the average nuclear charge by Bocquet *et al.* [15], and by the compilation by Wahl [2], respectively, both from thermal neutron induced fission. Dotted curve shows calculations in frame of Z_P-model by Wahl [2]. For clarity of display, the points of each data set are connected by continuous lines and 0.75 and 1 MeV data are omitted.

and 0.016 ch.u./1.2 MeV for ^{235}U and ^{239}Pu, respectively. Corresponding 1.2 MeV data calculated from the energy dependent Wahl's Z_P-formula are 0.017 and 0.016 ch.u./ 1.2 MeV. The Z_P-formula by Wahl [10] gives the same sort of correlation between the excitation energy E_x of the compound nucleus and the value of the nuclear charge polarization of fission fragments before neutron evaporation.

Nethaway's Z_P-formula does not reproduce the observed trend in the shift of the most probable charge due to the change in the excitation energy of the fissioning nucleus ΔE_x , giving a different correlation between the ΔE_x and Z_P values than it is observed in the present work. The energy dependence in Nethaway's Z_P formulas is governed by the coefficient c which has the same sign for the light and heavy fission products. This formula accounts only for the effect of prompt neutron emission. Furthermore, this equation does not take into account the different values of nuclear charge shift observed in primary fission fragments of different fissioning systems before prompt neutron emission.

C. Energy dependence of the odd-even effects.

It was shown that the odd-even effect strongly depends on the excitation energy of the compound nucleus [7,35,36]. The odd-even effects in the charge yield for the ^{235}U(n, f) reaction at 2 MeV neutron energy is reduced to 9–10%, compared to 22% at the thermal energy. At 3 MeV the effect is about 5%. The present method does not allow to estimate quantatively the magnitude of the odd-even effects in the charge distribution of fission fragments. It has been seen that the oscillations of $Z_P(A')$ and, consequently, $\Delta Z_P(A')$ dependence are determined by the dominant charge Z of fission fragments in the isobaric chain A'. The amplitude of these oscillations is proportional to the value of the odd-even effect for the individual nuclide. If the amplitude of $\Delta Z_P(A')$ and/or $\Delta Z_P(Z_P)$ oscillations can be taken as a measure of the magnitude of the odd-even effect (22% and 12% for thermal neutron induced fission of ^{235}U and ^{239}Pu respectively) one can conclude that the odd-even effect for 1.2 MeV neutron induced fission of these nuclides will be approximately 15 and 7%.

V. SUMMARY

The energy dependence of the cumulative yields of ^{87}Br, ^{88}Br, ^{89}Br, ^{91}Br, ^{93}Kr, ^{94}Rb, ^{95}Rb, ^{137}I, ^{138}I, ^{139}I, ^{140}I and the most probable charge $Z_P(A)$ in the appropriate isobaric β-decay chains have been obtained with a new method based on delayed neutron measurements in neutron induced fission of Z-even nuclides ^{235}U and ^{239}Pu. Results on the most probable charge of isobaric chains obtained for thermal neutron induced fission of ^{235}U and ^{239}Pu are in a good agreement with radiochemical data [2], and data obtained by direct physical methods [15,37]. It shows that the method based on the delayed neutron measurements used in the present work can be considered as reliable. The advantage of the method is that it can be easily adjusted to the measurements at a wider energy range of primary neutrons.

As a result of the studies the most probable charge of the investigated isobaric chains has been obtained for fission of ^{235}U and ^{239}Pu by thermal neutrons and neutrons with energies of 0.5, 0.75, 1, and 1.2 MeV. It was found that Z_P of light fission products decreases with the increase of the excitation energy of compound nucleus. Taking into account that the increase of the average number of prompt neutrons in low energy fission is related mainly to heavy fission fragments, this effect can be considered as the direct indication of the decrease of the charge polarization value of primary fragments. The deviation of the most probable charge of fission fragments from the unchanged charge distribution (charge polarization value) has been obtained as a function of fission fragment mass A' and the most probable charge Z_P, as function of the excitation energy of the compound nuclei. The magnitude of the nuclear charge polarization $|\langle \Delta Z_P \rangle|$ of fission fragments before prompt neutron evaporation is approximately the same for thermal neutron induced fission of ^{235}U and ^{239}Pu. As the excitation energy of the fissioning system increases a decrease of the charge polarization $|\langle \Delta Z_P \rangle|$ was observed. The results most likely indicate that the charge density of fission products is determined by two stage process: a redistribution of nuclear charge in the process of forming of fission fragments and emission of prompt neutrons from excited fragments. On the average over light fission fragments this decrease is 0.035 and 0.017 ch.u./ MeV for ^{235}U and ^{239}Pu, respectively.

A new empirical energy dependent Z_P-formula by Wahl [10] reproduces the effect of the linear decrease of Z_P in the mass region of light fission products as the excitation energy of compound nucleus increases but it predicts less pronounced energy dependence of the most probable charge of primary fission fragments in the case of ^{235}U. Besides, the absolute Z_P values from this formula, being based on the linear approximation of experimental data, do not reproduce the odd-even effects in the charge distribution.

If the amplitude of the oscillations in the $\Delta Z_P(A')$ or $\Delta Z_P(Z_P)$can be taken as a measure of the magnitude of the odd-even effects, one can conclude that the energy dependencies of $\Delta Z_P(A')$ and $\Delta Z_P(Z_P)$ obtained in the present work show an attenuation of the odd-even effects in the charge distribution as the excitation of the compound nucleus increases.

The obtained data may be useful for improvements of the data on the most probable charge of fission products needed for the development of the energy dependent fission yields data. Taking into account the different values of the charge polarization obtained in fission of ^{235}U and ^{239}Pu nuclides the energy dependent $Z_P(A)$ empirical models for each nuclide should use its own reference standard, instead of the standard used up to now namely—the $Z_P(A)$ data related to thermal neutron fission of ^{235}U.

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