Analysis of isotopic yield distributions in ^{11}B- and ^{22}Ne-induced fission of ^{238}U

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First moments and variances of measured rubidium and cesium isotopic distributions in the fission of ^{238}U by ^{11}B(E_{lab} = 85 MeV) and ^{22}Ne (E_{lab} = 132 and 162 MeV) are analyzed in the framework of the statistical model for nuclear fission and of evaporation theory. Excellent agreement with the data is obtained for ^{11}B-induced fission, assuming a compound-nuclear mechanism. The comparatively large isotopic variance observed in ^{22}Ne-induced fission can be explained if moderate contributions from fission following direct reactions are taken into account.

[NUCLEAR REACTIONS, FISSION ^{238}U with heavy ions (^{11}B, ^{22}Ne); isotopic distributions, calculated first moments, variances. Statistical model theory; good agreement with data; direct reaction contributions important.]

I. INTRODUCTION

A number of isotopic distributions of fragments from heavy-ion-induced fission have been measured[1,2] in the past and have been analyzed mainly in terms of the statistical theory of fission.[3] An important result[1] was that the variance of mass and charge in these fission reactions could be well understood for systems with $Z^2/A < 37$ (A,Z mass and charge of the fissioning nucleus). Beyond this value, however, a strong increase of the variances, well above statistical model expectations, was observed.

A number of possible reasons for this departure from statistical model predictions was given by the authors of Ref. 1. They showed that considerably larger fragment *mass* dispersions could be expected if one assumes, in contrast to the statistical model, a very rapid descent from saddle to scission point. The increased variance of the *isotopic* distributions was suggested to result from increased neck sizes at scission point for higher values of Z^2/A. Part of the isotopic variance was assumed to result from random distribution fluctuations of the nucleons in the neck at the moment of fission.

To these considerations one can add others by noting some of the features characteristic of heavy-ion-induced fission. First, these reactions are associated with large angular momenta of the fissioning systems. In the rotating liquid-drop model[4] the most important consequence of angular momentum is the lowering of the fission barrier. For nuclei lighter than the actinides this will strongly enhance the fission probability. For actinide nuclei, fission becomes the almost exclusive mode of deexcitation. While this influence of angular momentum on the fission probability seems to be at least qualitatively understood, the influence on the mass and charge distributions is less clear. For actinide nuclei liquid-drop model calculations predict the average fission barriers to be quite low or zero if shell effects are washed out, i.e., for excitation energies in excess of, say, 40 MeV. Under these conditions it is not clear if fission is still a compound nuclear process; especially, the transition state concept must break down. This may result in different charge and mass variances in fission.

A second aspect of heavy-ion-induced reactions, observed many times in the last years, is the appearance of direct reactions with deep inelasticity. In particular, ^{22}Ne- and ^{40}Ar-induced reactions on ^{232}Th have been found[5] to have important cross sections for large transfer of energy and mass between target and projectile. It is reasonable to expect the heavy actinide partner of such reactions to be highly excited and to fission. In the mass-yield distribution of Ar- and Kr-induced fission of ^{238}U, Kratz *et al.*[2] have found a major component believed to be due to fission following direct reactions.

Recently, complete independent-yield distributions for isotopes of rubidium and cesium in ^{11}B- and ^{22}Ne-induced fission of ^{238}U have been measured.[6] First moments and variances could be obtained with good accuracy. This makes it interesting to compare these data in more quantitative detail with the predictions of the statistical theory and to attempt to clarify the various aspects mentioned above.

In Sec. II we describe a simple statistical model for fission, using a two-spheroid configuration for the scission point and evaporation theory for the

fragment deexcitation. A comparison of the model calculations with the data of Ref. 6 is presented in Sec. III.

II. THEORY

A. Some characteristics of the investigated systems

In Table I we have summarized some of the characteristics of interest in the fissioning systems studied. The compound nuclei formed were ^{249}Bk and ^{260}No with excitation energies E^* larger than 60 MeV. Critical angular momenta l_c and average angular momenta $\langle l\rangle_{av}$ in the reactions were estimated with the Bass model.[7] The rotating liquid-drop[4] parameters x (fissility) and $\langle y\rangle_{av}$ (rotation parameter) are also given. The parameter $\langle y\rangle_{av}$ as well as the resulting average effective fission barrier $\langle B_f\rangle_{av}$ were evaluated for the average angular momentum. In the absence of shell effects, these barriers are seen to be close to zero. A fission-evaporation-competition calculation using ALICE[8] gives the average number $\langle \nu_{sc}\rangle_{av}$ of prescission neutrons (charged particle emission was found to be negligible). For ^{22}Ne + ^{238}U first chance fission is the dominating mode.

B. Approximate statistical theory for charge distribution in fission

The main assumptions of the statistical model for fission have been reviewed by Wilets.[9] The model is particularly attractive if shell corrections are small as we shall assume, because of the high excitation energies involved. As a consequence of the model assumptions one has to minimize the potential energy of the fissioning system at the moment, thought to be close to scission point, when statistical equilibrium is last established.

We approximate the scission point configuration by two spheroids in contact. This simple configuration has been used with some success in low energy fission.[10] We are here primarily interested in calculating the first moments and variances of isotopic distributions in fission. For the charge density (Z/A) degree of freedom the most important term in the potential energy at scission point is the asymmetry energy. In the Myers-Swiatecki mass formula[11] this term is predominantly (to about 75% for typical fission fragments) volume dependent, i.e., nearly independent of shape. Therefore it is believed that the restriction to spheroidal shapes is justified for the prediction of most probable fragment charge densities. Since a spheroid shape is determined by two parameters (c for the major half axis along the scission axis, and a for the half axis perpendicular to it), the coaxial two-spheroid configuration is described by eight parameters (A_i, Z_i, c_i, a_i; $i=1,2$). Mass and charge conservation and incompressibility of the two fragment volumes reduce these to four parameters. Two of these describe the mass (A_2/A_1) and charge split (Z_2/Z_1) and the remaining two are shape parameters. One can choose according to Nix[12]

$$P_s = 0.5\,[(a_1/c_1)^2 + (a_2/c_2)^2]\,, \tag{1}$$

$$P_{as} = (a_1/c_1)^2 - (a_2/c_2)^2\,, \tag{2}$$

i.e., one has a symmetry (P_s) and an asymmetry (P_{as}) parameter.

The potential energy to be minimized is written

$$W = E_{s_1}^{(0)}B_{s_1} + E_{s_2}^{(0)}B_{s_2} + 2x_1E_{s_1}^{(0)}B_{c_1} + 2x_2E_{s_2}^{(0)}B_{c_2} + Z_1Z_2(c_1+c_2)^{-1}B_{cc} - 4\pi d\gamma R_1R_2(R_1+R_2)^{-1}. \tag{3}$$

The first four terms are the usual liquid-drop terms for the surface energy and the Coulomb self-energy of the fragments (x fissility parameter; $E_s^{(0)}$ surface energy of a sphere; B_s and B_c shape parameters). The term proportional to Z_1Z_2 is the mutual Coulomb energy with the shape parameter B_{cc}.[13] The last term is a crude approximation to the mutual nuclear energy or proximity energy. We have chosen the form given by Bass,[7] where R is the curvature radius at the contact point, $d=1.35$ fm, and γ is related to the surface energy parameter of the Myers-Swiatecki mass formula. All terms associated with the spin of the system can be shown to be negligible for charge division equilibrium.

TABLE I. Characteristics of the fissioning systems studied. All symbols are explained in the text.

Fissioning system	E_{lab} (MeV)	E^* (MeV)	l_c	$\langle l\rangle_{av}$	x	$\langle y\rangle_{av}$	$\langle B_f\rangle_{av}$ (MeV)	$\langle \nu_{sc}\rangle_{av}$
^{11}B + ^{238}U → $^{249}_{97}$Bk	85.1	67.5	39.6	26	0.813	3.65×10^{-6}	2.5	0.7
^{22}Ne + ^{238}U → $^{260}_{102}$No	132.0	64.5	41.2	27.5	0.857	3.67×10^{-6}	1.0	0.3
	162.5	92.4	67.4	45.0		9.85×10^{-6}	0.24	<0.3

In the statistical theory the variances of any fission observable X (such as total kinetic energy, charge division, or mass division) are given by

$$\sigma^2(X) = T(2E_{pX})^{-1}, \tag{4}$$

where T is the nuclear temperature and

$$E_{pX} = \tfrac{1}{2}(\partial^2 W/\partial X^2)_{X=X_p}, \tag{5}$$

X_p being the most probable value of X found by minimization of the potential energy W. Equation (4) implies an expansion of W around its minimum to second order in X and classical Boltzmann statistics.

We see that the variances are directly related to the second derivatives of the potential in the vicinity of its minimum and can be found numerically by fitting a parabola around this minimum. For the nuclear temperature one can write approximately

$$aT^2 = E^*_{sc}, \tag{6}$$

where $a = \frac{1}{8}A$ (A mass of fissioning nucleus) and E^*_{sc} is the part of the energy at the scission point which can be identified with "heat." We shall come back to this definition later. Using Eqs. (4) and (6) one can use measured variances to obtain information on E^*_{sc}:

$$E^*_{sc} = \tfrac{1}{2}\sigma^4(X)AE_{pX}{}^2. \tag{7}$$

C. Time scale consideration

One of the main assumptions of the statistical theory for fission is that the descent from saddle point to scission point is a very slow process allowing nonfission degrees of freedom to be equilibrated at every step during the descent. It is important to check this assumption for charge equilibration.

Specializing Eq. (5) to mass and charge equilibrium, one can calculate two quantities, E_{pA} and E_{pZ}, which are connected respectively to the variation of mass keeping constant charges, and the variation of charge for a given mass division. The latter is the energy required, in the optimum configuration, to exchange a neutron from fragment 1 with a proton from fragment 2, while E_{pA} is the energy needed to shift a neutron from one fragment to the other. Calculations yield for E_{pA} values of 0.20–0.25 MeV, while E_{pZ} is found to be about 1.45–1.65 MeV. Using the uncertainty relation $\Delta\tau \approx \hbar/E_p$, one can estimate typical times to establish the equilibrium. They correspond to 0.4×10^{-21} s (constant A) and 2.8×10^{-21} s (constant Z). The charge equilibrium (constant A) establishes itself about 7 times faster than the mass equilibrium. The time for the mass equilibrium is so long that one has serious doubts about the justification of the statistical model. For the collective motion of a nonviscous liquid from saddle to scission Nix[12] obtains typical values of $(1-3)\times10^{-21}$ s which are no longer large compared to the 2.8×10^{-21} s mentioned above. We have therefore assumed that the fragment charge densities are determined by the faster mechanism, i.e., the adjustment to the optimum charge Z_p (A_1) for every given mass division $A_1/(A-A_1)$. Under this assumption one can also show that

$$E_{pA} \simeq E_{pZ}(Z/A)^2 \tag{8}$$

and, using Eq. (4),

$$\sigma_A{}^2 \simeq \sigma_Z{}^2(A/Z)^2, \tag{9}$$

where σ_A (σ_Z) is the mass (charge) variance for given Z_1 (A_1).

D. Fragment excitation energies

Before comparing calculated with experimental masses one has to take into account nucleon evaporation from the excited fragments. The sum of fragment excitation energies can be written

$$E^*_1 + E^*_2 = E_{c.m.} - \bar{E}_{kin} + Q_0, \tag{10}$$

where $E_{c.m.}$ is the center-of-mass kinetic energy in the entrance channel and $\bar{E}_{kin}$ is the average total kinetic energy in the exit channel which is calculated using

$$\bar{E}_{kin} = 0.680Z_1Z_2(A_1^{1/3}+A_2^{1/3})^{-1} + 22.2 \text{ (MeV)}, \tag{11}$$

which for $A_1 = A_2 = \frac{1}{2}A$ reduces to Viola's[14] semiempirical formula. For the ground state Q values Q_0, one can use the Myers-Swiatecki mass formula with the Lysekil parameters.[11] The excitation energy of one fragment is calculated in accordance with the prescriptions of the statistical model which require that the temperatures in both fragments must be equal at scission point. This leads to a subdivision of the heat energy proportional to the fragment masses. Furthermore, in the two-spheroid model also the deformation energy at scission point was found to be essentially proportional to the fragment mass. This can be understood from the known fact[10] that the softest fragments will always have the highest deformation energy in the minimum of the potential at scission point. In the liquid-drop model the "softness" is simply proportional to $Z_1{}^2/A_1$, and hence roughly proportional to A_1.

III. COMPARISON WITH EXPERIMENT AND DISCUSSION

A. Centroids of the isotopic distributions

Results of the two-spheroid calculation are shown in Table II. The potential, Eq. (3), was minimized numerically by varying the shape parameters, Eqs. (1) and (2), and the charge division ratio Z_1/Z_2 for a given mass division. One finds the well known fact that the light (heavy) fragments tend to have, on the average, lower (higher) most-probable masses A_p than the masses calculated from "unchanged charge densities (UCD)"

$$A_{\mathrm{UCD}} = Z_1 A/Z \,. \tag{12}$$

The difference $\Delta A_p = A_p - A_{\mathrm{UCD}}$ is given in column 5 of Table II. In addition to the systems studied here, we have done calculations for proton induced fission of the compound nucleus ^{239}Np, that was determined by Tracy *et al.*[15] in studies of ^{238}U fission induced by protons of 40 to 60 MeV. The calculated shift ΔA_p of -0.90 amu is in good agreement with the corresponding experimental average value of -1.04 ± 0.16 amu found in Ref. 15. The last column of Table II is the calculated total deformation energy D at scission point which we shall need later.

We performed evaporation calculations to obtain the average number ν of neutrons emitted using ALICE[8] (Table III). This calculation was done assuming zero intrinsic angular momenta for fragments. The column labeled ν_γ in the same table is a rough correction due to γ-n competition because the fragments have finite spins. It tends to decrease the number of emitted neutrons. Based on results published elsewhere,[16] we have estimated an upper limit on the intrinsic spins of the fragments to be given by

$$I_1 \approx 0.8 \langle l \rangle_{\mathrm{av}} A_1/A \,. \tag{13}$$

Assuming rigid rotor moments of inertia this leads to a rotation energy not available for neutron emission and hence a correction ν_γ for the emitted neutron number.

For ^{249}Bk the calculation was done assuming either no pre-scission neutron emission ($\nu_{sc} \doteq 0$) or $\nu_{sc} = 2$. Actually ν_{sc} is likely to be small (Table I). The theoretical mass A_s (the most probable mass after evaporation) is obtained by

$$A_s = A_{\mathrm{UCD}}(\nu_{sc}) + \Delta A_p - \nu + \nu_\gamma \,, \tag{14}$$

where

$$A_{\mathrm{UCD}}(\nu_{sc}) = Z_1 (A - \nu_{sc}) Z^{-1} \tag{15}$$

and

$$\Delta A_p = A_p - A_{\mathrm{UCD}}(\nu_{sc}) \,. \tag{16}$$

We have neglected the (very small) differences in the values of ΔA_p for the various fissioning nuclei $(A - \nu_{sc}, Z)$. As is shown in Table III, pre-scission emission of neutrons is not a critical factor in predicting theoretical masses. The indicated accuracy of the theoretical calculation is the cumulation of errors in the calculation of the fragment excitation energies, leading to an estimated error in ν of 0.3 amu, and of the uncertainties in ν_{sc} and ν_γ.

The theoretical masses A_s are compared with the experimental masses in the same table. First, we notice that for the ^{11}B induced fission we obtain excellent agreement between theory and experiment. No such agreement would have been obtained had we used for both fragments the masses A_{UCD} and ignored the shift ΔA_p. The agreement is also an indication that possible fission following direct reactions between ^{11}B and ^{238}U, which we ignored in the calculation, seems to have no measurable influence (within 0.4 amu) on the centroid of the distributions.

For ^{22}Ne induced fission (^{260}No) we see from Table II that the theoretical values tend to be lower than the experimental[6] ones. In the case of Cs the difference is as high as 2 amu, quite definitely outside the error estimates. We shall come back to this discrepancy later.

TABLE II. Results from a two-spheroid calculation.

Fissioning nucleus	Z_i	Z_1/Z_2	A_{UCD}	$\Delta A_p = A_p - A_{\mathrm{UCD}}$	E_{pZ} (MeV)	D (± 10 MeV)
$^{239}_{93}$Np	37 (Rb)	0.66	95.09	−0.90	1.65	28
$^{249}_{97}$Bk	37 (Rb)	0.62	94.98	−1.12	1.61	29
	55 (Cs)	0.76	141.19	+0.65	1.55	33
$^{260}_{102}$No	37 (Rb)	0.57	94.31	−1.35	1.56	39
	55 (Cs)	0.85	140.20	+0.44	1.48	38

TABLE III. Comparison of theoretical and experimental first moments of the isotopic distributions.

Compound nucleus	E^* (MeV)	Z_1	E_1^* (MeV)	ν_{sc}	ν (±0.3)	ν_γ	A_s (Calculated)	A_s (Measured)
$^{249}_{97}$Bk	67.5	37	36.1	0	4.11	0.14±0.1	89.85	
			29.3	2	3.41		89.79	
							89.8±0.4	89.84±0.05
		55	58.6	0	6.58	0.14±0.1	135.38	
			48.7	2	5.73		135.09	
							135.2±0.4	135.72±0.04
$^{260}_{102}$No	64.5	37	36.8	0	4.21	0.18±0.1	88.8±0.4	89.4 ±0.3
		55	65.7	0	7.18	0.20±0.1	133.5±0.4	135.8 ±0.4
	92.4	37	47.5	0	5.26	0.35±0.2	88.0±0.4	88.82±0.07
								88.0 ±0.3[a]
		55	80.3	0	8.48	0.35±0.2	132.3±0.4	134.36±0.09
								133.3 ±0.2[a]

[a] After correction for fission following deep inelastic collisions.

B. Variances

In Table IV we have calculated the heat energy at scission from the variances using Eq. (7) with the E_{pZ} values from Table II. One has to correct the experimental variances σ_s^2 for the variance due to the statistical fluctuations in the evaporation process (about 0.3 amu) obtained from the evaporation calculation and for the fluctuations in the fragment excitation energy which are a consequence of the experimentally known variance of the $\overline{E}_{kin}$. The corrected values σ^2 and the corresponding E_{sc}^* values are given in columns 5 and 6 of Table IV. We compare these values in the last column with the quantity $(E_1^* + E_2^* - D)$, i.e., the sum of the fragment excitation at infinity minus the deformation energy D at the scission point calculated with the two-spheroid model (Table II). This quantity can be considered to be an approximate estimate of the heat energy at the scission point. The indicated accuracy of about 10 MeV results primarily from the uncertainty in the calculation of D which cannot be expected to be very accurate in our simple model because of the restriction to spheroid shapes and the crude approximation of the proximity energy. Within these uncertainties it appears from a comparison of the last two columns of Table IV that the results are compatible with the statistical model interpretation for the ^{249}Bk compound system. The value of the level density parameter a used in Eq. (6) implies that there is full statistical equilibration of all the degrees of freedom. If some of these degrees of freedom were to be not fully equilibrated a smaller "effective" value for a would have to be taken, resulting in smaller values for E_{sc}^*. We note also that this type of comparison in Table IV has become meaningful primarily because of the good accuracy of the measured[6] isotopic distribution, since E_{sc}^* is proportional to (σ^4).

TABLE IV. Statistical interpretation of the variances of the isotopic distributions.

Compound nucleus	E^* (MeV)	Z_1	σ_s^2	σ^2	E_{sc}^*[a] (MeV)	$E_1^* + E_2^* - D$ (±10 MeV)
$^{249}_{97}$Bk	67.5	37	3.84±0.16	3.0 ±0.25	67±10	66
		55	3.95±0.14	2.95±0.25	60±10	70
$^{260}_{102}$No	92.4	37	5.41±0.22	4.54±0.30	155±10	92
			<4.2[b]	<3.3[b]	<82[b]	
		55	8.27±0.29	7.25±0.35	352±35	111
			4.25±0.28[b]	3.23±0.35[b]	70±16[b]	

[a] Using Eq. (7) with E_{pZ} from Table I.
[b] After correction for fission following deep inelastic collisions.

For the compound nucleus ^{260}No we have not used the data at the lower energy for this comparison because they are incomplete and thus do not have the required accuracy. At the higher incident energy for ^{22}Ne we calculate E^*_{sc} values which are totally unrealistic. The experimental variances[6] are too large to be compatible with statistical theory. This conclusion is in agreement with the results of Ref. 1.

C. Influence of fission following direct reactions

We have plotted in Fig. 1 the measured[6] isotopic distribution of Cs in the fission of ^{238}U induced by 162 MeV ^{22}Ne ions. The large variance of this distribution can be connected with the well developed shoulder visible on the heavy mass side of the yields. This suggests the possibility of a superposition of two different isotopic distributions originating from two fissioning mechanisms. In addition to compound nuclear fission there is, as mentioned in the introduction, the possibility of fission following direct reactions.

Some of the most prominent light reaction products found in the system ^{22}Ne+^{232}Th at 174 MeV (Ref. 5) are ^{22}Ne, ^{21}F, and ^{18}O nuclei; with a ^{238}U target the heavy partners would be ^{238}U, ^{239}Np, and ^{242}Pu (neglecting evaporation from the light partners). From the measured[5] kinetic energy distributions of the outgoing particles one can deduce that the excitation energies of the heavy fissioning partners were in a rather broad range in excess of their fission thresholds (~ 6 MeV). Tracy *et al.*[15] have measured Rb and Cs isotopic distributions in the reaction ^{238}U(p,f) at 40 MeV. Taking prescission evaporation into account,[15] the fissioning nuclei were essentially $^{238-n}$Np ($n = 2 \pm 1$) excited at 20 ± 10 MeV. These isotopic distributions should be approximately representative for fission following direct reactions of heavy ions such as ^{22}Ne with ^{238}U at incident energies of interest here. In Fig. 1 we have also plotted the cesium data of Ref. 15 with a normalization adjusted to reproduce the heaviest isotope cross sections ($A > 140$). The curve fits well into the bump and corresponds to (23 ± 6)% of the total area. An estimate of the complete fusion cross section with the Bass model and of the reaction cross section with standard optical-model parameters yields about 20% for nonfusion reactions. Recently, Viola *et al.*,[17] using the angular correlation technique, have determined a direct reaction fraction of (26 ± 5)% for the system ^{20}Ne+^{235}U at 175 MeV incident energy. We are encouraged to believe that the difference between the two curves in Fig. 1 represents approximately the Cs distribution for the fission of the compound nucleus ^{260}No. We have calculated for this difference distribution corrected first moments (Table III) and variances (Table IV). Similarly, from the Rb distribution at the same energy we have subtracted the same percentage amount (an approximation, since the Rb contribution from the second mechanism is likely to be larger) of the data of Ref. 15 for the Rb isotopic distribution in ^{238}U(p,f) at 40 MeV. The corrected first moments are now in better agreement with the theoretical values for fission after fusion. A drastic improvement (Table IV) is found for the corrected variances, which now lead to E^*_{sc} values approximately compatible with statistical model expectations.

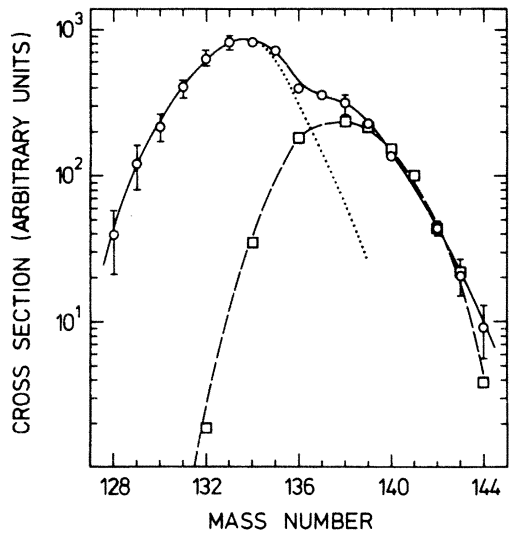

FIG. 1. Isotopic distribution of Cs in the fission of ^{238}U induced by ^{22}Ne [$E_{lab} = 162$ MeV, circles (Ref. 6)], and by protons [$E_{lab} = 40$ MeV, squares (Ref. 15)]. The distributions have been normalized relative to each other by adjusting the yields for masses heavier than 140. The dotted line is the difference distribution.

IV. CONCLUSION

We have analyzed rubidium and cesium isotopic distributions for ^{11}B and ^{22}Ne induced fission of ^{238}U. The highly excited compound nuclei in these systems have average liquid-drop barriers very close to zero. If one takes into account moderate contributions from fission following deep inelastic collisions, one can understand quantitatively the first moments and variances of the isotopic distributions in the framework of the statistical model of nuclear fission and of evaporation theory.

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