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Prompt neutron multiplicity distribution for $^{235}\text{U}(\text{n},\text{f})$ at incident energies up to 20 MeV

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Abstract: For the $\text{n}+^{235}\text{U}$ fission reaction, the total excitation energy partition of the fission fragments, the average neutron kinetic energy $\langle \varepsilon \rangle(A)$ and the total average energies $\bar{E}_\gamma(A)$ removed by γ rays as a function of fission fragment mass are given at incident energies up to 20 MeV. The prompt neutron multiplicity as a function of the fragment mass, $\nu(A)$, for neutron-induced fission of ^{235}U at different incident neutron energies is calculated. The calculated results are checked with the total average prompt neutron multiplicities $\bar{\nu}$ and compared with the experimental and evaluated data. Some prompt neutron and γ emission mechanisms are discussed.

Key words: fission fragment, excitation energy partition, prompt neutron multiplicity

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1 Introduction

The prompt fission neutron as the product of a fission reaction is an indirect probe of nuclear configurations near the scission point. Studying the prompt neutron multiplicity distribution (i.e. the prompt neutron multiplicity as a function of the fragment mass, $\nu(A)$) in detail can reveal some interesting characteristics of the fission process itself. In particular, it shines some light on how the total excitation energy (E_{tot}^*) available in the system is shared between the light and heavy fragments.

To the best of our knowledge, only a limited number of experimental data exist for $\nu(A)$. For ^{235}U , the $\nu(A)$ is only experimentally known in the case of $^{235}\text{U}(n_{\text{th}},\text{f})$. Although there are some theoretical calculations for the $\nu(A)$ of $^{235}\text{U}(\text{n},\text{f})$ [1–4], the partition of total excitation energy between the light and heavy fission fragments is a key and long-standing problem. In Ref. [2], the $\nu(A)$ of $^{235}\text{U}(n_{\text{th}},\text{f})$ was calculated with the point by point model, and a slow overestimation of the experimental points was observed in the heavy fragment mass region. In Ref. [4], the Monte Carlo approach was used to determine $\nu(A)$ for neutron-induced fission of ^{235}U at $E_{\text{n}}=0.53$ MeV with two different hypotheses for the partition of the E_{tot}^* . However, the results of $\nu(A)$ are not good enough, and the

calculated average $\bar{\nu}$ values are 2.73 and 2.67, respectively, and are larger than the experimental values.

This has stimulated the investigations into this topic. The main purpose of this paper is to focus on the $\nu(A)$ of higher incident neutron energies ($E_{\text{n}} > 5.0$ MeV). In this energy range, more than one fission chance will be involved, and the compound nuclei of $^{233-236}\text{U}$ undergoing fission can be formed. The fission channels of (n,f) , (n,nf) , $(\text{n},2\text{nf})$ and $(\text{n},3\text{nf})$ can open. In this context, the present work is the continuation of Ref. [5], where only the $\nu(A)$ distributions of $\text{n}+^{235}\text{U}$ at the incident neutron energy below 5.0 MeV are reported.

2 Theoretical approach

2.1 Total fission fragment excitation energy

The E_{tot}^* of the fission fragment is given as follows,

$$E_{\text{tot}}^* = E_{\text{r}} + B_{\text{n}}(A_{\text{c}}) + E_{\text{n}} - E_{\text{TKE}}(A_{\text{L}} + A_{\text{H}}), \quad (1)$$

where $B_{\text{n}}(A_{\text{c}})$ is the neutron separation energy of the fission compound nucleus, and subscript c refers to the compound nucleus. E_{n} is the kinetic energy of the neutron inducing fission. $E_{\text{TKE}}(A_{\text{L}} + A_{\text{H}})$ is the total kinetic energy of both light and heavy fragments, and

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is taken from experimental data. E_r is the energy released in the fission process, which can be calculated with Eq. (4) of Ref. [5].

2.2 Excitation energy partition

The key point for calculating $\nu(A)$ is to simulate the emission of neutrons from each fission fragment (FF), i.e., it is necessary to know how the E_{tot}^* is distributed among the light and heavy fragments. In previous work [5], we obtained the energy partition R_0 in the case of the thermal neutron, and this is the first time for giving an energy partition based on the experimental data without any theoretical hypotheses.

According to the temperature-dependent multi-mode fission model [6], the shell effect is responsible for the existence of the asymmetric fission mode, while the symmetric fission mode can be described by the liquid drop model. The probability of symmetric and asymmetric fission depends on the excitation energy of the fission compound nuclide. At a very low temperature (excitation energy), the asymmetric fission mode is dominant, and the energy partition is presented by the case of thermal neutron. With an increase in temperature (excitation energy), the shell effect becomes less important and disappears for high excitation energies (above 50 MeV), making the symmetric fission mode dominant. In this case, the energy partition R_L can be described as in the Los Alamos model [7]. If the excitation energy is between the two above-mentioned cases, both the shell effect and the liquid drop model will contribute to the energy partition.

Using this idea and the systematic parameters of the FF mass distribution of the $\text{n}+^{235}\text{U}$ fission system [8], in which the portion of symmetric and asymmetric fission as a function of incident energy was given, the energy partition at E_n can be deduced. In Ref. [8], the yields Y_s (symmetric fission) and $Y_{\text{as1}} + Y_{\text{as2}}$ (asymmetric fission) can be calculated for every energy point (< 20 MeV), here, $Y_s + 2(Y_{\text{as1}} + Y_{\text{as2}}) = 2$. With the above two energy partitions (R_0 and R_L) and the symmetric and asymmetric fission probabilities (Y_s and $Y_{\text{as1}} + Y_{\text{as2}}$), the energy partition R_{E_n} for a given energy $E_n (< 20 \text{ MeV})$ can be calculated as

$$R_{E_n} = (1 - R) \times R_0 + R \times R_L, \quad (2)$$

$$R = \frac{1}{2} \times \frac{Y_s}{Y_{\text{as1}} + Y_{\text{as2}}}.$$

With Eq. (2), the energy partitions at several energy points were calculated, and these are shown in

Fig. 1. One can see that the energy partition tends to the R_L line with increasing energy of the incident neutron.

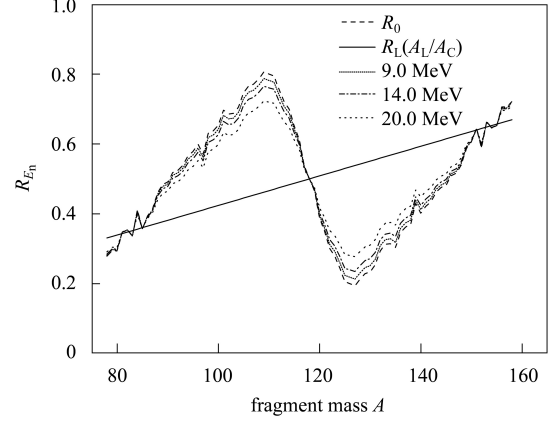


Fig. 1. The energy partition at different neutron energies.

With the energy partition R_{E_n} and the E_{tot}^* of FF, we can get the excitation energy of each FF, $E^*(A)$,

$$E^*(A) = R_{E_n} \times E_{\text{tot}}^*. \quad (3)$$

For a fragment with excitation energy $E^*(A)$, it could de-excite through emitting neutrons and γ rays. Let $\bar{E}_\gamma(A)$ denote the total energy released by γ ray, then the remaining energy $E_n^*(A) = E^*(A) - \bar{E}_\gamma(A)$ is available for neutron emission. The energy carried by the first emission neutron is $\langle \eta \rangle_{n1} = \langle \varepsilon \rangle_{n1}(A) + \frac{1}{2}B_{2n}(A)$ (where $B_{2n}(A)$ is the two neutron separation energy and $\langle \varepsilon \rangle_{n1}(A)$ is the average neutron kinetic energy), and the remaining energy can be expressed by $E_n^{1*}(A-1) = E_n^*(A) - \langle \eta \rangle_{n1}(A)$. If $E_n^{1*}(A-1) > 0$, then the second neutron emission will occur, and the remaining energy would be $E_n^{2*}(A-2) = E_n^{1*}(A-1) - \langle \eta \rangle_{n2}(A-1)$, where $\langle \eta \rangle_{n2}$ denotes the energy carried by the neutron emitted from $(A-1)$ nuclide, and can be written as $\langle \eta \rangle_{n2} = \langle \varepsilon \rangle_{n2}(A-1) + \frac{1}{2}B_{2n}(A-1)$. $E_n^{2*}(A-2)$ denotes the excitation energy of $(A-2)$ nuclide. This procedure continues until the remaining energy $E_n^{\nu*}(A-\nu) = 0$, then the emission neutron number $\nu(A)$ can be determined.

2.3 $\langle \varepsilon \rangle(A)$ and $\bar{E}_\gamma(A)$

For the thermal neutron case, the experimental data of $E_{\text{TKE}}(A_L + A_H)$, $\langle \varepsilon \rangle(A)$ and $\bar{E}_\gamma(A)$ are available. But no experimental data of $\langle \varepsilon \rangle(A)$ and $\bar{E}_\gamma(A)$ are available for the $\text{n}+^{235}\text{U}$ fission reaction at other energies. Therefore, some empirical approaches were used.

According to Ref. [9], an empirical expression for the average total γ emission energy of the $\text{n}+^{235}\text{U}$

system is as follows,

$$\langle E_{\gamma}^{\text{tot}}(E_n) \rangle = (6.600 \pm 0.03) + (0.0777 \pm 0.004)E_n (\text{MeV}). \quad (4)$$

Therefore, a linear relation in $\langle E_{\gamma}^{\text{tot}}(E_n) \rangle / \langle E_{\gamma}^{\text{tot}}(\text{th}) \rangle$ is used. $\langle E_{\gamma}^{\text{tot}}(\text{th}) \rangle$ is the experimental value for the case of a thermal neutron. For a given E_n ,

$$\bar{E}_{\gamma, E_n}(A) = \frac{\langle E_{\gamma}^{\text{tot}}(E_n) \rangle}{\langle E_{\gamma}^{\text{tot}}(\text{th}) \rangle} \times \bar{E}_{\gamma, \text{th}}(A). \quad (5)$$

Based on our investigation, Eq. (4) is only valid for (n,f) channel. For (n,nf), (n,2nf) and (n,3nf) channels, the numerator of Eq. (5) becomes

$$\begin{aligned} \langle E_{\gamma}^{\text{tot}} \rangle &= (5.297 \pm 0.03) + (0.0777 \pm 0.004)E'_n, \\ \langle E_{\gamma}^{\text{tot}} \rangle &= (6.845 \pm 0.03) + (0.0777 \pm 0.004)E''_n, \\ \langle E_{\gamma}^{\text{tot}} \rangle &= (5.743 \pm 0.03) + (0.0777 \pm 0.004)E'''_n. \end{aligned} \quad (6)$$

5.297, 6.845 and 5.743 are the neutron separation energy of the compound nuclei ^{235}U , ^{234}U and ^{233}U , respectively. E'_n , E''_n and E'''_n are the equivalent incident neutron energy for the (n,nf), (n,2nf) and (n,3nf) channels. For example, when a 9 MeV neutron induces ^{235}U fission, (n,f) and (n,nf) channels will open. For a (n,f) reaction, the compound nucleus is ^{236}U , and the excitation energy is $9 + 6.546 = 15.546$ (MeV). For a (n,nf) channel, ^{236}U emits a neutron (with energy $\varepsilon = 1.056$ MeV) and becomes ^{235}U with the excitation energy $E^* = 15.546 - 1.056 - 6.546 = 7.944$ (MeV). This can be considered as a neutron with energy E'_n which induces ^{234}U fission, and forms the compound nucleus ^{235}U with excitation energy of 7.944 MeV. Therefore, we can determine the $E'_n = 7.944 - 5.297 = 2.647$ (MeV). This method is also extended to other fission channels and other energy points.

For the average neutron kinetic energy $\langle \varepsilon \rangle_{E_n}(A)$ (at E_n) as a function of the mass number of the FF, according to the statistical theory [10], the average energy of a Maxwell evaporation spectrum $\langle \varepsilon \rangle = \frac{3}{2}T_M$. T_M is the nuclear temperature and is proportional to the excitation energy of the FF, $\sqrt{E^*}$. So a relation of $\langle \varepsilon \rangle_{\text{exp, th}}(A)$ and $\langle \varepsilon \rangle_{E_n}(A)$ is given for a given initial FF,

$$\langle \varepsilon \rangle_{E_n}(A) = \sqrt{\frac{E_{E_n}^*(A)}{E_{\text{th}}^*(A)}} \times \langle \varepsilon \rangle_{\text{exp, th}}(A), \quad (7)$$

where $E_{\text{th}}^*(A)$ is the excitation energy of the FF obtained from the E_{tot}^* for the case of a thermal neutron, and $E_{E_n}^*(A)$ is the excitation energy of the FF gained from E_{tot}^* at E_n . $\langle \varepsilon \rangle_{\text{exp, th}}(A)$ are the experimental values for the case of a thermal neutron.

According to the experimental data, it is found

that the total FF kinetic energy E_{TKE} almost does not change at incident energies up to 20 MeV in the $n + ^{235}\text{U}$ fission reaction, which is reasonable because E_{TKE} is the result of the Coulomb repulsion. Therefore, in this work, the E_{TKE} is assumed to be independent of the neutron incident energy.

3 Calculations of $\nu(A)$

For the $n + ^{235}\text{U}$ reaction, when incident neutron energy $E_n > 5.0$ MeV, the compound nuclei ^{236}U , ^{235}U , ^{234}U and ^{233}U undergoing fission will be formed in succession. Therefore, the multi-channel fission must be taken into account, and the calculations will become more complex.

Only the (n,f) channel opens at $E_n < 5.0$ MeV; (n,f) and (n,nf) channels will open when $5.0 \text{ MeV} < E_n < 10.0$ MeV; at $10 \text{ MeV} < E_n < 15.0$ MeV, (n,f), (n,nf), and (n,2nf) channels will open; and (n,f), (n,nf), (n,2nf) and (n,3nf) channels open when $E_n > 15.0$ MeV.

For the (n,f) channel, the compound nucleus is ^{236}U , so we calculated the $n + ^{235}\text{U}$ reaction with the neutron energy E_n , and the prompt neutron multiplicity as a function of fragment mass $\nu(A)_{236}$ is obtained.

For the (n,nf) channel, the compound nucleus is ^{235}U , so, the $n + ^{234}\text{U}$ reaction with the neutron energy E'_n is investigated, and $\nu(A)_{235}$ is obtained.

For the (n,2nf) and (n,3nf) channels, the $\nu(A)_{234}$ and $\nu(A)_{233}$ are determined in a similar way.

At a given E_n , the total $\nu(A)$ is calculated as a superposition of the $\nu(A)$ for each fission channel weighted by the fission cross-section, taking into account the multi-channel fission. It can be expressed as

$$\begin{aligned} \nu(A)_{\text{tot}} &= \left[\nu(A)_{236} \times \left(1 + \frac{0}{S_{236}} \right) \right] \times \frac{\sigma_{f236}}{\sigma_F} \\ &+ \left[\nu(A)_{235} \times \left(1 + \frac{1}{S_{235}} \right) \right] \times \frac{\sigma_{f235}}{\sigma_F} \\ &+ \left[\nu(A)_{234} \times \left(1 + \frac{2}{S_{234}} \right) \right] \times \frac{\sigma_{f234}}{\sigma_F} \\ &+ \left[\nu(A)_{233} \times \left(1 + \frac{3}{S_{233}} \right) \right] \times \frac{\sigma_{f233}}{\sigma_F}, \end{aligned} \quad (8)$$

where σ_F is the total fission cross-section, and σ_{f236} , σ_{f235} , σ_{f234} , σ_{f233} are the fission cross sections of the (n,f), (n,nf), (n,2nf) and (n,3nf) channels, respectively. The numerators 0, 1, 2 and 3 stand for the numbers of pre-fission neutrons evaporated from the compound nucleus of ^{236}U . The denominator S_i is a

factor to describe the pre-fission neutron and is assumed to be uniformly distributed for each mass A . It can be written as $S_i = \sum_A \nu(A)_i \times Y_{\text{CHN}}(A)_i$, with i standing for 236, 235, 234 or 233. $Y_{\text{CHN}}(A)$ is the chain yield of mass chain A . The chain yield of the $\text{n}+^{235}\text{U}$, $\text{n}+^{234}\text{U}$, $\text{n}+^{233}\text{U}$ and $\text{n}+^{232}\text{U}$ systems used in the calculations are taken from the ENDF/B-VII library [11]. In the cases where no evaluated data exist, the linear interpolation or extrapolation was adopted.

Since no experimental data are available for the calculated $\nu(A)$ at these energies except for the thermal neutron, the calculated results cannot be checked directly, but can be checked indirectly by calculating the total average prompt fission neutron multiplicity $\bar{\nu}$, according to Eq. (9). For $\bar{\nu}$, plenty of experimental and evaluated data are available,

$$\bar{\nu} = \sum_A [\nu(A)_{\text{total}} \times Y_{\text{CHN}}(A)_{236}]. \quad (9)$$

4 Results and discussions

With Eqs. (1–8) and the energy partitions at different neutron energies (Fig. 1), the prompt fission neutron multiplicity distributions $\nu(A)$ for the $\text{n}+^{235}\text{U}$ fission are calculated at 9.0 MeV, 14.0 MeV and 20 MeV, as shown in Fig. 2.

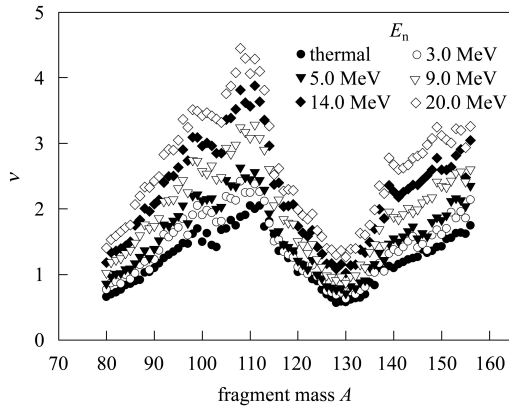


Fig. 2. The prompt neutron multiplicity ν as a function of the FF mass for the $\text{n}+^{235}\text{U}$ reaction.

The calculated results at $E_n < 5.0$ MeV in Ref. [5] are also presented for comparison. The behavior of the calculated sawtooth data $\nu(A)$ shows an obvious dependence on the incident neutron energy consisting in the increase of the prompt neutron numbers emitted by each individual fragment and in a slow flattening of the sawtooth character at mass number $A \approx 100$ when E_n is increasing. The same behavior is found in Ref. [3] for $^{237}\text{Np}(\text{n},\text{f})$.

A comparison of the calculated $\bar{\nu}$ (Eq. (9)) with the evaluated values [12](line) and experimental val-

ues [13](open symbols) are shown in Fig. 3. We also present the results of the thermal neutron, 3.0 MeV and 5.0 MeV, which are taken from Fig. 6 of Ref. [5]. These show that the present calculations reproduce the total average prompt fission neutron multiplicity $\bar{\nu}$ well, and the largest difference is less than 3.0% at 9.0 MeV.

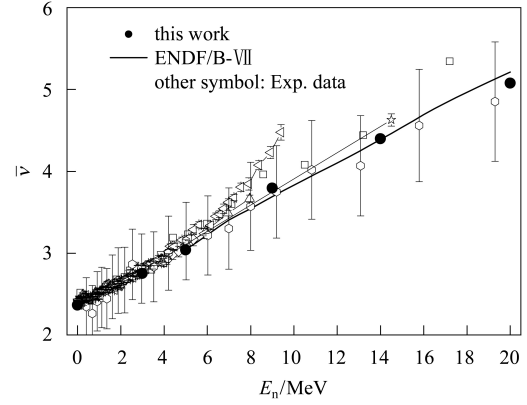


Fig. 3. Comparison of the calculated $\bar{\nu}$ with the experimental data and the evaluated data.

The good agreement of the total average prompt fission neutron multiplicity $\bar{\nu}$ illustrates that the calculated results of $\nu(A)$ are acceptable, and also indicates that the total excitation energy partitions among the light and heavy fragments in this work are reasonable.

According to Eqs. (4) and (6), although the $\langle E_{\gamma}^{\text{tot}} \rangle$ depends on the incident neutron energy E_n , the main component of $\langle E_{\gamma}^{\text{tot}} \rangle$ is neutron separation energy, and the factor on the E_n is small. Therefore, we can conclude that there exists γ competition in the course of neutron emission, but the competition is very weak.

The experimental data of $\langle \epsilon \rangle(A)$ are not available for the $\text{n}+^{235}\text{U}$ fission except for the thermal neutron case. The $\langle \epsilon \rangle(A)$ distributions used in this work are obtained according to the statistical theory. The present results show that this method is reasonable.

The agreement between the calculated $\nu(A)$ and the experimental data at thermal neutron energy could be given with the sum of neutron numbers emitted by the light fragment and heavy fragment of a FF pair as function of the FF mass number. It can be seen from Fig. 4(a) that the calculated results are in good agreement with the experimental data.

Figure 4(b) shows the neutron multiplicity of the FF pair at higher incident neutron energies where experimental data do not exist. It can be seen that the number of neutrons emitted by the FF pair increases with the incident neutron energy E_n , and the number

of neutrons emitted by each of the complementary fragments is approximately constant (i.e. independent of A) for a given E_n , except for some structure near symmetric mass region. In addition, the dips occur near $A_H=130$, and in the complementary FF,

near $A=104$. This is probably related to the proximity of Z and N of the heavy fragment of the spherical $Z=50$ and $N=82$ shells, where the fragment kinetic energy is high, therefore, the excitation energy and neutron emission are small.

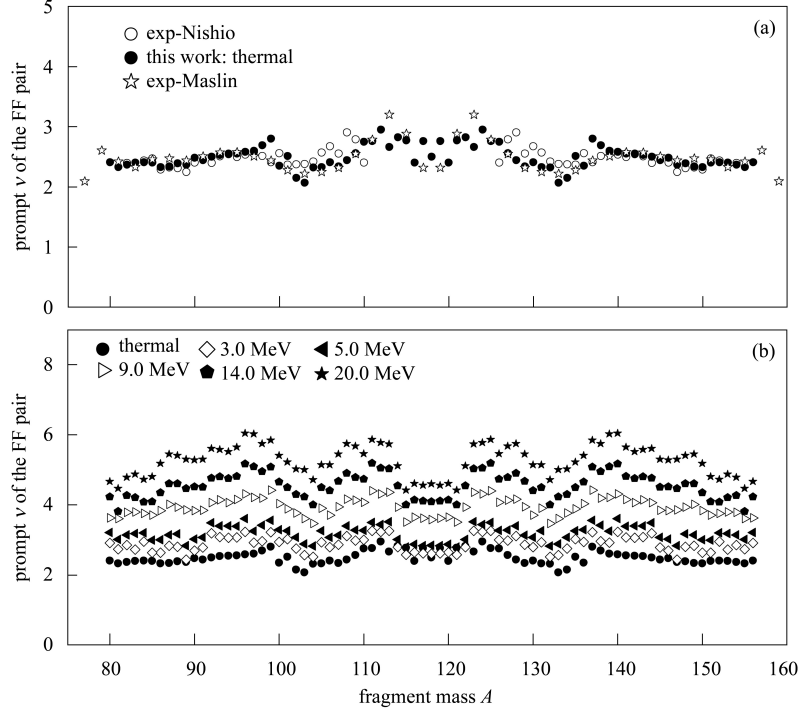


Fig. 4. FF pair multiplicity of the $^{235}\text{U}(n,f)$ reaction at thermal neutron energy (a) and higher neutron energies (b).

5 Conclusion

The total excitation energy partitions between the complementary light and heavy fission fragments for the $n+^{235}\text{U}$ fission reaction are given at incident energies up to 20 MeV. The average neutron kinetic energy $\langle\epsilon\rangle(A)$ and the total average energies removed by γ rays $\bar{E}_\gamma(A)$ as a function of FF mass at different incident neutron energies are presented. The prompt neutron multiplicity distribution $\nu(A)$ for $n+^{235}\text{U}$ fis-

sion at different incident neutron energies are calculated. The results are checked with the total average prompt neutron multiplicities $\bar{\nu}$ and compared with the experimental and evaluated data. These results can be used for the prompt fission neutron multiplicity and spectrum evaluation, and the calculated $\nu(A)$ distribution provides the data to deduce the post-neutron emission mass yields from the already known pre-neutron emission mass yields. The former is very important for the application.

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