Isotopic distributions in deep inelastic reactions induced by heavy ions and the relaxation of the neutron excess degree of freedom

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The available experimental results on the equilibrium of the N/Z degree of freedom are recalled. New available experimental results for very asymmetric systems are presented and compared to a simple classical model including the mass diffusion, the relaxation of the neutron excess degree of freedom, and the subsequent evaporation. The agreement between this classical calculation and the experiment is shown to be satisfactory.

[NUCLEAR REACTIONS $^{40}Ar+^{197}Au$, $^{32}S+^{238}U$, $^{40}Ar+^{238}U$, $^{63}Cu+^{232}Th$, $^{40}Ar+^{232}Th$. Deeply inelastic reactions. Comparison of experimental distributions with a classical model.]

I. INTRODUCTION

Among all the collective degrees of freedom involved in deep inelastic reactions, the most important are certainly the neutron to proton ratio (or neutron excess), the relative motion, the rotational degrees of freedom, and the mass asymmetry.[1]

When two nuclei characterized by different neutron to proton ratios (N/Z) come into contact, and if the N/Z degree of freedom is relaxed, then the most probable ratio Z_1/Z_2 for a given mass asymmetry is the one which minimizes the potential energy of the system. This ratio can be very different from the ones of the projectile and target nuclei. This degree of freedom has been shown to be the most rapidly relaxed.[2] Experimental results give a relaxation time close to $1-2\times10^{-22}$ s. In contrast, the slowest relaxation mode corresponds to the mass asymmetry degree of freedom ($t_r \sim 60\times10^{-22}$ s).[1]

Isotopic distribution measurements in deep inelastic reactions are needed for studying the relaxation of the neutron excess collective degree of freedom. Several experiments were performed some years ago at Orsay,[3-5] and more recently at Darmstadt[6] on medium-mass systems (Ar+Ni, for example). The study of such systems with masses close to symmetry in the entrance channel and large bombarding energies have nevertheless some disadvantages. Indeed, the damping of the relative motion leads to relatively high excitation energies of the composite system. Furthermore, in a symmetric system, the two fragments are on the average equally highly excited and the subsequent evaporation process can be very complex. Therefore knowledge of the primary distributions cannot be easily estimated. Moreover, in such systems the N/Z ratios for the reaction products are not very far from that of the projectile (or the target), and it becomes difficult to conclude whether the final N/Z is reflecting the conditions of equilibration or of the entrance channel.

For these reasons, we have chosen to study very mass-asymmetric systems with relatively light projectiles such as S or Ar, and heavy, neutron-rich targets such as ^{238}U at energies just above the interaction barrier. In such cases, the N/Z equilibration should lead to a considerable neutron enrichment of nuclei close to the projectile and even to the production of new nuclides.[7] In such conditions, it should be easier to make a clear distinction between deep inelastic and quasielastic products (as the isotopic distribution will often be different). Furthermore, if equal temperatures are reached for both light and heavy fragments,[8] then the excitation energy of the light one will be very low and light particle decay following the primary process will be minimized. Thus, in considering the observed isotopic distributions we expect to have almost a direct view of the primary processes, only weakly perturbed by the secondary evaporation processes.

Several authors[9,10] have suggested that the excitation of the giant dipole resonance could be responsible for relaxation of the N/Z degree of freedom. In this respect, the calculated relaxation time appears to be compatible with the experimental one.[2] It has also been suggested that

quantum effects could be seen if the associated phonon energy is larger than the temperature of the system. In this case, the width of the distribution would rather reflect the zero point oscillation than the Brownian dispersion. In the following we shall show that it is not necessary to invoke the dipole giant resonance to explain the experimental data. A classical statistical model close to the one first used by Braun-Munzinger and Barrette[11] is shown to reproduce quite nicely the isobaric distributions.

In this paper, we will first review the main characteristic features on the equilibrium of the N/Z degree of freedom in the light of the first published experimental data.[2-5,12-15] Then a simple classical model including mass diffusion, the relaxation of the neutron excess degree of freedom, and the subsequent nuclear evaporation is developed. The model is applied to recent results obtained by our group for the systems Ar + Au (Ref. 12) and Ar + U (Ref. 7). Furthermore, new results concerning the system S(241 MeV) + U are presented (for experimental setup see Ref. 7). Comparison is also extended to the two systems (Cu + Au) and (Ar + Th) investigated by others.[13,14] It is shown that the agreement is satisfactory between this classical model and all the experiments.

II. EXPERIMENTAL EVIDENCE FOR THE FAST EQUILIBRATION OF THE NEUTRON TO PROTON RATIO DEGREE OF FREEDOM

Several experiments were performed during the past three years in order to study the relaxation of this collective mode.[2-7,12-15] One of the first sets of data is shown in Fig. 1 where potassium isotopic distributions are compared for the two systems (^{40}Ca + ^{64}Ni) and (^{40}Ar + ^{58}Ni).[3] For these two systems, the N/Z ratios for the composite system are very similar; however, in the first case, the projectile is neutron-poor as compared to the target and in the second case it is neutron-rich. The quasielastic components are very different in the two cases; in the first one, the most probable transfer corresponds to one proton stripping leading to ^{39}K whereas one proton has been picked up by the projectile in the second system, leading to ^{41}K. On the other hand, in the energy relaxed components, the mass distributions are very similar as one would expect from an equilibration of the neutron-excess collective degree of freedom.

Another convincing case for this equilibration has been given by the study of the systems (^{40}Ar + ^{64}Zn) and (^{40}Ca + ^{64}Ni) at the same bombarding energy and with exactly the same number of protons and neutrons.[5] If this relaxation mode is thought to be the fastest of all collective modes and in particular faster than the mass asymmetry mode, then the two systems are expected to reach the same N/Z asymmetry before any mass rearrangement could appear. Therefore, with this picture the same primary product distributions should result and then the same final distribution should be observed after evaporative deexcitation because the excitation energy and angular momentum distribution are very much the same (same c.m. bombarding energy). The results, given in Fig. 2, show clearly that the distributions are very similar. This strongly suggests that the neutron to proton ratio asymmetry relaxation mode is reached much faster than the mass asymmetry mode.

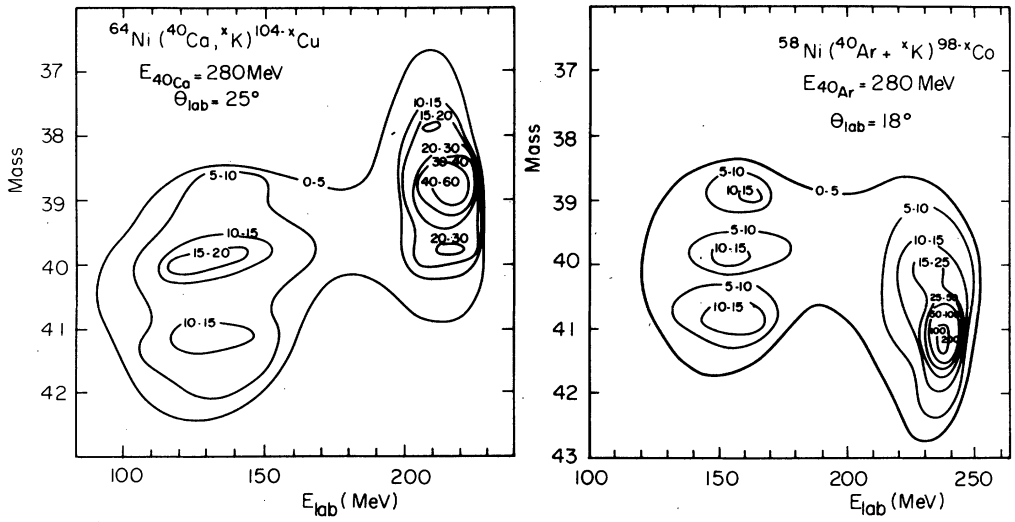

FIG. 1. Contour plot of the isotope yields for K in the mass-energy plane for the reactions ^{40}Ar (280 MeV) + ^{58}Ni and ^{40}Ca (280 MeV) + ^{64}Ni (Ref. 3).

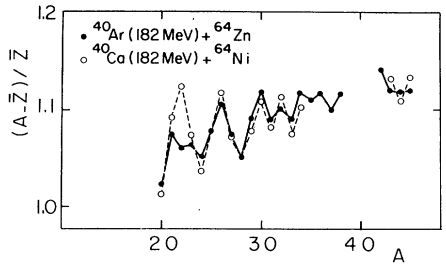

FIG. 2. Most probable ratio $(A-\bar{Z})/\bar{Z}$ as a function of product mass for the two systems ^{40}Ca + ^{64}Ni and ^{40}Ar + ^{64}Zn at 182 MeV bombarding energy (Ref. 5).

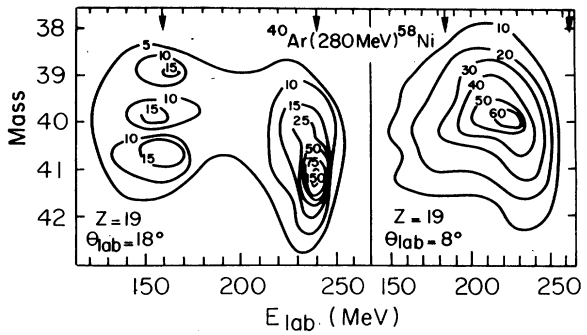

FIG. 3. Contour plot of K isotope yields in the mass-energy plane for the reaction ^{40}Ar (280 MeV) + ^{58}Ni at 18° (close to the grazing angle) and 8° (below the grazing angle, Ref. 2).

From these studies (Figs. 1 and 2), it appears that the N/Z equilibration is reached for completely damped collisions, but it is far from being attained for quasielastic reactions (Fig. 1). An important aspect not yet addressed is the transition between these two situations.[2] The equilibration of the N/Z degree of freedom has been investigated for incompletely damped intermediate collisions in the reaction Ar + ^{58}Ni at 280 MeV bombarding energy. As shown in Fig. 3, near the grazing angle (θ_{lab} = 18°) the two components are clearly separated; on the other hand, for θ_{lab} = 8°, even when as little as 15 to 20 MeV have been lost in the relative motion, the mass distribution looks very similar to the one observed at θ = 18° for completely damped events. This shows very clearly that the N/Z degree of freedom is relaxed much more readily than the relative motion. A characteristic relaxation time has been estimated to be of the order of 10^{-22} s by considering the average rotational velocity and the angle to which the products are detected.[2]

Recently, other experiments have been performed with heavy targets (for example, Ar + U,[7] and this work) which will be quantitatively described in Sec. IV. Nevertheless, an overview of results obtained for the system 263 MeV ^{40}Ar + U is presented in Fig. 4. The number of events for each nuclide (measured at θ_{lab} = 60°) is indicated in the mass-charge space. Very n-rich nuclei have been produced in this experiment which clearly demonstrates that deeply inelastic reactions with heavy ions are excellent tools for producing new n-rich isotopes.[7] Several other results have been obtained recently for other very asymmetric systems. They will be presented in Sec. IV for comparison with model calculations. They concern, in addition to the Ar + U system, the following reactions: ^{40}Ar + ^{197}Au (E_{lab} = 217 MeV),[12] S + U (E_{lab} = 241 MeV) (this work), ^{63}Cu + ^{197}Au (E_{lab} = 445 MeV),[13] and Ar + Th (295 MeV).[14] It should be mentioned that the experimental technique used for the S + U reaction study is that described in Ref. 7. These reactions have usually been studied at relatively low energies above the interaction barrier. Thus

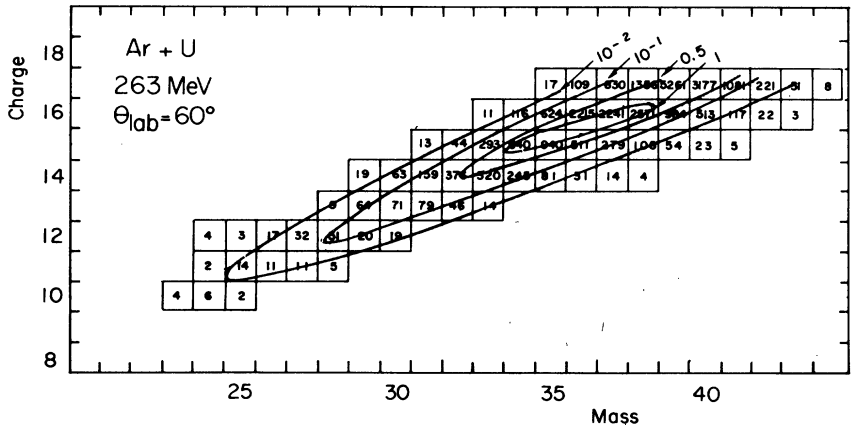

FIG. 4. Experimental nuclide distribution in deep inelastic collisions induced by the reaction ^{40}Ar (263 MeV) + ^{238}U (in number of events). These results are compared to the calculation represented by contour plots of isoprobability (for more details, see Sec. IV).

the light primary fragments (projectile-like) are left relatively unexcited, and the subsequent deexcitation process plays a minor role. Furthermore, in such very asymmetric systems, the light fragments are very neutron-rich and decay mainly by neutron and γ-ray emission. These systems are thus well chosen to investigate whether or not the N/Z degree of freedom is relaxed for the widely varying initial N/Z ratios corresponding to the projectile and the target (respectively 1 and 1.59 for S+U, 1.22 and 1.59 for Ar+U, 1.17 and 1.49 for Cu+Au). Additionally, it is interesting to study the influence of the bombarding energy. For this we can choose two neighboring systems for comparison in which the bombarding energies to interaction barrier ratios (E/V_I) are quite different: Ar+Au ($E/V_I = 1.13$) and Ar+Th ($E/V_I = 1.42$).

III. DESCRIPTION OF THE CLASSICAL MODEL

The aim of the analysis is to account for both isotopic and isobaric distributions by application of very simple classical concepts. The basic idea, suggested by the experimental data, is to assume that the N/Z degree of freedom is much more rapidly relaxed than the mass asymmetry mode. Therefore, at any step of the nucleon exchange between the nuclei, i.e., for any mass asymmetry, the N/Z degree of freedom has had enough time to equilibrate. Our model calculations are based on three assumptions:

(i) For a given mass ratio in the exit channel, the N/Z distribution is governed by the potential energy of the composite system. If a full statistical equilibrium is reached for this degree of freedom, then the charge distributions should obey the Boltzmann equation.

(ii) In order to account for the mass distribution of the fragments, a classical diffusion model has been utilized.[18]

(iii) Finally, to account for the excitation energy left in the fragments and for the subsequent particle emission, a very simple evaporation model is employed.

A. The potential energy of the composite system and the charge distribution

The shape of the composite system is approximated by a configuration of two spherical liquid drops with a separation distance d. The potential energy of the system is then given by the following expression:

$$V(Z_1, A_1, Z_2, A_2) = V_{\mathrm{LD}}(Z_1, A_1) + V_{\mathrm{LD}}(Z_2, A_2) + V_C + V_{\mathrm{ROT}} - V_{\mathrm{EC}} . \quad (1)$$

Z_1, A_1, Z_2, A_2 are respectively the charge and mass of each drop, V_{LD} denotes the liquid drop potential energy of each fragment, V_C the Coulomb potential between the two drops, V_{ROT} the rotational energies of the system. The total potential energy in the entrance channel is V_{EC} (the potential energy of the system is normalized to zero for the exit channel identical to the entrance channel). All liquid drop potential energies have been calculated using the parameters of Myers and Swiatecki.[16]

As will be seen from the experimental kinetic energies of the fragments, the composite system has an extended configuration at the scission point. Therefore, the spherical approximation with the separation distance d used in the calculation of the potential energy might be very crude. Nevertheless, the deformation energies (and also the nuclear potential energy) have not been taken into account in the calculation. As will be seen further, the mass distribution for a given Z does not depend on the absolute value of the potential energy V but on the derivative dV/dM. We have considered that the deformation energies and the nuclear potential energy vary very slowly with the mass asymmetry, and therefore we take it to be constant in the narrow zone of interest, which seems to be a reasonable approximation.

It is very well known that, owing to the tangential friction, the entrance channel orbital angular momentum is shared between intrinsic angular momentum of the reaction products and relative angular momentum. If one assumes sticking, i.e., rotation of a rigid dinuclear system, the orbital angular momentum in the exit channel is found to be the following fraction (l_f) of (l_i), the initial one:

$$\langle l_f \rangle = \langle l_i \rangle \frac{\mu R^2}{\mu R^2 + \frac{2}{5} M_1 R_1^2 + \frac{2}{5} M_2 R_2^2} , \quad (2)$$

where R is the distance between the two fragments, μ the reduced mass, and R_1, R_2 the radius of each fragment. The mean value of the entrance channel orbital angular momentum for these deep inelastic collisions is denoted by $\langle l_i \rangle$

$$\langle l_i \rangle = [(l_{\max}^2 + l_{\mathrm{crit}}^2)/2]^{1/2} .$$

The critical angular momentum for fusion l_{crit} has been calculated assuming the critical distance concept.[17] The Coulomb potential V_C has been adjusted so as to reproduce the experimental kinetic energies in the exit channel. Since complete damping of the relative motion has been achieved, the kinetic energy E_1 of a fragment (Z_1, A_1) can be expressed as the sum of Coulomb and centrifugal energies,

$$E_1 = E_C + E_l = \frac{A_2}{A_1 + A_2} \left(\frac{Z_1 Z_2 e^2}{R} + \frac{l_f(l_f+1)\hbar^2}{2\mu R^2} \right), \quad (3)$$

where R is the distance between the centers of mass:

$$R = r_0(A_1^{1/3} + A_2^{1/3}) + d .$$

We take r_0 to be 1.225 fm and d is an adjustable parameter in the calculation. For a configuration of two spherical liquid drops in contact, the parameter d should be close to 2 fm. A value of d of 4 ± 0.5 fm was chosen as this value reproduced fairly well the mean experimental kinetic energies for all the systems under consideration in this study. (This rather large value of d shows clearly the extended configuration of the composite system at the scission point.)

With this expression for the potential energy of the composite system, the corresponding charge distribution $P(Z)$ for a given mass asymmetry is expressed by a Boltzmann distribution:

$$P(Z) \propto e^{-V(Z)/T} . \quad (4)$$

The nuclear temperature T is related to the thermal excitation energy E^* ($E^* = AT^2/8$). The value of E^* is obtained by assuming two-body breakup with kinetic energy of the fragments calculated from the expression (3).

B. Diffusion along the mass asymmetry degree of freedom

The probability for producing a fragment (Z_1, A_1) can be expressed by the equation

$$\mathcal{P}(Z_1, A_1) = \frac{P(Z_1, A_1)}{\sum_Z P(Z, A_1)} P(A_1) , \quad (5)$$

where $P(Z, A)$ is obtained according to Eq. (4) and $P(A_1)$ is extracted from a diffusion model.

The transport theory of Nörenberg[18,19] has been used to describe the time evolution of the mass distributions. Thus $P(A_1)$ can be expressed by the relation

$$P(A_1) = (4\pi D_A t_R)^{1/2} \exp[-(A_1 - A_p - V_A t_R)^2 / 4 D_A t_R] , \quad (6)$$

where A_p is the projectile mass. The reaction time t_R can be easily calculated from θ the angle of rotation, θ_{gr} the grazing angle, and ω the angular velocity of the system as taken from the sticking model:

$$t_R = \frac{1}{\omega}(\theta_{gr} - \theta) .$$

The drift velocity V_A and the diffusion coefficient D_A have been obtained from the simple analytical expressions given in Ref. 18. When experimental mass distributions were compared to those calculated from Eq. (6), it was found that the theoretical transport coefficients can deviate from those deduced from the experiment by as much as a factor of 2. Nevertheless, the calculated isotopic distributions are not very sensitive to the choice of the transport coefficients (as will be shown later). Accordingly, theoretical values of V_A and D_A have always been used in these calculations of isotopic distributions.

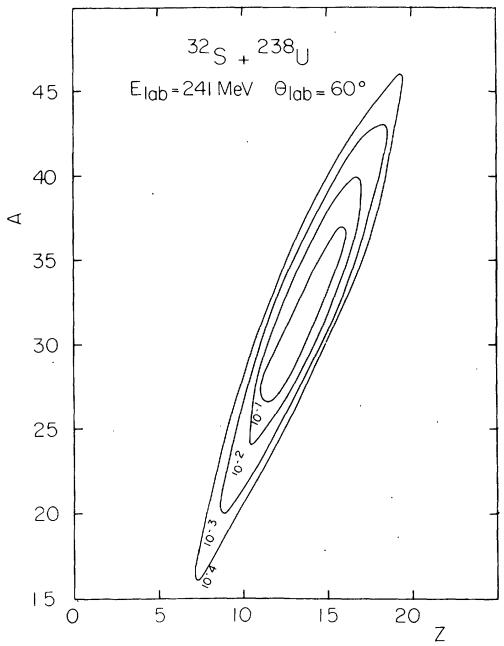

FIG. 5. Calculated yield distribution before the sequential decay process, for the $^{270}_{108}X$ composite system ($^{32}S + ^{238}U$ reaction).

An example of a calculated yield distribution is given in Fig. 5 for the composite system $^{270}_{108}X$. This distribution, centered around $Z_1 = 14$, $M_1 = 32$, resembles very much the experimental one, although it does not take into account the secondary particle evaporation process from the excited fragments.

C. The sequential decay process

Most of the experiments under consideration have been performed at relatively low bombarding energies above the interaction barrier; therefore the total excitation energy of the composite system does not exceed 60 MeV. Moreover, since thermal equilibrium between the fragments was indicated by various experiments,[20-22] the mean excitation energy of the light fragment is never expected to be >10 MeV. In addition, since heavy neutron-rich targets have been used, the light fragments are very n-rich and neutron evaporation is their primary mode of decay. At this low excitation energy, proton and α-particle evaporation can be neglected.

We have assumed that the primary kinetic en-

TABLE I. Mean entrance channel characteristics for all systems under study and associated diffusion model coefficients.

System	E_{lab}	$E_{c.m.}/V_I$	$\langle l_i \rangle_{DIC}$	t_R (10^{-22} s)	D_A (10^{22} u^{2}s^{-1})	V_A (10^{22} u s^{-1})	Ref.
^{40}Ar + ^{197}Au	217	1.13	68	11.8	3.8	1.5	12
^{40}Ar + ^{232}Th	295	1.42	134	7.5	3.9	1.6	14
^{40}Ar + ^{238}U	263	1.24	106	2.8	3.9	2.0	7
^{32}S + ^{238}U	241	1.33	99	1.8	3.6	1.5	This work
^{63}Cu + ^{197}Au	445	1.34	178	10.3	4.5	1.0	13

ergy distribution of the fragments is the same for all isotopes of a given element. Moreover, we have taken the energy spectrum for the most neutron deficient isotope to be representative of the primary energy distributions. Indeed, for this isotope both n, p, and α binding energy plus Coulomb energy (for the charged particles) are large enough to ensure essentially only γ decay. On the other hand, and for the same reasons, the feeding of this species by evaporation from the slightly heavier isotopes should also be very weak.

These primary energy spectra were converted into excitation energy distributions, and the neutron emission was then taken into account in a very simple way. An energy threshold for the emission of 1, 2, or 3 neutrons has been defined, equal to the binding energy for 1, 2, or 3 neutrons (taken from Ref. 23). We did not introduce any competition: As the threshold is reached for the emission of the ith neutron, the probability for the evaporation of this ith neutron becomes equal to unity. The cross sections of the daughter isotopes were then integrated accordingly and summed over all the possible feedings. It is only after having applied these corrections that comparison with the experimental data was meaningful.

IV. DISCUSSION

The systems for which the experimental isotopic distributions have been compared with the model are listed in Table I together with the entrance-channel parameters and the values of the transport coefficients used in the calculation. As mentioned before, these model calculations give the production probability for each nuclide. As an example, in Fig. 4 we show a contour diagram for the calculated production of the different nuclides as a function of A and Z; this calculation appears to be in good agreement with the experimental distribution.

A closer view of any such comparison between the calculation and the experiment can be obtained from the isotopic distributions for each element, or from the charge distribution for a given mass. The first is more revealing as the isotopic distribution widths are broader.

These experimental isotopic distributions (dots), compared with primary (solid lines) and secondary (histograms) calculated distributions are presented in Figs. 6–10 for all the systems listed in Table I. At first glance, an excellent agreement between experiment and calculation is generally observed provided that the secondary neutron emission is taken into account. The first and second moments of the distribution are generally very well reproduced except for two cases. First for S + U if good agreement is attained for the lower Z's ($Z \leq 12$) then rather large discrepancies result for $Z \geq 13$. We propose a rather simple explanation. The detection angle was rather close to the grazing angle, and it was very difficult to make a clear distinction between the quasielastic and the deep inelastic components. In this case, the reaction time is probably too short to allow for the N/Z degree of freedom to be equilibrated. For the Ar + Th system,[14] the width of the experimental distribution is substantially broader than that from the calculation For this case the rather high excitation energies may allow significant evaporation of charged particles, which have been neglected in our model.

Before looking further at the results, it is useful to discuss the role of the different parameters of the model. For this purpose four parameters have been successively varied in the calculation of the isotopic distribution for $Z = 16$ in the reaction Ar + U. Results are plotted in Fig. 11 for three such variations; the drift velocity V_A, the $D_A t_R$ product, and the parameter d by which the deformation of the fragment is simulated (the values of V_A, D_R, and t are those indicated in Table I). The fourth parameter, the mean orbital angular momentum in the entrance channel, has been varied between the estimated values of l_{crit} for fusion[17] and the l_{max} value corresponding to a grazing collision. Within these two limits the first moment of the isotopic distribution for $Z = 16$ does not change appreciably and the full width at half maximum

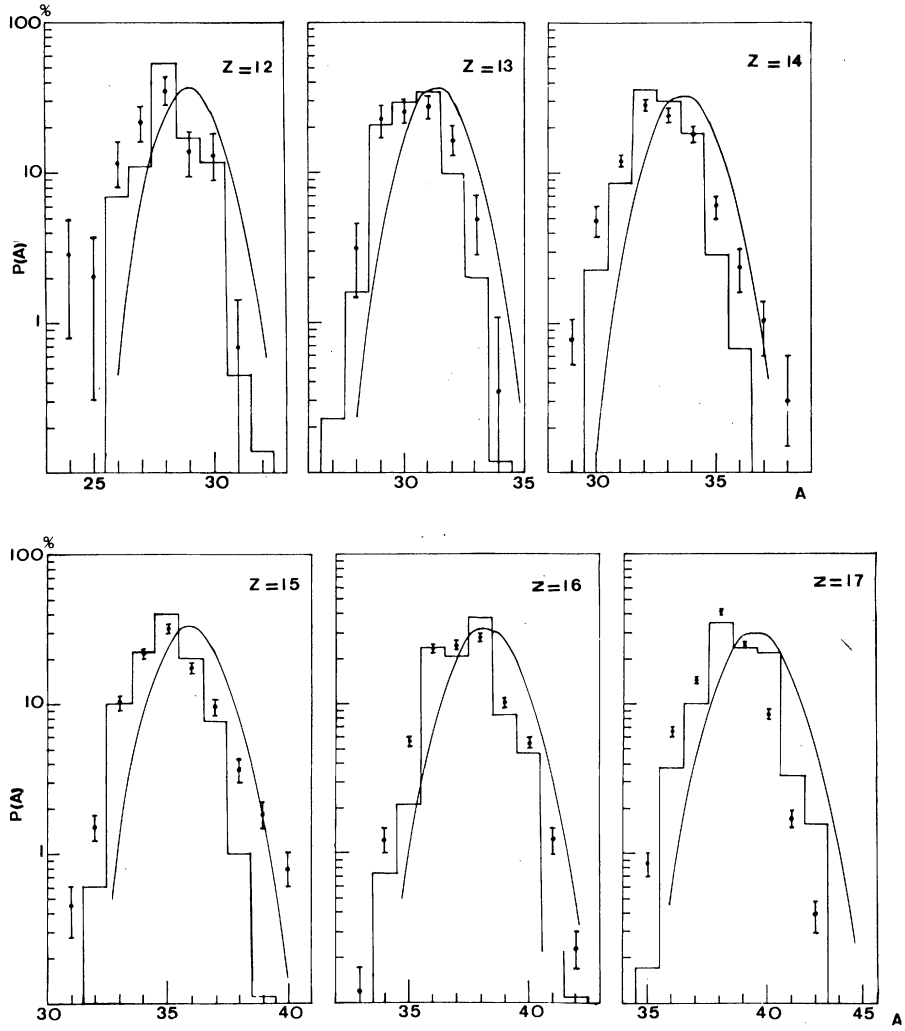

FIG. 6. Isotopic distributions for the reaction ^{40}Ar (265 MeV)+^{238}U. The experimental results (dots) are compared with the primary (solid lines) and secondary (histograms) distributions.

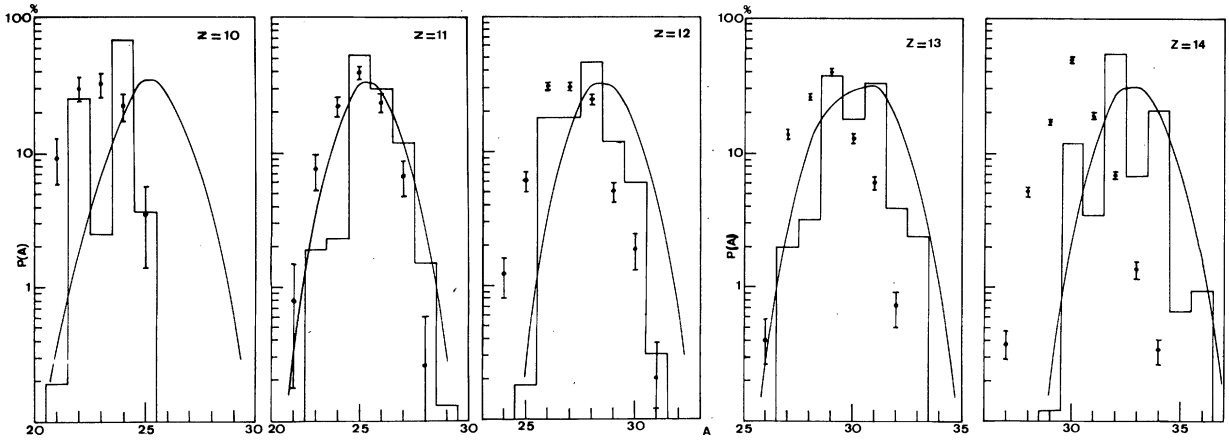

FIG. 7. Isotopic distributions for the reaction ^{32}S (241 MeV)+^{238}U. For definitions, see Fig. 6.

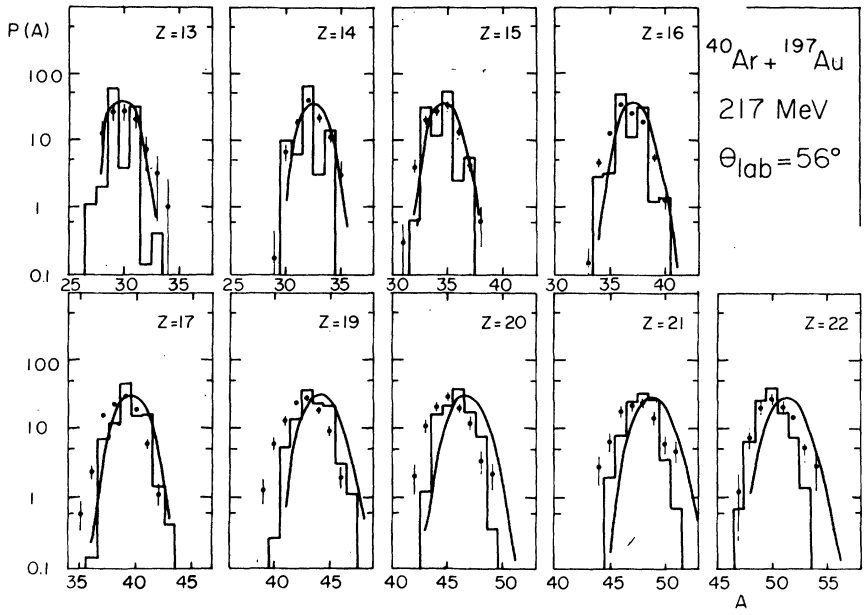

FIG. 8. Isotopic distributions for the reaction ^{40}Ar (217 MeV)+^{197}Au. For definitions, see Fig. 6.

(FWHM) varies by less than 0.2 u. From Figs. 11(a) and 11(b), we see that the transport coefficients are not critical for the isotopic distributions for neighboring masses even though they have been shown to be of great importance for describing the mass distributions. Indeed, even with a change of a factor of 3 in $D_A t$ or V_A, the calculated isotopic distributions remain unchanged. By contrast the distribution obtained for the extreme case of full equilibration [Fig. 11(b)] in the mass asymmetry degree of freedom gives a shift of two u. This shows clearly that in this model the mass equilibration is far from being achieved.

Although the parameter d was obtained by a fit to experimental energy data, it is nevertheless interesting to check its influence on the isotopic yields. In Fig. 11(c), it is shown that a variation in d of 25% (i.e., 1 fm) leads to only a small shift in A (less than half a mass unit) and essentially no change in the width.

In sum, for this model the first and second moments of the isotopic distributions are essentially parameter independent within a reasonable physical range.

The results presented in Figs. 6–10 call for three main remarks. The first concerns the influence of the evaporation on the second moment of the distribution. For all the systems which have been studied and independent of the number of evaporated neutrons, the deexcitation process does not

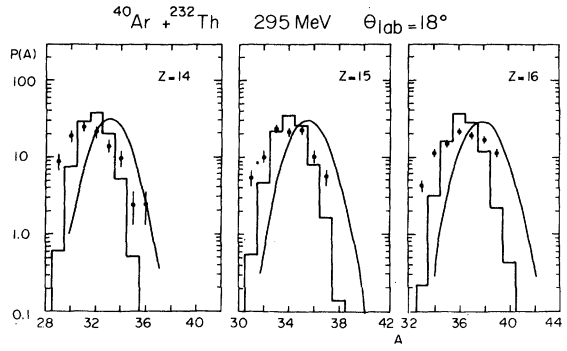

FIG. 9. Isotopic distributions for the reaction ^{40}Ar (295 MeV)+^{232}Th. For definitions, see Fig. 6.

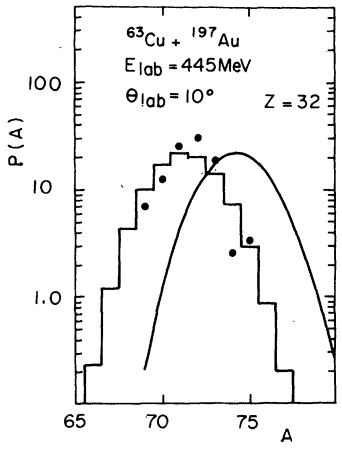

FIG. 10. Isotopic distributions for $Z=32$ in the reaction ^{63}Cu (445 MeV)+^{197}Au. For definitions, see Fig. 6.

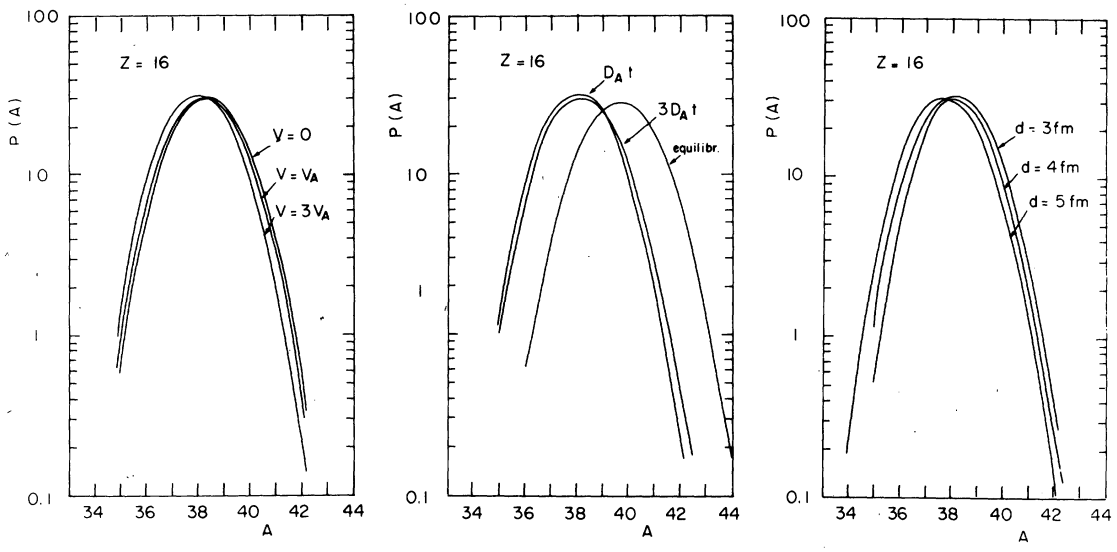

FIG. 11. Influence of the different parameters of the model ($D_A t$, V_A, d) on the isotopic distribution for $Z=16$ in the reaction ^{40}Ar (263 MeV) + ^{238}U ($V_A = 2.0\times10^{22}$ u sec^{-1}, $D_A t = 11$ u^2).

introduce any significant broadening of the isotopic distributions. This is the advantage of studying these reactions at low energy where only a few neutrons can be emitted. In this situation, the experimental width is that of the primary process as there is essentially no effect from the deexcitation process. In this sense the situation is very different for light systems such as ^{40}Ar + ^{58}Ni. Braun-Munzinger and Barrette[6] showed very clearly that for this system the broadening effect due to evaporation was more than a factor of 2. The second point is related to even-odd effects. The calculated primary distributions are, of course, very smooth as no pairing or shell effects have been taken into account in the calculation of either the potential energy or of the diffusion process. On the other hand, discontinuities may be observed in the distribution after the deexcitation process since binding energies for neutrons which have been used include both pairing and shell effects. These effects are emphasized when the mean number of evaporated neutrons is lower than unity.

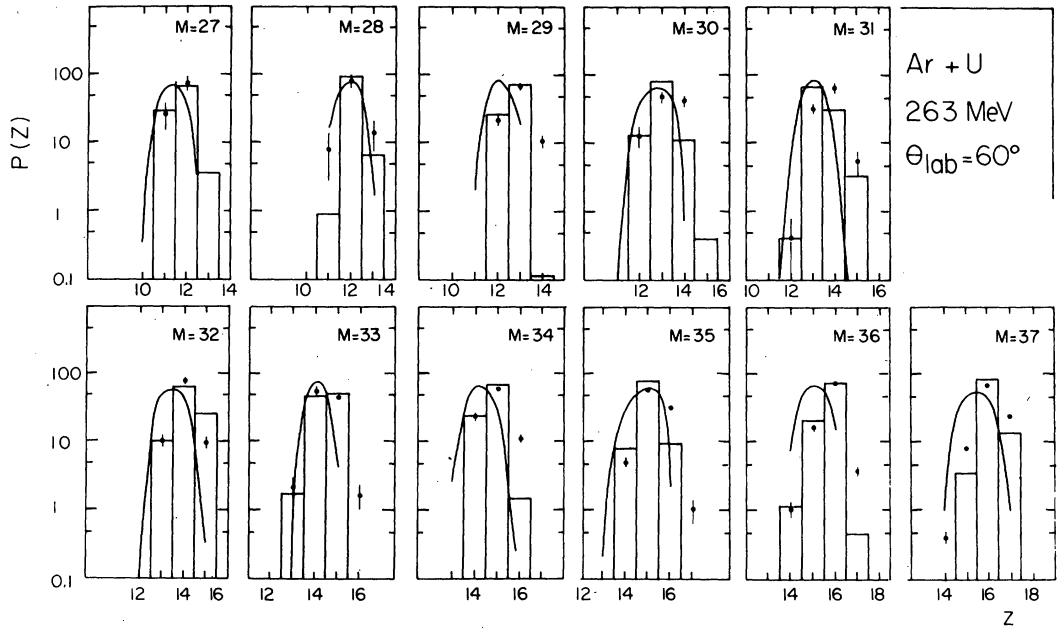

FIG. 12. Isobaric distributions for the reaction ^{40}Ar (265 MeV) + ^{238}U. For definitions, see Fig. 6.

(The example of the distributions for $Z = 13, 14$ in the Ar + Au reaction is very significant from this point of view.) Indeed, when the excitation energy is so low, the crude evaporation model used here fails as one should take into account the n-γ competition. On the contrary, these effects completely disappear when more than 3 neutrons are emitted (Fig. 10). Generally, the experimental distributions do not exhibit any large odd-even effects. Nevertheless, the weak effects observed for the Ar + U case are reasonably well reproduced by the calculation.

The last point refers to the production of new exotic nuclides in deeply inelastic reactions. Is the model sufficiently accurate to provide significant predictions for productions of such new n-rich isotopes? The answer is probably no. Experimental distributions are reasonably well described up to one order of magnitude below the maximum, but for the lowest part experimental widths are much larger than the calculated ones. Since we have used only a very crude model to try to account for the main features of the phenomenon, a bad description of the rate of production for the very rare events which can lead to exotic nuclei and which represent a few thousandths of the total production is not surprising. For example, the experimental production rate of four new isotopes produced in the Ar + U reaction[7] (^{37}Si, ^{40}P, $^{40\text{-}41}$S) is not at all well reproduced by this model calculation.

As mentioned before, the calculation describes also the charge distributions for a given mass asymmetry as shown in Fig. 12 for the system Ar + U. It is, of course, more difficult to make the comparison here as the widths of these distributions are very narrow, nevertheless the agreement is rather good.

V. CONCLUSION

Isotopic distribution measurements in deeply inelastic reactions are of great interest to test the hypothesis of equilibration for the N/Z degree of freedom. Moreover, the choice of very asymmetric systems in the entrance channel leads to very large exchanges of particles before the attainment of equilibrium for this degree of freedom (about 9 charge units according to the hydrodynamical model of Brosa and Krappe[9]). Thus, such systems give stringent tests of this hypothesis; the choice of a rather low energy above the interaction barrier minimizes the particle decay of the fragments.

Experimental results have been compared with a classical potential-energy calculation based on the liquid drop model coupled with a diffusion model and taking into account the subsequent decay processes. All the data which have been explored in this paper are in good agreement with this simple model (provided that only the deep inelastic component has been taken into account in the experimental data). The first and second moments of the isotopic distributions are well reproduced. On the other hand, and this is not surprising, the calculation does not predict very accurately the production of very n-rich nuclei. Their cross sections are often 2 or 3 orders of magnitude lower than that of the most probable isotope.

The good agreement we have always observed between the experimental widths and the calculated ones gives strong support for a statistical origin of the observed fluctuations. The very same conclusion was recently given by Wirth *et al.*[24] when studying the Xe + Au system. However, Berlanger *et al.*[25] have been led to interpret the fluctuations measured in the Kr + Mo system as quantal fluctuations rather than statistical ones. At this point, additional data are required to settle the question about the nature of the observed distributions.

In any case, all experimental results presented in this paper, and their comparison with the model calculations, give strong evidence that the neutron excess degree of freedom is achieved very readily in deeply inelastic reactions, independent of the number of charges exchanged between the two nuclei.

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