

# Charge Distributions According to the Cluster Model of Nuclear Fission

H. FAISSNER

III. Physikalisches Institut der Technischen Hochschule Aachen

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*Dedicated to Professor Dr. W. GENTNER on the occasion of his 60th birthday*

The cluster model of nuclear fission makes definite predictions about the charge distributions. The assumptions made in the computation of mass distributions have to be supplemented by the postulate of statistical sharing of the surplus protons and neutrons between light and heavy cluster. Given the mass distributions, the charge distributions are completely determined by statistical considerations. The calculated average charge  $\langle Z_p \rangle$  as a function of fragment mass number  $A_p$  for thermal neutron induced fission of  $U^{235}$  agrees rather well with the radiochemical results and some recent mass spectroscopic measurements. The shape of the charge distribution around  $\langle Z_p \rangle$  depends on  $A_p$ , and is in fair agreement with the available data.

The first measurements of the kinetic energy distribution of fission fragments were performed by FLAMMERSFELD, JENSEN and GENTNER<sup>1</sup>, and independently by JENTSCHKE and PRANKL<sup>2</sup>. They demonstrated already clearly the salient features of low energy nuclear fission: two broad regions of allowed masses, and a rapid decrease of yield towards symmetric or very asymmetric mass division. For a long time this striking mass asymmetry defied theoretical explanation. The liquid drop model<sup>3</sup>, so successful in explaining the energetics and the main features of the dynamics of fission, was unable to give any clue.

It was only after the success of the shell model<sup>4</sup> that a simple physical explanation seemed possible\*: The empirical data suggested a distinction of the magic numbers  $Z=50$  and  $N=82$  and  $50$ . Consequently it was argued, most convincingly by LISE MEITNER<sup>6</sup>, that asymmetric nuclear fission of heavy nucleides is an effect of nuclear shell structure. However, it was an open question in which way shell effects could influence nuclear fission, and at which stage of the process they would enter. Two models have been widely discussed: The statistical model<sup>7</sup> assumed that shell effects enter via the level density

of the final fragments. It is in violent disagreement with the experimental data, notably on the excitation energies of fragments, and should be discarded. Another approach is to consider shell effects in the energetics of the nascent fragments<sup>8,9</sup>.

A simple model of nuclear fission was recently proposed by FAISSNER and WILDERMUTH<sup>10,11</sup>. It is based on the cluster model of the nucleus<sup>12</sup>, which was rather successful in giving a physical explanation of level structures and reaction probabilities in light nuclei.

The idea is that large clusters (for instance with the above mentioned magic numbers) can also be formed within the fission prone compound nucleus during its migration through the many energetically possible nucleon configurations. Admittedly this is an improbable configuration — about as improbable as the concentration of the excitation energy on one neutron, which then can escape. But as soon as such a two-cluster-state is reached, the energy gained by cluster formation enhances the probability for fission considerably.

The cluster model provides a natural framework for a discussion of the many aspects of nuclear

<sup>1</sup> A. FLAMMERSFELD, P. JENSEN, and W. GENTNER, Z. Phys. **120**, 450 [1942].

<sup>2</sup> W. JENTSCHKE and F. PRANKL, Z. Phys. **119**, 696 [1942]. — W. JENTSCHKE, Z. Phys. **120**, 165 [1942].

<sup>3</sup> N. BOHR and J. A. WHEELER, Phys. Rev. **56**, 426 [1939]. — J. A. FRENKEL, J. Exp. Theor. Phys. USSR **9**, 641 [1939] and Phys. Rev. **55**, 987 [1939].

<sup>4</sup> M. GOEPPERT-MAYER, Phys. Rev. **75**, 1969 [1949]. — O. HAXEL, J. H. D. JENSEN, and H. E. SUESS, Phys. Rev. **75**, 1766 [1949].

\* Note however the early conjecture of FLÜGGE and von DROSTE<sup>5</sup>.

<sup>5</sup> S. FLÜGGE and G. VON DROSTE, Z. Physik. Chem. (B) **42**, 274 [1939].

<sup>6</sup> L. MEITNER, Nature London **165**, 561 [1950], and Ark. Fys. **4**, 383 [1952].

<sup>7</sup> P. FONG, Phys. Rev. **102**, 434 [1956]; Phys. Rev. Letters **11**, 375 [1963] attempts to save the model.

<sup>8</sup> W. BRUNNER and H. PAUL, Z. Naturforschg. **15 a**, 1018, and **1109** [1960]; Ann. Phys. Leipzig **6**, 267 [1960].

<sup>9</sup> R. VANDENBOSCH, Nucl. Phys. **46**, 129 [1963].

<sup>10</sup> H. FAISSNER and K. WILDERMUTH, Phys. Letters **2**, 212 [1962].

<sup>11</sup> H. FAISSNER and K. WILDERMUTH, Nucl. Phys. **58**, 177 [1964].

<sup>12</sup> K. WILDERMUTH and TH. KANELLOPOULOS, Nucl. Phys. **7**, 150 [1958]. — K. WILDERMUTH, CERN-Report 59-23 [1959].



fission from one unified point of view. The connection between mass asymmetry and angular anisotropy has been dealt with in an earlier note<sup>10</sup>. Asymmetric mass distributions were treated in some detail in a paper<sup>11</sup> (henceforth referred to as *I*). It contains also a discussion of the physical foundation of the model.

It is the purpose of the present paper to give the gross features of the charge distributions. As it will turn out, the prescriptions used in *I* to calculate the mass distributions fix the charge distributions too. One has only to add the very natural postulate that the sharing of surplus protons and neutrons between heavy and light cluster happens in a purely statistical way. This is discussed in Sec. 1. In order to obtain a true picture of the charge distribution one has also to consider cluster states with a small number of nucleons removed from the original "closed shells". Their relative contributions can be read off directly from the wings of the mass distributions (Sec. 2). Actual computations have been done for thermal neutron induced fission of  $U^{235}$ , where the most accurate experimental data are available. The results are given in Sec. 3. They agree quite well with the radiochemical data and also with some recent mass spectroscopic measurements.

### 1. Statistical distribution of surplus protons

The proposed way of determining charge distributions is based on one observation: that the sharing of surplus nucleons between heavy and light cluster has a vanishingly small influence on transition probabilities. This feature of nuclear fission has been discussed in paper *I*, in connection with the mass distributions. It manifests itself in the flatness of the mass distributions inside the allowed mass bands. This is, in a large number of cases, experimentally verified with great accuracy.

In the computation of mass distributions it was not necessary to specify, if a given surplus nucleon was a proton or a neutron. But in the spirit of the model this cannot influence transition probabilities in any significant way. Consequently, the charge distribution at a given fragment mass is completely determined by simple statistical considerations: One has only to compute the probability to obtain a

certain number of protons, if one allots to the original cluster a fixed number of the available surplus nucleons.

Let the light cluster have  $Z_1$  protons and  $A_1$  nucleons, the heavy cluster correspondingly  $Z_h$  and  $A_h$ . If the fissioning nuclide is characterized by  $Z_0$  and  $A_0$ , the number of surplus nucleons is

$$a = A_0 - A_1 - A_h, \quad (1a)$$

and the number of surplus protons is

$$z = Z_0 - Z_1 - Z_h. \quad (1z)$$

Consider a nuclide in the heavy group with mass number  $A_p$ :

$$A_p = A_h + n, \quad 0 \leq n \leq a. \quad (2A)$$

This mass number obtains by adding a fixed number  $n$  of surplus nucleons to the cluster mass number  $A_h$ . Amongst these  $n$  nucleons there may be  $k$  protons, where  $k$  runs from zero to the smaller of the two numbers  $n$  and  $z$ . Consequently, at this mass number  $A_p = A_h + n$  there is a spectrum of charges

$$Z = Z_h + k, \quad 0 \leq k \leq \min(n, z). \quad (2Z)$$

The statistical probability of getting  $k$  protons out of  $n$  nucleons, drawn at random from a sample of  $z$  protons and  $(a - z)$  neutrons, is

$$W_{nk} = \binom{n}{k} \frac{z}{a} \frac{z-1}{a-1} \cdots \frac{z-k+1}{a-k+1} \cdot \frac{(a-z)}{a} \frac{(a-z-1)}{a-1} \cdots \frac{a-z-(n-k)+1}{a-(n-k)+1}. \quad (3)$$

[For  $k=0$  the  $k$  factors of the type  $(z-\nu)/(a-\nu)$  have to be replaced by 1. The same applies to the  $n-k$  factors of the type  $(a-z-\mu)/(a-\mu)$  in case  $n-k=0$ .]

For a given cluster configuration, characterized by the four numbers  $(A_h, Z_h, A_1, Z_1)$ , the modified BERNOULLI distribution (3) is already the answer to our problem. The factor  $\binom{n}{k}$  is responsible for the sharp maximum of the charge distribution near the average charge  $\langle Z_p \rangle$ . This (in general nonintegral) expectation value is simply given by

$$\langle Z_p \rangle = Z_h + \frac{z}{a} n. \quad (4)$$

To the extent that the charge distribution is symmetric it agrees with the most probable charge  $Z_p$ , as defined in radiochemical studies<sup>13-15</sup>.

<sup>13</sup> A. C. WAHL, R. L. FERGUSON, D. R. NETHAWAY, D. E. TROUTNER, and K. WOLFSBERG, Phys. Rev. **126**, 1112 [1962].

<sup>14</sup> A. C. WAHL, Physics and Chemistry of Fission, Vol. I, p. 317, Int. Atomic Energy Agency, Vienna 1965.

<sup>15</sup> G. HERRMANN, Radiochim. Acta **4**, 173 [1965].

## 2. Determination of the cluster states involved

If there were only one heavy cluster and one light cluster involved in nuclear fission, the calculation was finished. But there is a complication discussed in detail in *I*: Indeed the formation of the heavy fragment seems always to be dominated by one cluster configuration, namely  $Z_h = 50$  together with  $N_h = 82$ . But to the formation of the light fragment contribute in general several cluster configurations. In case of  $U^{235}$  ( $n_{th}$ , fi) they are  $A_l = 90$  (with  $Z_l = 36$ ) and  $A_l = 84$  ( $Z_l = 34$  plus  $N_l = 50$ ). The relative contribution of these different cluster states has been taken from experiment (see Fig. 7 of *I*). Since fission via different cluster configurations does correspond to different fission modes, one has to compute the charge distributions for  $A_l = 90$  and  $A_l = 84$  separately, and adds them, properly weighted, together.

Another complication enters in the present context, which was not present in *I*: Asymmetric fission does also occur, though with reduced probability, when the above mentioned clusters are broken. In computing the mass distributions this was taken into account phenomenologically by adjoining GAUSS-functions to both edges of the allowed mass bands. We have to recognize that these "broken cluster states" have an appreciable influence on the charge distribution also inside the allowed mass bands. This follows immediately from the independence of transition probabilities from the distribution of surplus nucleons discussed in the preceding section.

Let us assume that  $r$  nucleons have been removed from the heavy cluster with original  $A_h$  nucleons. This bare broken cluster still leads with a finite probability to asymmetric fission, as evidenced by the finite yield of the fragment with mass number  $A_h - r$ . But the addition of any number of the now available surplus nucleons, according to our model, does not change the transition probability. The same argument applies for the light cluster.

For the quantitative formulation we consider only one unbroken light cluster (say  $A_l = 90$ ). Let us denote the probability that a heavy cluster with  $r$  missing nucleons is formed, and leads to fission, with  $p_r$ . The corresponding probability for the light cluster with  $s$  missing nucleons be  $q_s$ . The fractional fission yield involving these two configurations is

$$y_{rs} = p_r q_s. \quad (5)$$

It is uniform for a mass band extending from  $A_h - r$  to  $A_h + a + s$  in the heavy fragment group, (and of course from  $A_l - s$  to  $A_l + a + r$  in the light group). The total mass yield at a given  $A$  is given by the sum over all possible contributions (5):

$$y(A) = \sum_{r,s} p_r q_s. \quad (6)$$

Inside the flat top of the mass peaks (extending from  $A_h$  to  $A_h + a$  for the heavy, and from  $A_l$  to  $A_l + a$  for the light group), the summation extends from  $r, s = 0$  over all configurations considered. For a heavy fragment mass  $A_h - R$  the summation starts with  $r = R$ , for a light fragments mass  $A_l - S$  with  $s = S$ . The nearly trapezoidal mass distributions presented in *I* are, according to the present viewpoint, composed of a large number of rectangles of different length. The mass yields  $y(A)$  have already been computed in *I*, under the reasonable, though approximate, assumption  $p_r \sim q_r$ . From there we get the relevant probabilities  $p_r$ , as listed in Table 1. (Slightly deviating from *I* we used the constant yields  $y_0(A_l = 90) = 4.53\%$  and  $y_0(A_l = 84) = 1.0\%$  throughout the flat top as defined above.)

Finally we have to specify the charge of the broken cluster configurations. It does depend on the nature of the cluster, if protons and neutrons can be removed with comparable probabilities: The cluster with  $A_l = 84$  is associated with  $N = 50$ , whereas  $Z = 34$  is no magic number. Consequently, in breaking this structure up, protons are more likely to be removed. None such preference exists in  $A_l = 90$ . In  $A_h = 132$  both, proton and neutron number are magic. However, as has been outlined in *I*, the abnormally low  $Z_h/A_h$  of 0.3788, ( $Z_0/A_0 = 0.3898$ ), is energetically unfavourable, because of the symmetry force. Therefore, it is much more favourable to remove neutrons than protons.

These considerations have been rationalized in the following way:  $A_h = 132$  loses only neutrons,  $A_l = 84$  loses only protons, and  $A_l = 90$  loses both of them with equal probabilities. Since only the first four broken cluster configurations need to be taken into account (because of the decrease in yield), these assumption are presumably well justified. (Of course they become wrong if pushed too far.) The numbers used in the computations are given in Table 1. They fix the charge distribution of  $U^{235}$  ( $n_{th}$ , fi) completely.

$A_h = 132$				$A_1 = 90$				$A_1 = 84$				alternative $A_1 = 84$			
$r$	$A_r$	$Z_r$	$p_r$ %	$s$	$A_s$	$Z_s$	$q_s$ %	$s$	$A_s$	$Z_s$	$q_s$ %	$s$	$A_s$	$Z_s$	$q_s$ %
0	132	50	10.24	0	90	36	8.40	0	84	34	1.84	0	84	34	1.84
1	131	50	4.78	1	89	36	1.96	1	83	33	0.86	1	83	34	0.43
2	130	50	3.69	1	89	35	1.96	2	82	32	0.67	1	83	33	0.43
3	129	50	2.44	2	88	35	3.02	3	81	31	0.44	2	82	33	0.67
4	128	50	2.39	3	87	35	1.00	4	80	30	0.43	3	81	33	0.22
				3	87	34	1.00					3	81	32	0.22
				4	86	34	1.96					4	80	32	0.43

Table 1. Masses, charges and relative contributions of the "cluster" configurations used in the computation. — Differential yield of a particular cluster combination  $y_{rs} = p_r q_s$ .

### 3. Results and discussion

Some examples of computed charge distributions for fixed fragment mass number  $A_p$  are given in Fig. 1. As usual the yields are given as fractions of the chain yield. The calculated independent yields are indicated by the uninterrupted curve, the cumulative yield by the dashed curve. Experimental independent yields are given as open circles, measured cumulated yields as filled circles. Of course these radiochemical data were obtained after neutron emission, whereas the computation refers to primary masses. For the examples chosen the correction is easy, because for  $A_p = 92$  and  $94$  the number of emitted neutrons is about one, and this is also approximately true for the two other masses<sup>16</sup>.

The agreement with the radiochemical data of WAHL and coworkers<sup>13, 14</sup> is not too bad. It is actually quite good in the regions of high yield. The only definite discrepancy is that the computed distributions have broader wings, in particular towards larger charges for light fragments (i.e. smaller charges for the heavy fragments). It is not quite clear what this discrepancy means. The main contribution to the yield comes from the combination of different states of the  $A_1 = 90$ -cluster with the states of  $A_h = 132$ . For  $A_1 = 90$  the mass yields are well known, and the cluster states are essentially unambiguous (see Table 1). There is no parameter to adjust. If one assumes that the  $A_1 = 84$  cluster is broken in a different way, namely by removing both, protons and neutrons, instead of protons alone, the discrepancy is slightly enhanced. Charge distributions wider than obtained by radiochemical methods have recently been found by KONECNY et al.<sup>17</sup>,

using a high resolution mass spectrometer. It seems best to wait with a final judgement until the experiments form a consistent pattern.

The average charge value  $\langle Z_p \rangle$  is only little affected by this discrepancy in the far wings. Consequently the computed  $\langle Z_p \rangle$  values agree rather well with the empirical  $Z_p$ -function, as derived by WAHL et al.<sup>13</sup> on the basis of radiochemical data. The comparison is done in Fig. 2. As usual we plot  $Z_p$  minus  $Z_p^0$ , the charge which would result, if all fragments had the same charge density as the parent nucleus:

$$Z_p^0/A_p = Z_0/A_0.$$

The results of KONECNY et al.<sup>17</sup> are also given; (filled circles are averages over two adjacent points). The well established parts of the theoretical curve are given by an uninterrupted line. They result from considering only the  $A_1 = 90$ -configurations. Point-dashed portions were obtained by smooth interpolation between high-yield asymmetric fission and the extreme cases, symmetric and very asymmetric fission, which have to give an "unchanged charge distribution"  $\langle Z_p \rangle = Z_p^0$ . A possible influence of the  $A_1 = 84$ -configuration is indicated by the dashed curves. The upper one corresponds to the  $N = 50$ -shell remaining intact, the lower one to the case of having protons and neutrons removed with the same probability. Which of these possibilities is favoured by nature cannot be decided on the basis of present experiments. Some radiochemical measurements seemed to indicate an increase of  $|\langle Z_p \rangle|$  after the well established dip around  $A = 148$ . For this reason WAHL's curve bends over again. But the experimental errors are too large for any definite conclusion. On the other hand, in the region of high mass yield the

<sup>16</sup> J. TERRELL, Phys. Rev. **127**, 880 [1962].

<sup>17</sup> E. KONECNY, H. OPOWER, H. GUNTHER, and H. GÖBEL, Physics and Chemistry of Fission, Vol. I, p. 401, Int. Atomic Energy

Agency, Vienna 1965. Points from Diplomarbeit GÖBEL (unpublished), cited in Ref. <sup>19</sup>.



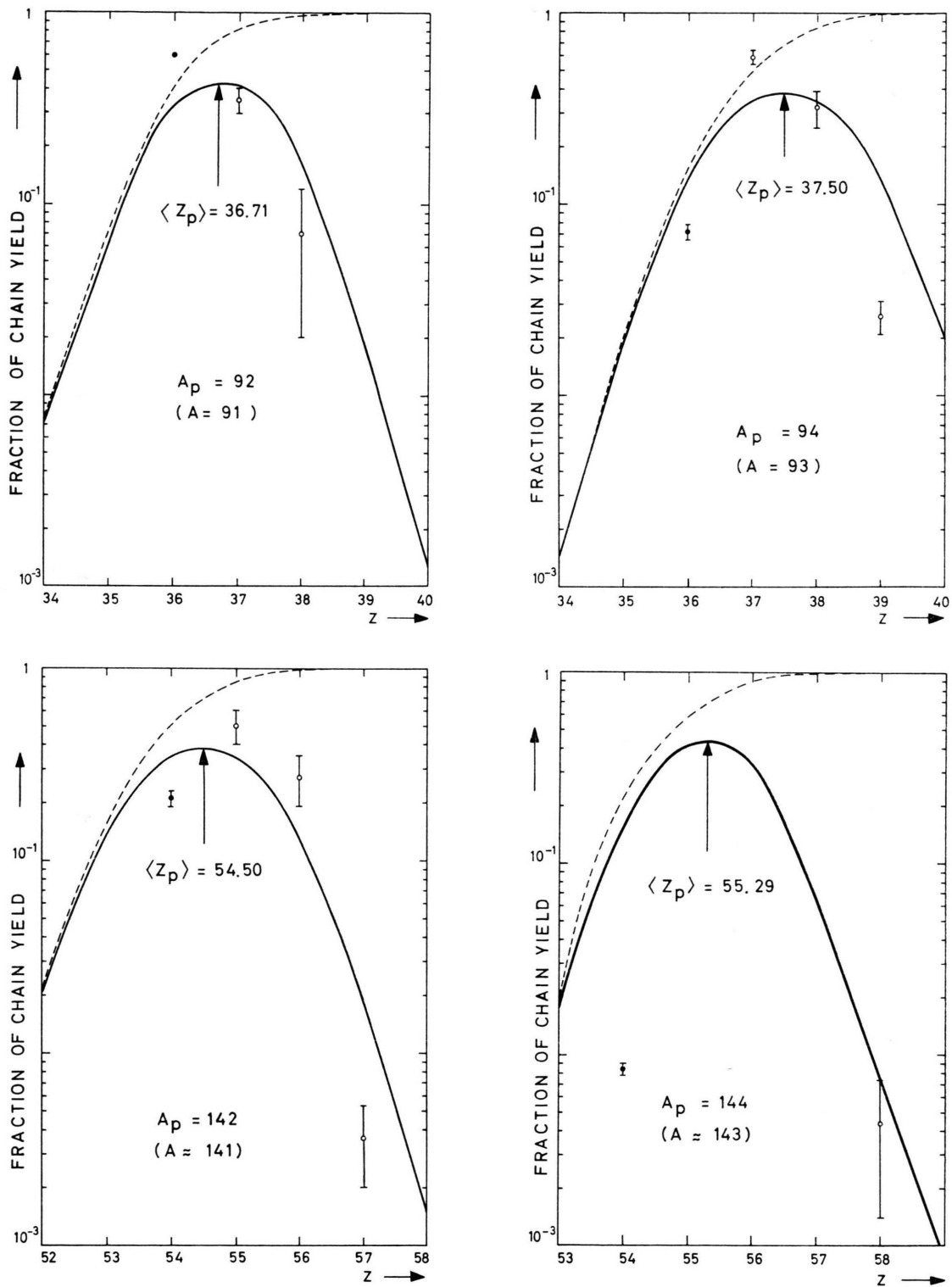


Fig. 1. Calculated fractional yield of fragments with primary mass number  $A_p$  as a function of  $Z$ : ——— independent, — — — cumulative yield. Experimental points from WAHL<sup>13, 14</sup> (○ independent, ● cumulative yield).

agreement between prediction and experiment is very satisfactory. (This does not apply to the earlier low mass resolution measurements of ARMBRUSTER et al.<sup>18</sup>.)

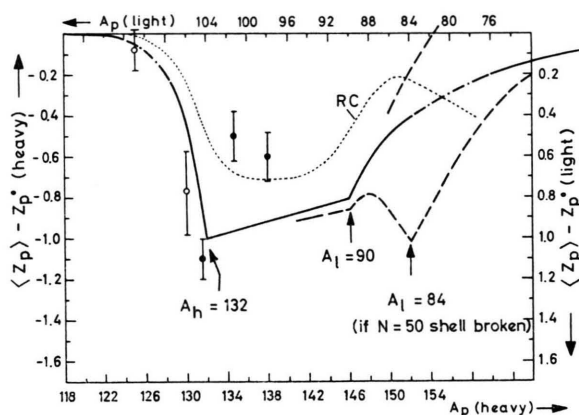


Fig. 2. Average charge as a function of primary mass number  $A_p$ : ——— computed from  $A_l=90$  alone (— · — interpolated). — — — possible influence of  $A_l=84$ . ····· Radiochemical data according to WAHL et al.<sup>13</sup>. ○ ● mass spectroscopic measurements of KONECNY et al.<sup>17</sup>.

Previous attempts to understand the empirical  $Z_p$  values have not been very successful. Neither the empirical rule of “equal charge displacement”, nor

the postulate of “minimum potential energy”, nor the statistical model give an adequate description of the data. Surveys of these models may be found in Refs. 13 and 18. A quite ambitious calculation was recently done by NÖRENBERG<sup>19</sup>. It is based on the COOPER-BARDEEN-SCHRIEFFER model, and yields a narrow, symmetric peak around  $A_h=132$ . With respect to peak height and descend towards symmetric fission this calculation agrees with ours. It fails however to give the flat portion of the curve between heavy fragment masses of 132 and 146. This flat top corresponds precisely to the flat top in the mass distribution. In our model it has the same physical origin: The gradual addition of surplus nucleons to a cluster structure. Since this appears to be the basic process in all low energy fission reactions, all these charge distributions are expected to show a similar behaviour. Also this is indicated by the available data<sup>13, 14</sup>.

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<sup>18</sup> P. ARMBRUSTER, D. HOVESTADT, H. MEISTER, and H. J. SPECHT, Nucl. Phys. **54**, 586 [1964].

<sup>19</sup> W. NÖRENBERG, Phys. Letters **19**, 589 [1965].