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LAWRENCE RADIATION LABORATORY
UNIVERSITY of CALIFORNIA BERKELEY

31

DISCLAIMER

SOME LIMITATIONS ON THE PRODUCTION OF VERY NEUTRON-DEFICIENT NUCLEI[†]

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Abstract

Our present method of producing very-neutron-deficient nuclei--evaporation of neutrons following compound nucleus formation--is limited by the eventual predominance of protons over neutrons in the evaporation spectrum. We have made measurements to determine the effective Coulomb barrier for protons in the evaporation process, and have then explored mathematically the nature and location of this limit.

[†] Work performed under the auspices of the U. S. Atomic Energy Commission.

1. Introduction

Heavy-ion reactions have been used for a number of years as a means of producing and studying neutron-deficient nuclei. The fusion of a target and projectile nucleus inevitably tends to produce a neutron-deficient compound system due to the increasing neutron excess of heavier nuclei along the valley of beta stability. For this reason, very heavy projectile nuclei will generally produce the most-neutron-deficient products. Also, these compound systems are produced with considerable excitation energy, which usually results in the evaporation of a few particles. For heavier nuclei, the Coulomb barrier inhibits the evaporation of charged particles resulting (normally) in a strong preference for evaporating neutrons. This tends to produce still more-neutron-deficient products. As the compound system becomes very neutron-deficient, however, proton emission increasingly competes with, and eventually dominates over, neutron emission, resulting in less neutron-deficient products1). This competition constitutes a limit on our ability to produce very neutron-deficient products using this method, and our aim here is to find the nature and location of this limit in the region of the periodic table between tin and lead. Above Pb, fission begins to compete with particle evaporation from the compound system, and this changes the situation rather completely. It should also be noted that we are only considering compound nuclear reactions; surface reactions are being neglected in the calculations.

2. General Method

In order to find the limit in producing neutron-deficient product nuclei, we must evaluate two things. These are (1) which compound system can be produced and (2) what particles will be evaporated from this system. The first of these is easy to evaluate under any given conditions for a particular projectile-target combination. Since accelerators that can accelerate any nucleus will soon become available, we have chosen, as a reasonable condition, only that both target and projectile must exist in nature. This is not an ultimate limit, by any means, but seems likely to be the best achievable in the near future. One can then easily determine the lightest compound nucleus (LCN) than can be produced for each element. In fig. 11, which will be discussed later, this limit is shown as the heavy dashed line. We have actually constructed this line from only even-Z nuclei, but, since protons are readily evaporated in this region, it does not effectively differ for the odd-Z nuclei.

The question of which particles are evaporated from a compound nucleus clearly depends on the relative effective binding energies. For neutrons, these effective binding energies do not differ from the usual binding energy, but for protons they must include an effective Coulomb barrier. We believe this effective barrier can be best determined empirically from the relative proton-neutron evaporation rates near the point where they are equal. We have used a very simple relationship[2])

$$P_n/P_p = e^{-\left(\frac{B_n-B_p^*}{T}\right)} , \qquad (1)$$

where: P_n and P_p are the probabilities of evaporating a neutron and proton, respectively; B_n is the neutron binding energy; B_p^* is the effective proton binding energy; and T is the nuclear temperature. One can see that B_p^* can be determined from P_n/P_p since B_n is available from mass tables[3]) and T is not very important if $P_n/P_p \sim 1$. We then obtain the effective Coulomb barrier, E_c^*, from

$$B_p^* = B_p + E_c^* \quad , \tag{2}$$

where B_p is the proton binding energy from the mass tables. With the assumption that

$$E_c^* = \frac{kZ}{1 + A^{1/3}} \text{ MeV} \quad , \tag{3}$$

where k is a number (presumably near one) that is evaluated from the measured E_c^*, we can scale E_c^* to other elements. This method has the enormous advantage that errors in B_n, B_p, or eq. (3) are normalized out at the point where P_n/P_p is measured, and only relative errors from this point are important. For the region we are interested in, around $P_n/P_p \sim 1$, the value chosen for T is not very important, but it is critical for calculating P_n/P_p far from this region. We have used T = 1.5 MeV.

In principle, alpha emission relative to neutron or proton emission should be treated in just the way described above. However, we have found (1) that alpha evaporation is considerably smaller than proton evaporation over the region of nuclei we are interested in and (2) that around the region where $P_n/P_p \sim 1$ the ratio P_p/P_α does not change very much. For the region of nuclei around Ce, we have therefore simply used an empirical value of 3 for P_p/P_α, and in the Os region we have used 2 for this ratio. If one were mainly interested in the products from alpha evaporation, this would not be a very good procedure, but we are more concerned with the main products which come from proton and neutron evaporation.

Using the above-outlined procedure we can, in a given case, find the lightest compound nucleus that can be produced, and calculate the probabilities for the emission of protons, neutrons and alpha particles. Each of the product nuclei can again emit particles with calculable probabilities, and so on, until the excitation energy remaining becomes too small to permit the evaporation of further particles. If this is done for a number of cases, using different excitation energies, we can find the maximum probability with which a given product can be made. We need now to discuss the relationship between excitation energy of the compound system and the number of particles evaporated. We have adopted the point of view that one can use an appropriate bombarding energy so that any preselected number of particles will be evaporated. This means that we consider, for example, the evaporation of four particles from a compound system, and leave it as a separate problem to determine at what bombarding energy this occurs for a given target-projectile pair. Alpha evaporation requires about twice the energy needed for proton or neutron evaporation, so that we just consider an alpha particle to replace two nucleons. This is not very important as we are not primarily interested in the products of the alpha evaporations, but only in deciding for which steps alpha evaporation can compete with nucleon evaporation.

It is, of course, true that even at the optimum bombarding energy for evaporating, say, four nucleons, three or five particles may be evaporated. For light projectiles the latter cross sections are relatively small (~ 10%), but they can be around a factor of two larger for presently-available heavy ones[4]) (due to angular momentum effects). They also become larger the greater the number of nucleons evaporated. This correction has not been included in our calculations, nor do we allow for the fraction of the total reaction cross section going into surface reactions (20-40%). Thus an absolute accuracy of a factor of two is the best we can expect, although relative cross sections from a given compound nucleus might be somewhat better.

One further point concerning excitation energies needs to be mentioned. This is the minimum number of nucleons that can be evaporated in a given case. Since, as discussed below, the maximum yield of a given product usually results from evaporation of the fewest possible number of nucleons (from the appropriate compound nucleus) it is important to know what this number is. It depends on the excitation energy of the compound nucleus, which is determined by the Q value for the reaction and the bombarding energy. The minimum bombarding energy possible is just the Coulomb barrier energy for the projectile-target system, so that the minimum excitation energy, $E^*_{CN}(\min)$, can be estimated as:

$$E^*_{CN}(\min) \approx Q + \frac{Z_1 Z_2}{\Sigma A^{1/3}} \text{ MeV} \quad . \qquad (4)$$

The quantity $E^*_{CN}(\min)$ has been plotted in fig. 1 for several target projectile systems that produce light Ce compound nuclei. The usefulness of this can be seen if one uses the rough estimate that evaporation of a nucleon requires about 15 MeV. Thus production of ^{128}Ce, for example, by the ^{16}O + ^{112}Sn reaction at the minimum bombarding energy should result in good yields for the evaporation of only two nucleons; whereas production by ^{20}Ne + ^{108}Cd or by ^{32}S + ^{96}Mo should result mainly in three or more evaporated nucleons. The lower E_{CN}^{*}(min) for the lighter Ce nuclei, using a given target-projectile pair, results from the greater instability of these Ce nuclei and hence a lower Q value. This is a very fortunate circumstance, as it means that evaporating only two nucleons may often be possible for the very lightest systems, where evaporation of a third particle might cost a factor of 5 or 10 in yield.

3. Experimental

The experimental problem is, then, to measure the relative yields of two or more reaction products in a case where protons and neutrons are both strongly evaporated, and from these ratios obtain values for k using eqs. (1-3) above. We chose first the Ba-Ce region for this measurement, as we were interested in studying the energy levels of very light even-even isotopes of these elements. We made a second determination in Os in order to check the validity of the extrapolation according to eq. (3). The primary data obtained were Ge(Li) gamma-ray spectra taken both during and between the 5 msec Hilac beam bursts. From these two spectra a net in-beam spectrum could be obtained by subtracting the appropriate fraction of the out-of-beam spectrum, and the out-of-beam spectrum could also be corrected for the portion of the time it was not gated on. The yield of an in-beam (prompt) transition could thus be directly compared with an out-of-beam (after one or more β-decay) transition. The spectra were sometimes rather complex, and to aid in identifying the various products coincidence measurements were made between a particle detector, sensitive to the evaporated protons and alpha particles, and the gamma-ray detector.

In fig. 2 are shown the net in-beam spectrum and the out-of-beam spectrum (not normalized) following bombardment of ^{112}Sn with 88 MeV ^{16}O ions. A number of lines have been identified, but there are also many others which have not been. We are mainly interested here in a set of lines$^{5)}$ belonging to the ground-state rotational band of ^{124}Ba. These occur in both the net in-beam and out-of-beam spectra and are indicated near the center of fig. 2. In the in-beam spectrum these must result from the prompt de-excitation of ^{124}Ba formed in the ^{112}Sn(^{16}O,2p2n)^{124}Ba reaction (an alpha particle rather than two protons and two neutrons is excluded, see below). In the out-of-beam spectrum these lines result from the β-decay of ^{124}La which is produced either by the ^{112}Sn(^{16}O,p3n)^{124}La reaction or from the β-decay of ^{124}Ce, the ^{112}Sn(^{16}O,4n)^{124}Ce product. To use the ^{124}Ba lines in the out-of-beam spectrum as a measure of the total yields of the p3n and 4n reactions, we must be sure that: (1) the β-decay lifetimes of ^{124}Ce and ^{124}La are short compared with the bombardment time; and (2) that all the β-decay of ^{124}La goes through the transition used. In this case, the bombardment time was ~ 1 hour and, although the lifetimes of ^{124}Ce and ^{124}La are not known, they are very likely to be short compared to this, as estimated from the known β-decay lifetimes in this region. Thus (1) above is likely to be fulfilled. Regarding (2), since the decay of ^{124}La leads to population of the 6^+ and 8^+ states in ^{124}Ba, it seems quite likely that essentially all the decays go through the $2^+ \to 0^+$ transition. Thus comparison of the yields of this transition in-beam to out-of-beam should give directly the relative yields of the 2p2n to p3n+4n reactions, respectively. This ratio is about 1.2 and determines rather sensitively the quantity k in eq. (3). Using the $4^+ \to 2^+$ transition rather than the $2^+ \to 0^+$ one makes essentially no difference in the result. The ^{124}Xe line[6]) in the out-of-beam spectrum results from any of the 4n, p3n, 2p2n, or 3pn reaction products. We have abbreviated this by 3xn where 3x can be three protons or three neutrons or a mixture of these. In this case, however, neither of the above two conditions is likely to be fulfilled, so that the yield is not useful. It does mark the exact location of the ^{124}Xe line in the in-beam spectrum, however, which enables an upper limit to be set on the 4p reaction, and this limit is consistent with the calculated yield, based on the same value of k.

In order to be sure that our assignments are correct, and that the outgoing particles are 2p and 2n rather than an α particle, we have made coincidence measurements with a thin particle detector in the backward direction. An Al foil, 0.025 mm thick, covered the detector and prevented any projectiles from reaching it. The proton and alpha particle spectrum resulting is shown in fig. 3. The proton spectrum is cut off at ~ 8 MeV, since the counter was thick enough for the deposition of only this much energy from protons. This helps considerably in separating the protons and alpha particles, and, as fig. 3 shows, this separation is quite adequate. Figure 4 contains the gamma-ray spectra in coincidence with the protons and alpha particles. The proton-coincident spectrum is rather clean and shows that the ^{124}Ba is, indeed, essentially completely produced by the 2p2n reaction. The spectrum in coincidence with alpha particles is more complex, and we have identified only the lines from ^{122}Ba, which have been characterized in other experiments$^{7)}$. In both of these spectra the results from several bombarding energies have been added together in order to improve the statistics, as is indicated in fig. 4.

In the Ba-Ce region we have ten reasonably good ratios of the type described above, all of which are consistent with a value for k (eq. (3)) of about 0.85. Although the variation in the ratios is no doubt due to experimental error and failure of the cases to meet the two conditions outlined above, we can, for orientation, ascribe it all to variations in k, and find then that $k = 0.85 \pm 0.05$. We have measured one ratio in Os as a further check on eq. (3). Figure 5 shows the net in-beam and out-of-beam spectra from 155 MeV Si ions on ^{144}Sm. Here the rotational lines$^{7)}$ of ^{168}W are very analogous to those of ^{124}Ba, described above, and their ratio in- and out-of-beam is in excellent agreement with the calculation based on the above value for k. We thus feel that one can calculate with reasonable confidence the proton-neutron relative evaporation rates over a broad region of nuclei.

4. Results

In this section we will try to indicate what the above considerations imply about producing very neutron-deficient nuclei with heavy ions. In fig. 6 the effective binding energies for protons and neutrons are plotted against the mass number for Os and Ce nuclei. These curves are based on the even-A nuclei only, so that the changes in pairing energy should be similar for removal of either a proton or a neutron. For the odd-A nuclei, we have interpolated from these curves rather than using the mass tables, since we feel that pairing effects will be essentially absent for very highly-excited nuclei. It is apparent that the neutron binding energies are much lower for the heavier nuclei, resulting predominantly in neutron evaporation (eq. (1)). However, as the neutron number decreases this situation changes, so that proton evaporation dominates for the lighter nuclei shown. Also indicated in fig. 6 is an effective alpha-particle binding energy, where the effective Coulomb barrier was taken to be the same fraction of the full Coulomb barrier as was found for protons. (Actually, pairing or other effects might shift this line somewhat relative to the proton and neutron lines.) One can see from this why: (1) alpha evaporation relative to proton evaporation does not change very rapidly with A, for a given Z, and (2) the proton to alpha-particle evaporation ratio changes only slightly between Ce and Os.

Using eq. (1) and the fixed ratio for P_p/P_α, the effective binding energies of fig. 6 can be converted into relative probabilities for the emission of neutrons, protons, and alpha particles. For Ce these are shown in fig. 7. Although these probabilities do not seem to change so rapidly, the cumulative changes after emitting several particles can be very large.

When plots like fig. 7 have been constructed for all the elements in a given region, one can follow the evaporation of particles from a particular compound nucleus. Choosing ^{128}Nd, for example, at an excitation energy such that four nucleons (or an alpha particle and two nucleons) are emitted we can calculate the yields Ba, La, Ce, Pr, and Nd nuclei with mass 124 (or Ba, La, Ce with mass 122). These yields, and all those discussed below, are per compound nucleus formed, and involve the approximations discussed in sec. 2. The curve for these yields as a function of Z is shown in fig. 8 as the one labeled 124 for A_{Final}. The largest yield (around 25%) is for ^{124}La and the yields fall off rather rapidly away from this value for Z (~ 6% for ^{124}Ba and less than 0.01% for ^{124}Nd). For this case the probability of emitting an alpha particle is about 15%, but since alphas can compete at each of three steps, almost half of the total yield involves evaporation of an alpha particle. Thus the α2x yields are rather large, as one can see qualitatively from the data in figs. 2, 4, and 5. Figure 8 also shows how the above pattern changes, with increasing mass number, into the more familiar situation where the HI,4n product dominates. Since alpha emission behaves like proton emission, it also becomes small at large mass numbers.

If we are interested, for example, in Ce final nuclei, then fig. 8 will give the relative yields expected, as a function of mass number, for producing Ce nuclei in 2p2n reactions. These peak at 30% near ^{128}Ce, and fall off rather slowly at higher and lower mass numbers. We can now try to compare these yields with those obtained by producing these same Ce nuclei using other reactions. Figure 9 shows the results of such a comparison, where the line labeled 2p2n is the one just described above. Since ^{128}Nd is the lightest compound nucleus (LCN) of Nd that can be made (see sec. 1), it follows that ^{124}Ce is the lightest possible 2p2n Ce product, and this is indicated in fig. 9 by termination of the 2p2n curve with a short perpendicular line. It should be realized that in fig. 9 we have not corrected for possible differences in surface-reaction cross section, or for the extent to which the entire compound-nucleus cross section goes into evaporation of a particular number of nucleons. We have tried to emphasize this by labeling the ordinate of fig. 9 as the maximum cross section that could be expected (σ_{max}) divided by the compound-nucleus cross section (σ_{CN}). Fortunately, our conclusions are not very sensitive to small shifts in these curves.

Three other features of fig. 9 require explanation. The first is that we have considered only the evaporation of an even number of nucleons. This was done for the sake of simplicity in the figure; and, in fact, the p2n and 2pn reactions give yield curves similar to those of the 2p2n reaction. Neither of these reactions can give Ce nuclei lighter than mass 124, however. The second omission from fig. 9 is the reactions where alpha particles are evaporated. In general, these are not so important because alpha emission moves along a line roughly parallel to the LCN line, and it is usually better to produce a lower-Z compound nucleus at a lower excitation energy than to evaporate an alpha particle. This is not always possible, however, so that there can be cases where these reactions should not be omitted. Finally, the dashed lines in fig. 9 for the 2p and the 2n reactions are meant to indicate that they may or may not have such high yields depending on whether the desired excitation energy is above or below E^{*}_{CN}(min)--(see sect. 1).

A number of conclusions may be drawn from fig. 9 (or similar figures for other elements). The first is that above mass 128 the HI,xn reactions are the best, and should give yields of 50% or higher. A second is that the HI,xp reactions never have large yields in this region because sufficiently-neutron-deficient compound nuclei cannot be made. If one wants to make a particular Ce nucleus, ^{126}Ce for example, then fig. 9 immediately shows that the best reactions to try are 2p, 2n, or 2p2n (p2n should also be in this group). In fact, the 2n reaction has recently been found to work very well in this case[7]). We further see that ^{124}Ce can only be produced in 2 or 3 times poorer yield--by 2n or 2p2n (or p2n) reactions. Such knowledge is extremely useful in trying to identify these products. Figure 9 also illustrates the important fact that the highest possible yields for producing the Ce nuclei begin to drop off rather sharply below mass ~ 126. Below mass 120, there is no significantly better way to produce the Ce nuclei than by the 4n reaction and the highest possible yield is less than 0.01%. This is already well below the yield required (~ 10%) for in-beam spectroscopic studies of the type currently being made, so that such studies can only be made down to about mass 124. However, some types of out-of-beam experiments, such as those using on-line isotope separators or the study of alpha or proton emitters, can work with much lower yields, so it seems useful to extend these calculations to lower yields.

At lower mass numbers the decrease in yield becomes much steeper as is indicated in fig. 10. Here we see that the HI,4n curve terminates at mass 118, since the LCN in Ce is at mass 122. To get lower masses, one must evaporate a larger number of neutrons. Effectively, then, the curve of highest yield for a Ce nucleus is the line connecting the termination points of the 4n, 5n, 6n, etc. curves, and that is just the dashed line in fig. 10. One can see that the yield is below 10^{-9} at mass 116, and dropping over two orders of magnitude per mass number at that point. Since yields of 10^{-10}-10^{-12} are required at present for the most sensitive out-of-beam experiments, it seems these will terminate at mass 115 or 116. At a cost of over a factor of 100 per mass number, very great improvements in sensitivity will be required to go much beyond this point. Figure 10 is based on the belief that the LCN of Ce (^{122}Ce) is the best starting point to produce very neutron-deficient Ce nuclei, and calculations using the Nd LCN instead as a starting point do, indeed, give considerably lower yields for the appropriate 2pxn, and αxn products. This trend is in accord with the proton spallation results, where the compound nucleus is very far away, and in that case yields (σ/σ_{CN}) of $\sim 10^{-8}$ occur[8]) where the highest yields calculated here would be $\sim 10^{-1}$.

The situation we have just described is summarized in fig. 11, which is a section of the chart of nuclides for the region under discussion. The stable isotopes are shown for reference as the black squares, and the LCN line, previously described, is the dashed line. The black circle on this line corresponds to ^{128}Nd, the only LCN we have produced in this region. The heavy solid line is the place where P_p/P_n $(\Gamma_p/\Gamma_n) = 1$. The accuracy in locating this line may be estimated from the uncertainty in k to be ~ 1 AMU in the Ce region, and ~ 2 AMU in the Os region. An excited compound nucleus will tend toward this line from either side by preferentially evaporating the appropriate nucleons; and its location, together with the LCN line, mainly determines the behavior we have described. That these lines are so closely parallel is somewhat accidental, as far as we can see, and it means that the situation presented here for Ce is qualitatively similar throughout this region. The highest yield we can expect for producing nuclei along the $P_p/P_n = 1$ line, using HI reactions, is ~ 30% of σ_{CN} and the drop-off to 10^{-6} and 10^{-12} is roughly indicated in fig. 11. The two widely spaced solid lines are the neutron and proton drop lines, where the latter assumes the proton to be unbound by the Coulomb-barrier energy. Although there are many very-neutron-deficient nuclei which cannot be produced in useful amounts by this method, almost all of these will have very short proton-decay lifetimes. Using the estimates of Goldanski$^{9)}$, one finds that for a given Z, these lifetimes drop from 10^{2} sec to 10^{-12} sec in just two or three mass numbers and this change is expected to occur somewhere between the LCN and $Y \leqslant 10^{-12}$ lines.

These calculations have been extended to lower Z nuclei and give results that are in good accord with data in the Kr region$^{10)}$. It appears, therefore, that this rather simple approach seems to have validity over a large portion of the periodic table.

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Figure Captions

Fig. 1. Minimum excitation energies for Ce compound nuclei resulting from various target-projectile systems.

Fig. 2. Net in-beam and out-of-beam spectra resulting from the bombardment of ^{112}Sn with 88 MeV ^{16}O ions.

Fig. 3. Particle spectrum resulting from bombardment of ^{112}Sn with 76 MeV ^{16}O.

Fig. 4. Gamma-ray spectra in coincidence with evaporated protons (above) and alpha particles (below).

Fig. 5. Net in-beam and out-of-beam spectra resulting from bombardment of ^{144}Sn with 155 MeV ^{28}Si ions.

Fig. 6. Effective binding energies (see text) for Ce and Os nuclei.

Fig. 7. Relative probabilities for emission of neutrons, protons, and alpha particles from Ce nuclei.

Fig. 8. Relative yields (calculated) for different elements (Z values) resulting from evaporation of four nucleons from Nd compound nuclei.

Fig. 9. Expected yields (see text) of Ce nuclei resulting from various reactions.

Fig. 10. Continuation of fig. 9 to lower yields. The dashed line connects the points of highest yields for each mass number.

Fig. 11. Section of the chart of nuclides showing the location of the various lines discussed in the text.

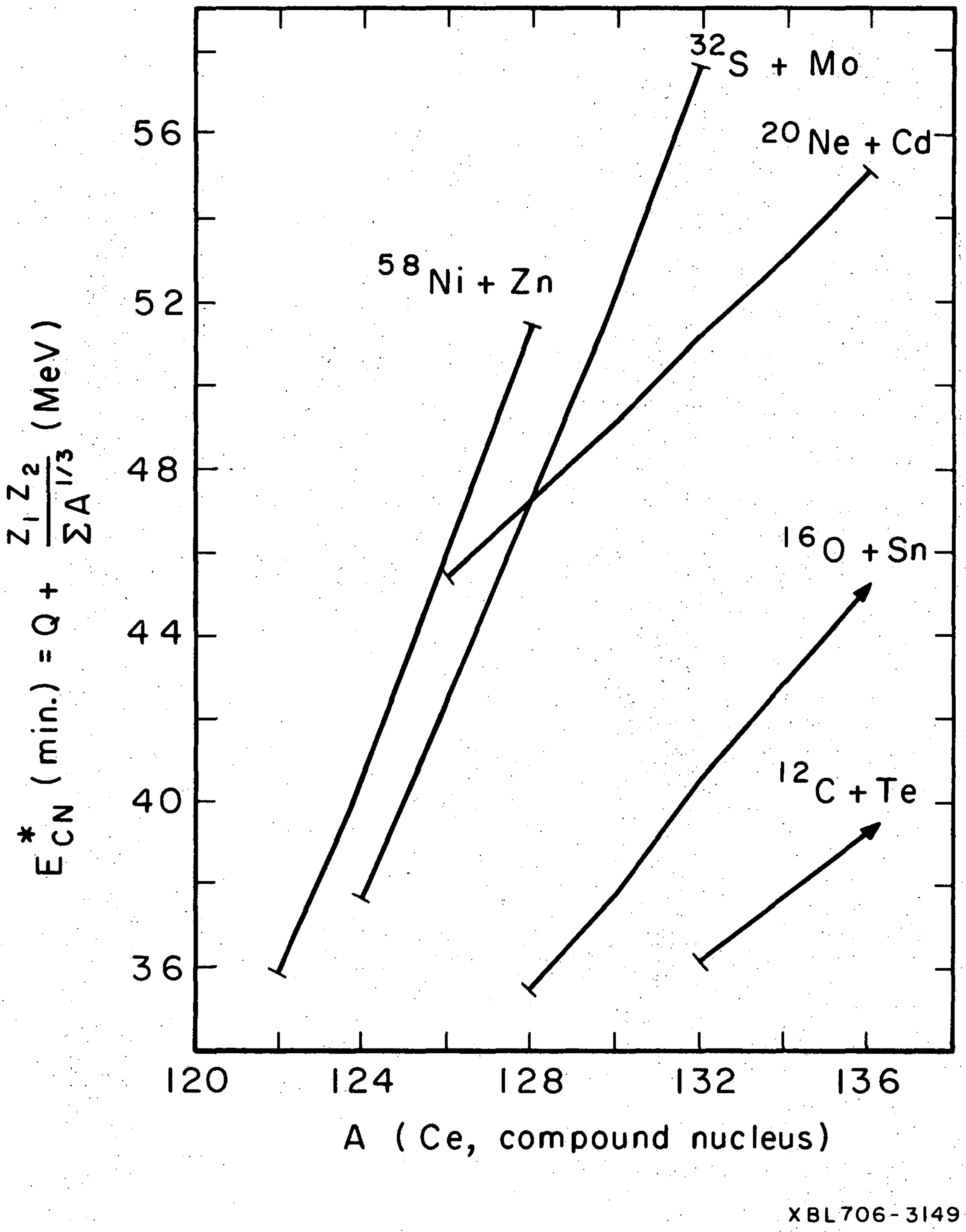
^{32}S + Mo
^{20}Ne + Cd
^{58}Ni + Zn
^{16}O + Sn
^{12}C + Te
56
52
48
44
40
36
120
124
128
132
136
A (Ce, compound nucleus)
E^*_{CN} (min.) = Q + $\frac{Z_1 Z_2}{\Sigma A^{1/3}}$ (MeV)
XBL706-3149

Fig. 1

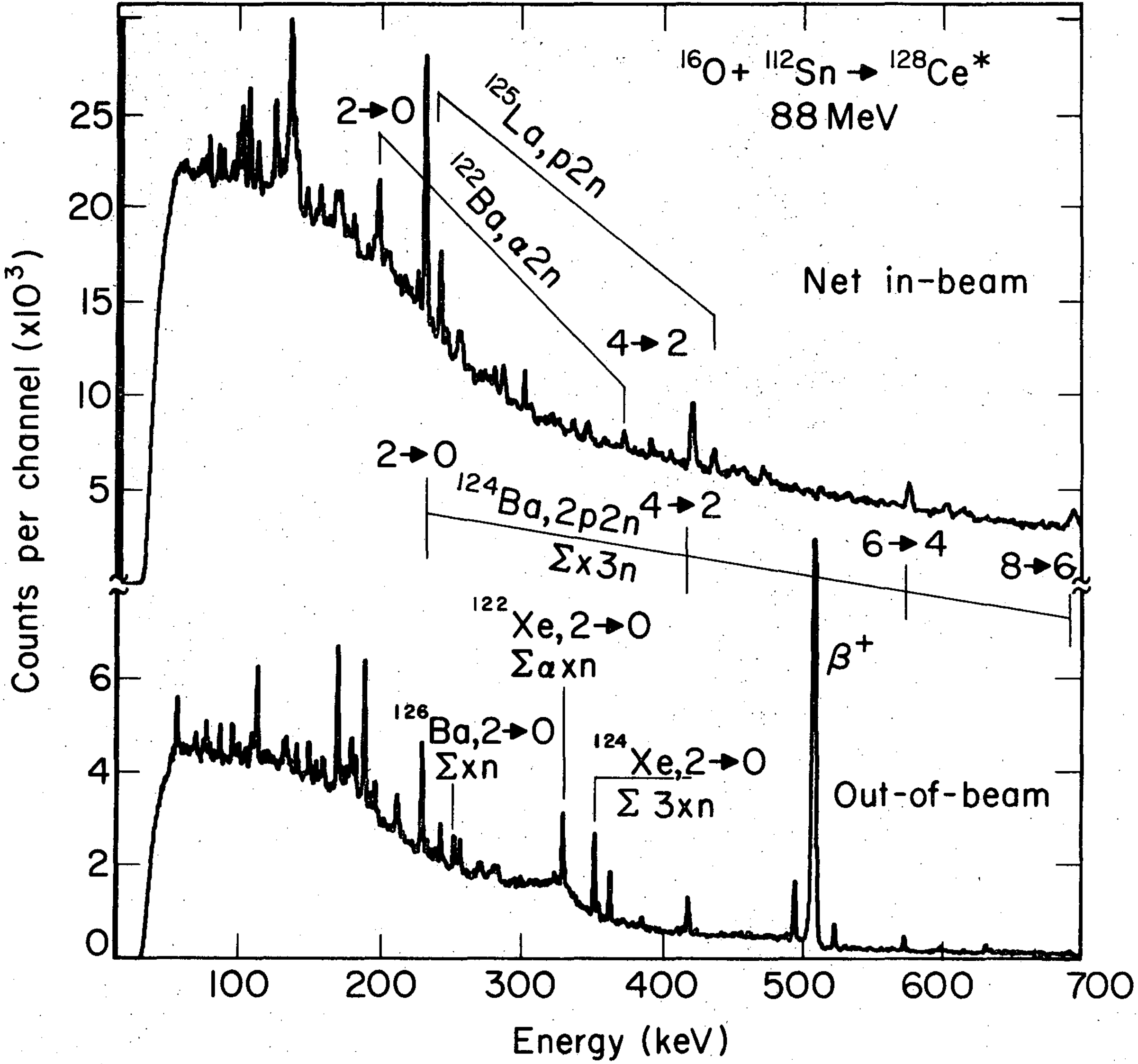

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Fig. 2

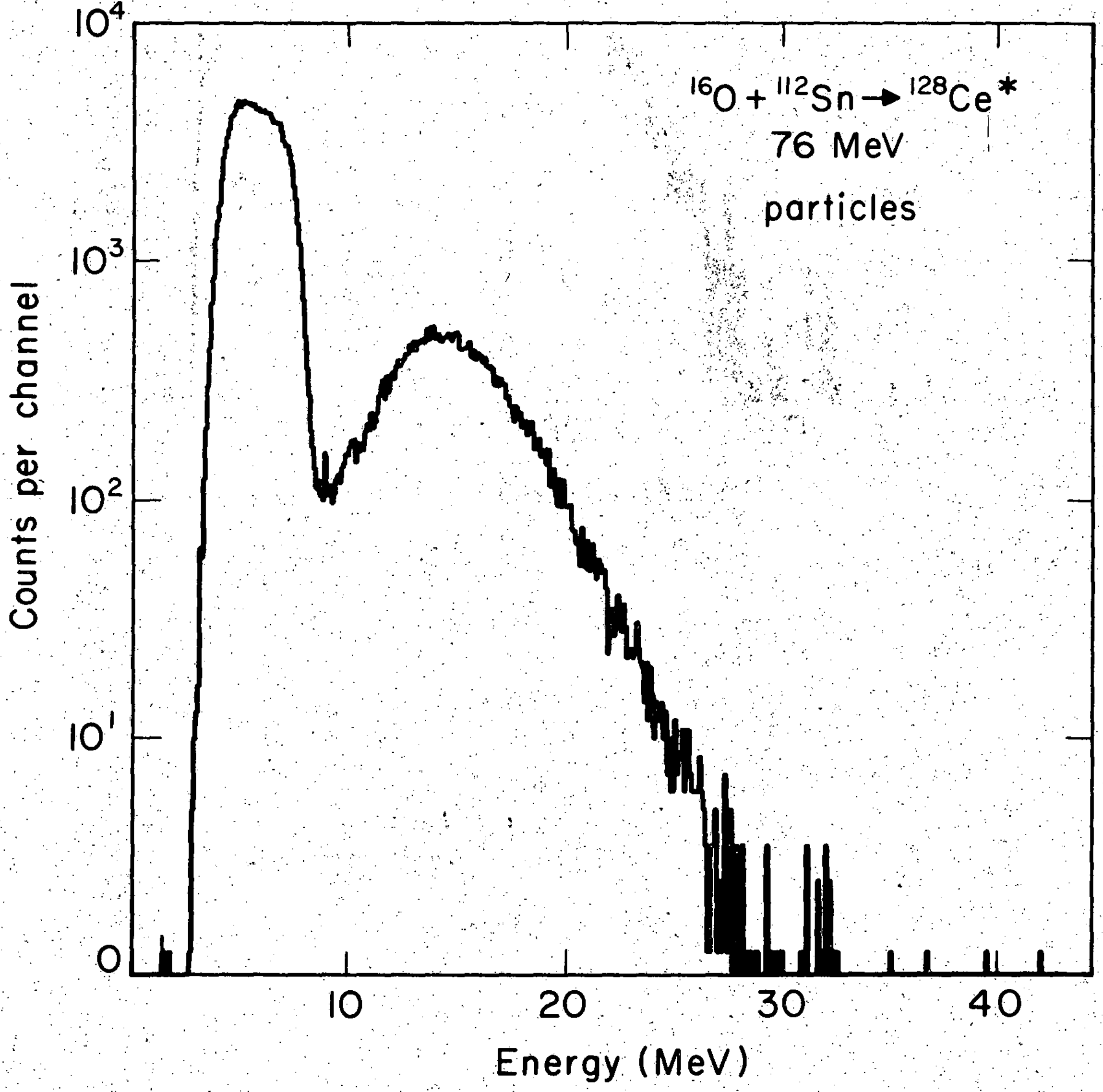

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Fig. 3

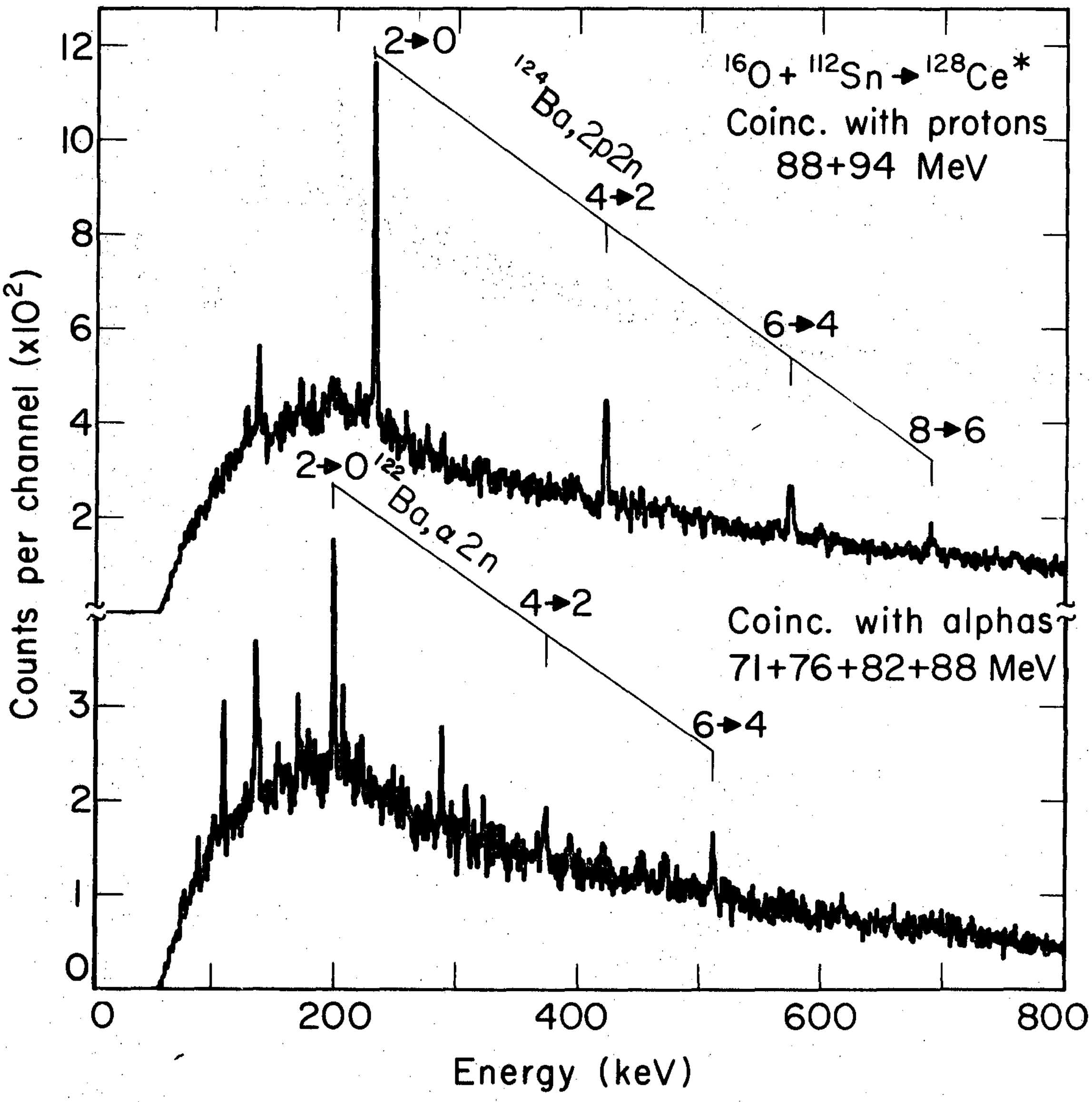

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Fig. 4

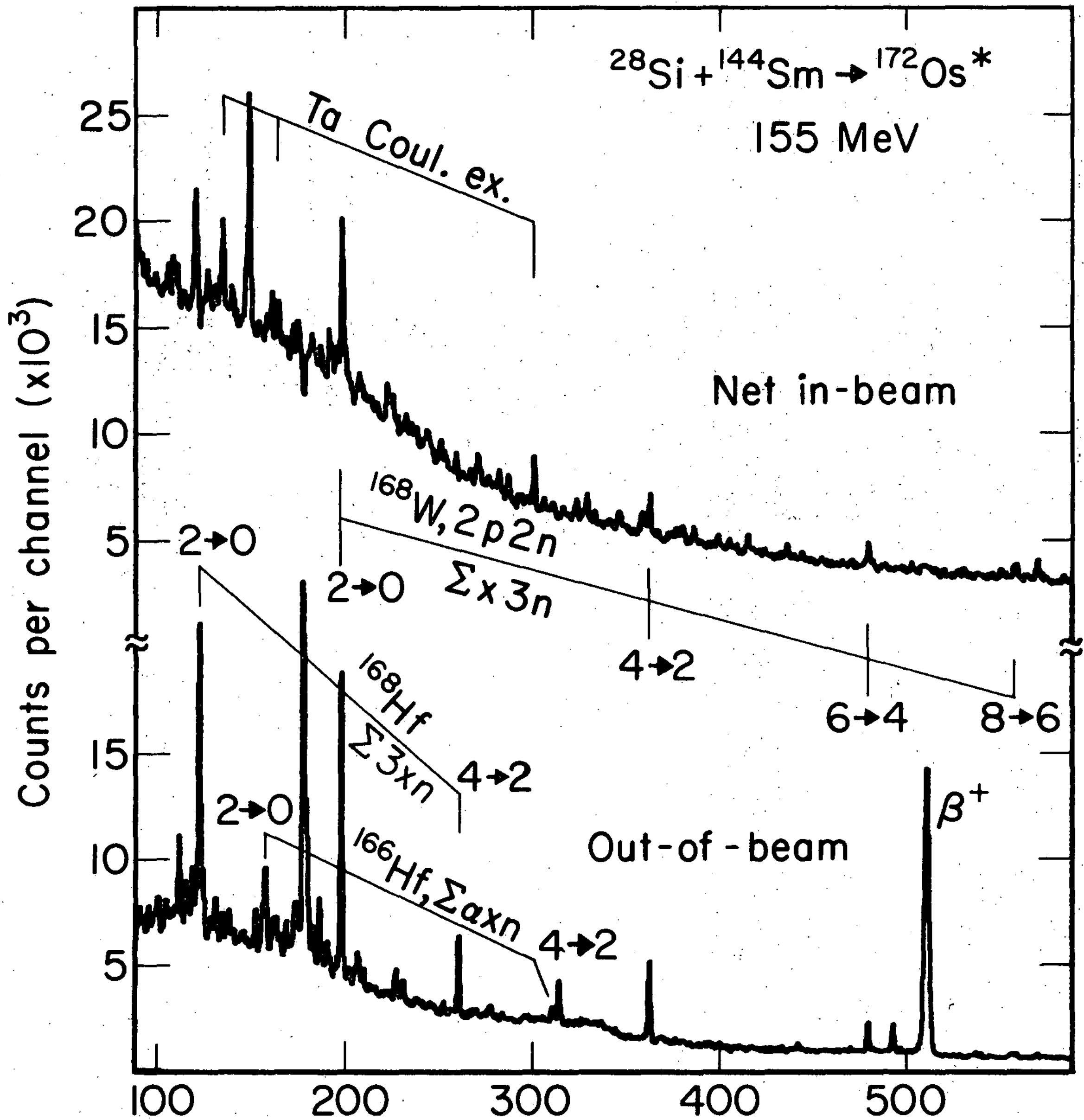

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Fig. 5

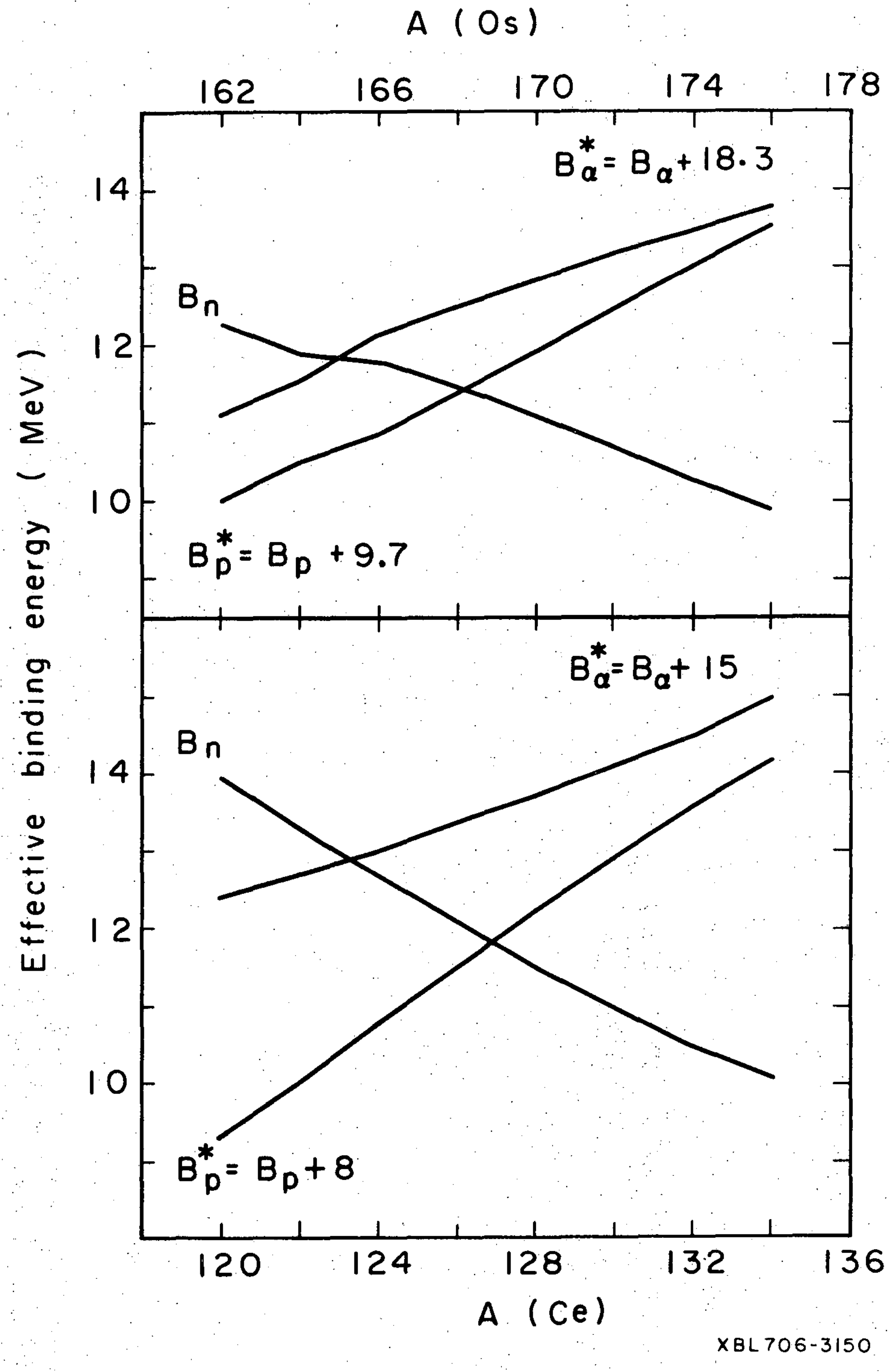
A (Os)
162
166
170
174
178
$B^*_\alpha = B_\alpha + 18.3$
B_n
$B^*_p = B_p + 9.7$
14
12
10
Effective binding energy (MeV)
$B^*_\alpha = B_\alpha + 15$
B_n
14
12
10
$B^*_p = B_p + 8$
120
124
128
132
136
A (Ce)
XBL706-3150

Fig. 6

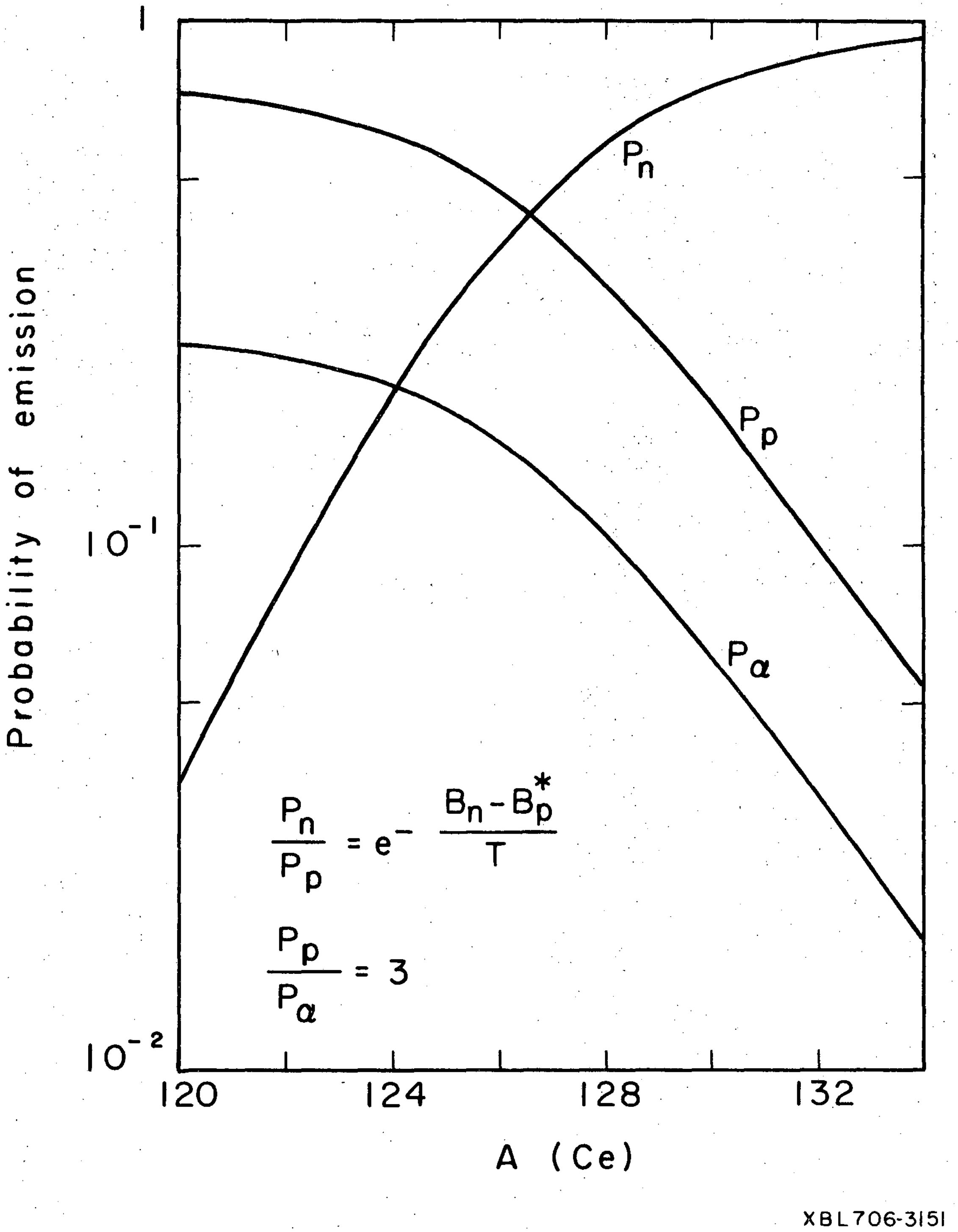
1
10^{-1}
10^{-2}
Probability of emission
P_n
P_p
P_α
$\frac{P_n}{P_p} = e^{-\frac{B_n - B_p^*}{T}}$
$\frac{P_p}{P_\alpha} = 3$
120
124
128
132
A (Ce)
XBL706-3151

Fig. 7

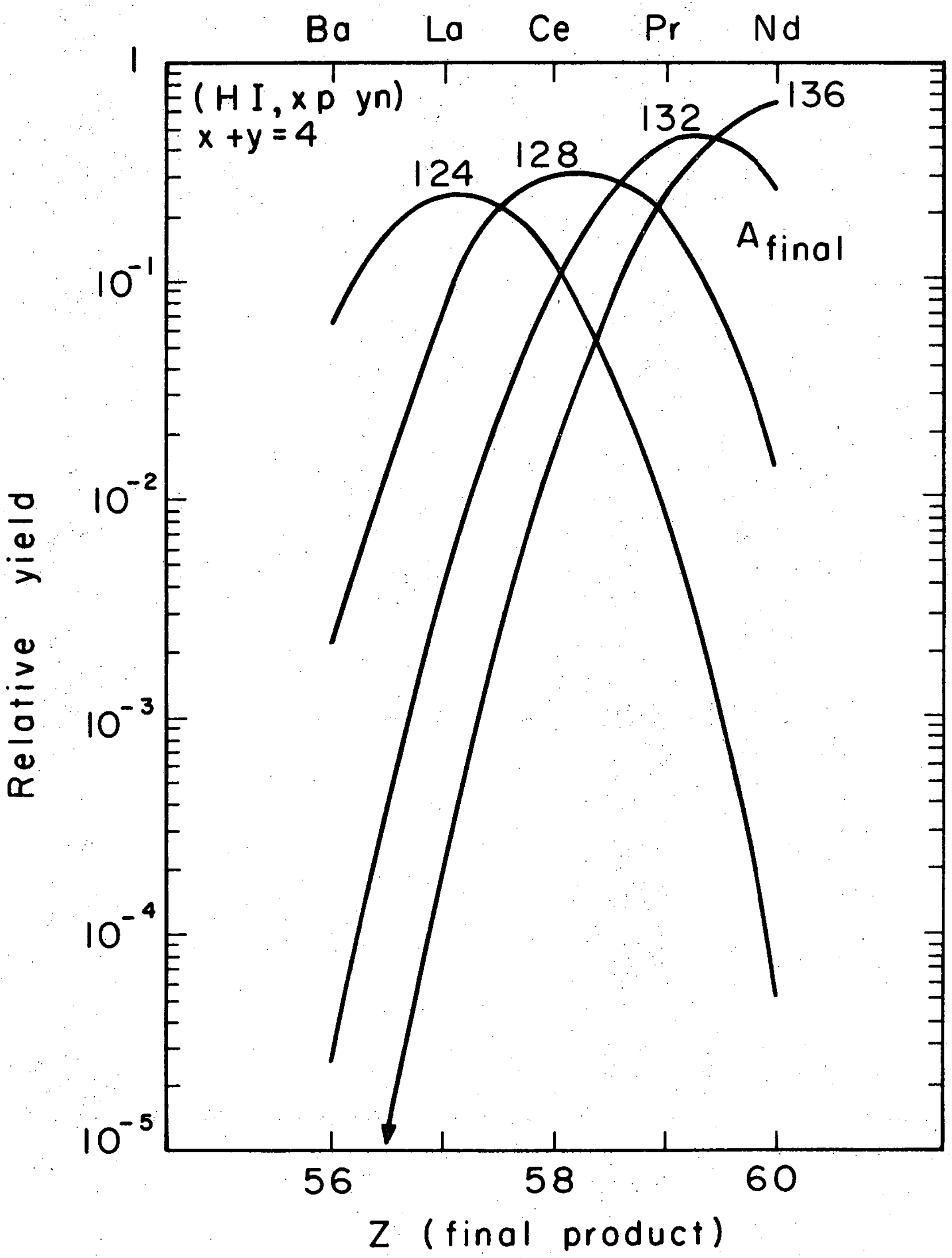

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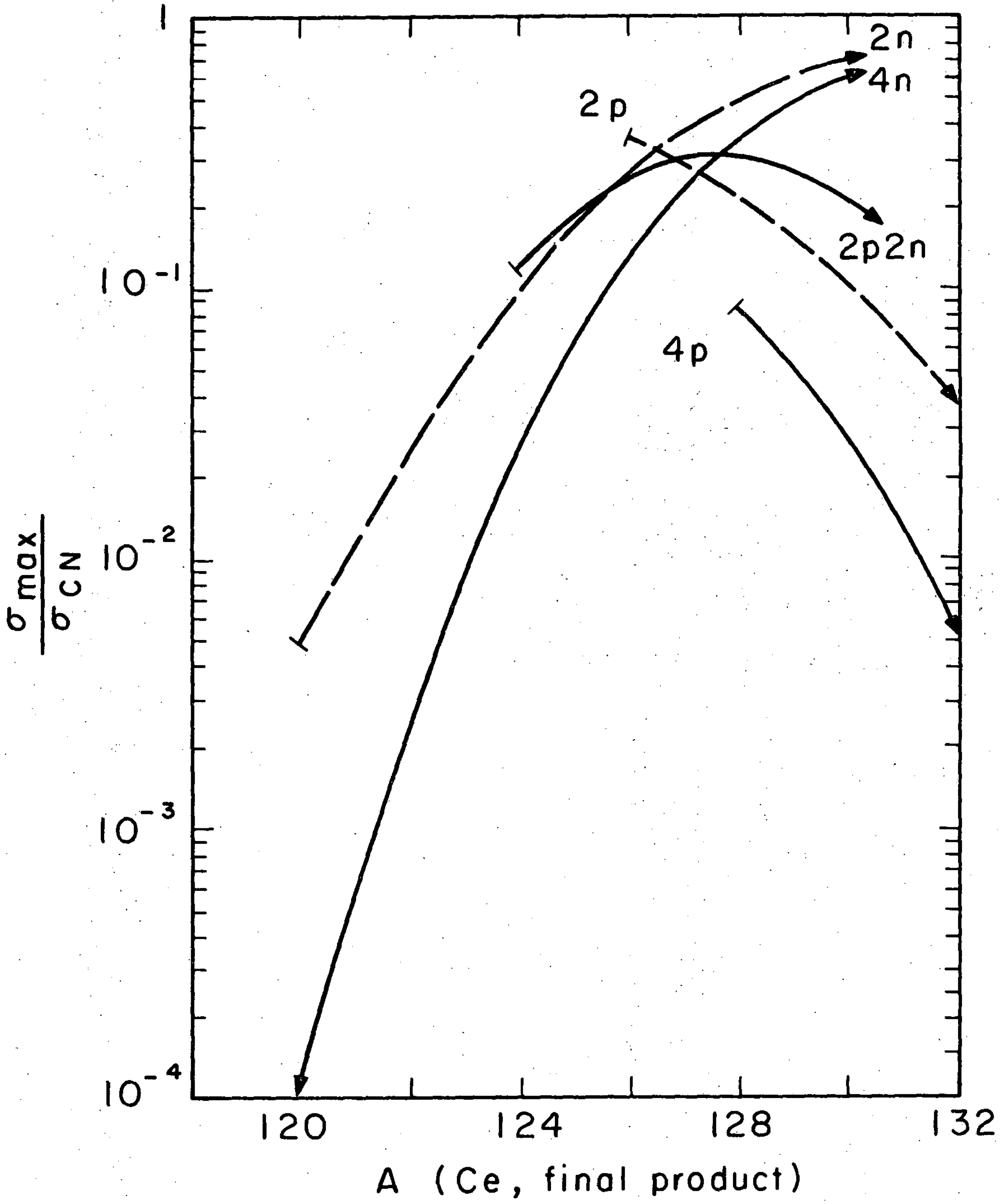
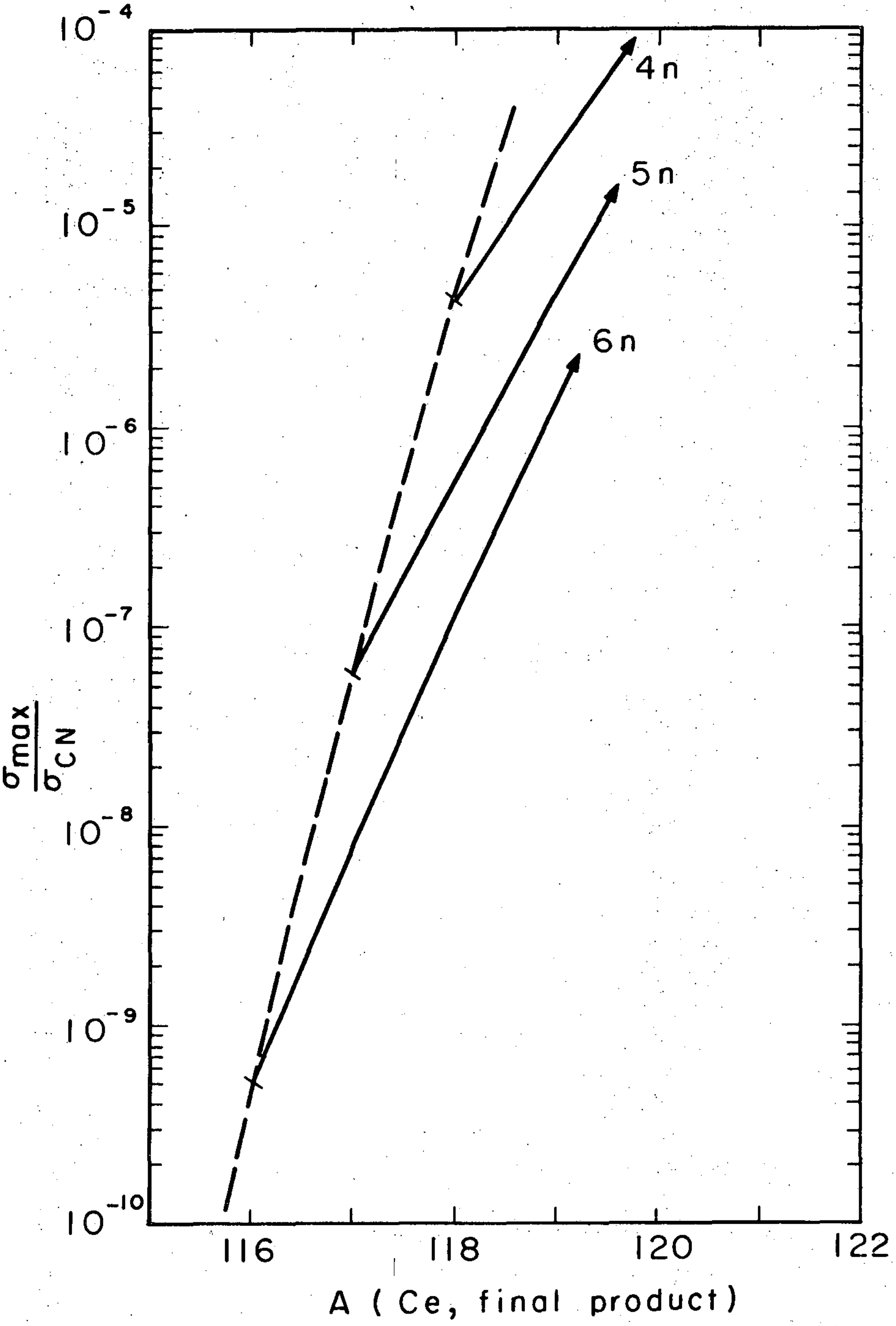
Fig. 8



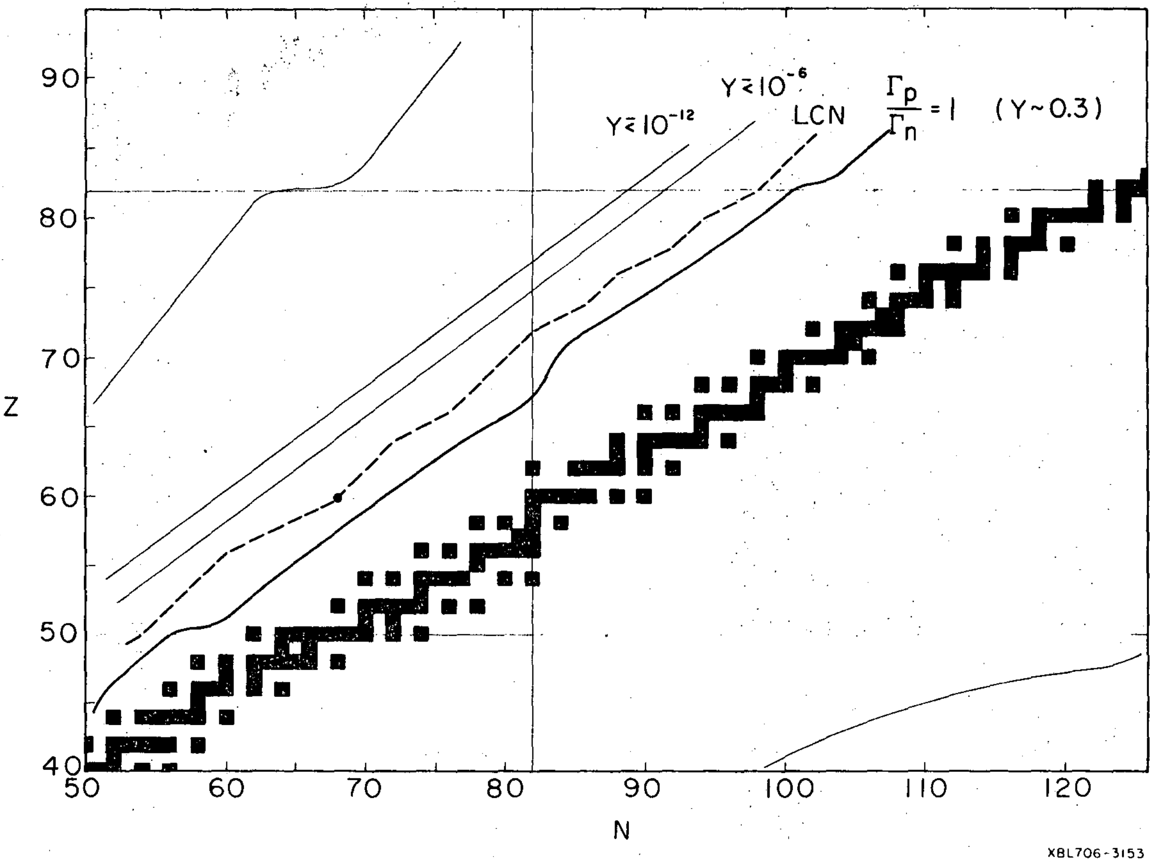
XBL706-3148

Fig. 9



XBL706-3147

Fig. 10



90
80
70
60
50
40
Z
50
60
70
80
90
100
110
120
N
Y ≲ 10⁻¹²
Y ≲ 10⁻⁶
LCN
Γp/Γn = 1 (Y~0.3)
XBL706-3153

Fig. 11

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