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May 1992

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Nuclear Science Division,
Lawrence Berkeley Laboratory
MS 70A-3307
University of California
Berkeley, CA 94720

and

Department of Chemistry
University of California
Berkeley, CA 94720

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The Spontaneous Fission Properties of ^{259}Lr

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Nuclear Science Division, Lawrence Berkeley Laboratory, MS 70A-3307, and Chemistry Department, University of California, Berkeley, CA 94720

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Abstract

We have measured the mass and kinetic-energy distributions of fragments from the spontaneous fission of ^{259}Lr. The ^{259}Lr was produced via the ^{248}Cm(^{15}N,4n) reaction with a production cross section of 100 nb using 81-MeV projectiles. The kinetic energies and times of the alpha particles and coincident fission fragments were measured using our rotating wheel system. From these data the half-life and mass and kinetic-energy distributions were derived. The total kinetic energy (TKE) distribution appears to consist of a single component with a most probable pre-neutron-emission TKE of 215±3 MeV. The mass distribution is predominantly symmetric with a full-width at half-maximum (FWHM) of about 20 mass numbers. These results are consistent with trends observed for other trans-berkelium spontaneously fissioning isotopes. We determined the half-life to be 6.14±0.36 seconds by measuring its alpha decay and the observed spontaneous fission half-life was consistent with that value. An energy of 8.439±0.010 MeV was measured for the main alpha transition of ^{259}Lr. We measured a spontaneous fission to alpha-decay ratio of 0.25±0.03 which results in a partial half-life for spontaneous fission of 31±4 seconds, if there are no other appreciable modes of decay.

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I. INTRODUCTION

Since the discovery of increased yield for symmetric mass division in the spontaneous fission (SF) of ^{257}Fm[1], many studies have been done to explore the SF properties of transberkelium elements. Asymmetric mass division was believed to be the most probable mode for SF until narrowly symmetric mass division was seen in the heavier Fm isotopes. The transition from asymmetric to symmetric mass division in Fm isotopes is shown in the schematic of mass-yield curves for the SF of trans-Bk isotopes shown in Fig. 1. ^{257}Fm is considered the transition nucleus between the clearly asymmetric mass division of ^{256}Fm and the dramatic change to narrowly symmetric mass division in ^{258}Fm. A similar transition is seen in the No isotopes with ^{256}No being the corresponding transition nucleus. The heaviest Fm and No isotopes, as well as the two Md isotopes depicted in Fig. 1, also have a very high total kinetic energy (TKE) release accompanying the narrowly symmetric mass splits. The average TKE released in the SF of the lighter actinides appears to be a linear function of the Coulomb parameter, $Z^2/A^{1/3}$ (Fig. 2). The heaviest Fm's and ^{260}Md have abnormally high most probable TKE values, 30-40 MeV higher than might be expected from the linear plot. The TKE distributions for some trans-Es isotopes are shown schematically in Fig. 3. ^{262}No has a narrowly symmetric mass-yield distribution[2] wth a high TKE component of about 237 MeV and some evidence for a lower intensity component around 200 MeV.

The SF properties of ^{259}Lr are of interest for several reasons. Prior to this study, no complete measurements of the SF properties of any of the Lr isotopes had been made. SF data for this element are important for a complete understanding of trends and changes in the SF properties of the heaviest elements. Also, as evidenced by Fig. 1, relatively few SF measurements had been done on odd Z elements. One reason for this is the fact that odd Z fission is believed to be hindered by the odd

proton[3] and thus fewer odd Z isotopes have sufficiently large SF probabilities for study. Measurements of any odd Z isotope are valuable additions to existing SF systematics. Thus is it is important to measure the SF properties of ^{259}Lr and determine if they are consistent with trends in the SF properties of other trans-Bk isotopes.

The first observation of ^{259}Lr was reported by Eskola *et al.*[4] in 1971. They produced ^{259}Lr in ^{15}N bombardments of ^{248}Cm targets and determined a half-life of 5.4±0.8 s. The mass assignment was based on the excitation function and was not conclusive. The mass assignment was later confirmed by Gregorich *et al.*[5] using an alpha-daughter recoil catcher technique. The ^{255}Md daughter was caught on aluminum foils placed above the decaying ^{259}Lr parent. The $6.35^{+0.46}_{-0.42}$ s half-life measured for the Lr parent corresponded to the distribution of the 20.07-hr ^{255}Fm granddaughter activities on the Al foils. From measurements of the alpha decay and single fission fragments from the SF of the ^{259}Lr, the SF branch was determined to be 23±2% with the rest decaying by alpha emission. Based only on the single fragment kinetic-energy distribution derived from the data, an average TKE of 200±10 MeV and a broadly symmetric mass distribution were deduced.

In the current experiment we have produced ^{259}Lr by the ^{248}Cm(^{15}N,4n) reaction and have measured the SF-to-alpha ratio, the alpha and SF half-lives, and the energy of the main alpha transition to ^{255}Md. Also, we have measured the kinetic energies of coincident fission fragments and from these derived the TKE and mass-yield distributions.

II. Experimental Techniques

The ^{259}Lr was produced at the Lawrence Berkeley Laboratory 88-Inch Cyclotron via the $^{248}Cm(^{15}N,4n)$ reaction. The target contained 0.50 mg/cm^2 ^{248}Cm (97% isotopic purity) as the oxide (0.55 mg/cm^2 Cm_2O_3) deposited by the molecular plating method[6] in a 0.6 cm diameter circle on 1-mil thick beryllium foil. The 95-MeV beam of $^{15}N^{4+}$ ions passed through a 2.75 mg/cm^2 Be entrance window, 0.3 mg/cm^2 of N_2 cooling gas, and the Be target backing to result in a beam energy in the target material of 81 MeV (laboratory system). The maximum cross section was calculated using SPIT[7] (an evaporation code) to be at 83 MeV for the 4n excitation function. The beam current ranged from 0.50-0.63 particle μA (3.1-3.9 x 10^{12} particles/second).

The recoiling products were stopped in the target chamber in He gas at 1.3 bar. The products were attached to potassium chloride aerosols contained in the He gas and swept through a 1.2 mm i.d. polyvinyl chloride capillary tube to the vacuum chamber of our MG rotating wheel system[8] five meters away. The aerosols containing the activity were collected on 40±10 μg/cm^2 polypropylene foils equally spaced around the periphery of an 80-position horizontal wheel. The efficiency of the He gas jet system was estimated to be 75%. The wheel was stepped at 6-second intervals to position the foils sequentially between pairs of passivated, ion-implanted planar silicon (PIPS) detectors (100 mm^2 active area) located above and below the wheel at 180° to each other. Six pairs of dectectors were used to measure the kinetic energies of the coincident fission fragments and alpha particles. Five pairs were located in the first five positions following the collection position. The sixth pair was located 7 positions away from the fifth pair to more adequately measure the longer-lived background activities. The source-to-detector distance was about 2.0 mm, resulting in an efficiency for the detection of alpha particles in a

given detector of 30% and 60% for the detection of a fission fragment. The alpha-particle energy resolution was about 40 keV in the top detectors and about 60 KeV in the bottom detectors; the latter being larger because of energy degradation in the polypropylene foil. The wheel containing the foils was changed every 40 minutes (5 revolutions of the wheel) to minimize the buildup of long-lived activities.

Off-line alpha-energy calibrations were obtained by measuring the known alpha groups from ^{212}Bi and ^{212}Po in equilibrium with a ^{212}Pb source. On-line alpha-energy calibrations were obtained using ^{213}Fr and ^{213}Rn activities produced from the small amount of Pb impurity present in the target. Activity measurements in each detector were normalized to the known decay of the 34.6-second ^{213}Fr activity to correct for any differences in efficiency from one detector to the next. Decay curves were obtained by integrating the activities measured in the top detectors in the regions of interest for appropriate sets of time intervals. The fits were performed using the Maximum Likelihood Decay by the Simplex method (MLDS) code[9], a code appropriate for relatively small numbers of events. Off-line fission fragment energy calibrations were obtained by measuring ^{252}Cf sources on similar polypropylene foils and using the calibration procedure of Schmitt, Kiker, and Williams[10](SKW) with the constants determined by Weissenberger *et al.*[11] Pulses from alpha particles between 5 and 10 MeV and fission fragments up to 200 MeV were digitized and stored in list mode on magnetic tape by our real-time data acquisition and graphics system (RAGS).[12] The timing requirement for coincidence in off-line sorting was about 2 μs.

III. Results

The alpha-energy spectrum of the ^{259}Lr from a 46-particle μAh bombardment measured in the top detectors of the last five stations is presented in Fig. 4. The most prominent peak in the spectrum at 6.775-MeV is due to ^{213}Fr, the alpha daughter of ^{217}Ac. The ^{217}Ac was produced via the ^{208}Pb(^{15}N,6n) reaction from the small amount of lead impurity in the target. The peak at 8.09-MeV is due to ^{213}Rn, produced by the EC decay of ^{213}Fr (0.55% branch). The alpha-multiplet between 8.56 and 8.65 MeV is from ^{258}Lr, produced via the competing ^{248}Cm(^{15}N,5n) reaction. An energy of 8.439±0.010 MeV was obtained for the alpha particle emitted in the main alpha transition of ^{259}Lr to ^{255}Md. This value is consistent with the less precise previous best value of 8.45±0.02 MeV.[4] The alpha decay of the ^{259}Lr parent imparts enough momentum to the ^{255}Md daughter for it to recoil into the top detector. Off-line measurements of the alpha decay of the ^{255}Fm (produced from the EC decay of the ^{255}Md) in the top detectors were made. The measured alpha energy and half-life were consistent with the known values for ^{255}Fm, another confirmation that the SF activity studied was ^{259}Lr.

The most probable half-life for ^{259}Lr was determined by fitting[9] a two-component decay curve to the 8.439 MeV alpha activity, with the long component half-life fixed at 10^5 seconds. From the fit, presented in Fig. 5, the most probable value for the half-life is 6.14±0.36 seconds. The stated error limits indicate the interval of equal likelihood chances corresponding to a confidence level of 68%.[13] This value is consistent with, but somewhat shorter than, the previously published value of $6.35^{+0.46}_{-0.42}$ seconds[5], probably because of inclusion in the fit of the second, longer-lived component. The two-component fit is more appropriate in this study because the on-line background was adequately measured in the sixth detector

station. A one-component fit to the decay data for coincident SF fragments resulted in a half-life of $6.23^{+0.51}_{-0.53}$ seconds, consistent with the half-life measured for the alpha decay. The one-component fit, presented in Fig. 6, was appropriate because the SF background was found from detector station 6 to be an insignificant contribution (< 1%). The SF to alpha ratio was determined by comparing the initial activities from the decay curves of the alpha activity and the SF coincidences measured in the top detectors. When the half-life in the fit to the SF activity is fixed at the value of 6.14 seconds measured for the alpha activity, the resulting SF to alpha ratio is 0.25±0.03, consistent with the previously reported value of 0.30±0.03 %.[5] The resulting partial fission half-life of 31±4 seconds indicates a SF hindrance factor, calculated[3] relative to both adjacent even-even neighbors, of 6.3×10^3, consistent with the SF hindrance factors observed for other heavy actinides with an odd proton.[3]

The kinetic energies of 442 pairs of coincident fission fragments were measured. The pre-neutron-emission TKE distribution is shown in Fig. 7. The measured post-neutron-emission fragment kinetic energies and derived masses were corrected to pre-neutron-emission values using a saw-toothed $\bar{\nu}(M)$ distribution similar to that measured for ^{252}Cf[14] and ^{256}Fm[15] and used by Balagna *et al* [1] for ^{257}Fm. The average number of neutrons emitted per fission, $\bar{\nu}_T$, was normalized to 4.5, a value estimated for ^{259}Lr from a plot of $\bar{\nu}_T$ vs. mass number.[16] The best Gaussian fit to the TKE distribution gave a most probable pre-neutron-emission TKE of 215.5±2.6 MeV with a FWHM of 40 MeV. The distribution appears to consist of only one component. A summary of the kinetic-energy measurements for ^{259}Lr and the ^{252}Cf standard measured in the same system is given in Table I.

The post-neutron-emission and pre-neutron emission mass-yield distributions are shown in Fig. 8. The mass-yield data are expressed as yield (%) per mass number with the fragment yield normalized to 200%. The post-neutron emission

mass distribution, represented by closed circles, is symmetric with a FWHM of about 20 mass numbers. The pre-neutron-emission mass yield distribution, represented by open triangles, was derived from the kinetic-energy coincidence data using the neutron correction described earlier and shows a decrease in yield near symmetry. The contour plot in Fig. 9 shows the pre-neutron-emission TKE distributions and average TKE as a function of mass fraction. The contours are lines representing equal numbers of events based on data groupings of 10 MeV X 0.02 units of mass fraction. The relative numbers of events are represented by 5 equal increments of 10 events each. Intermediate values were obtained to help smooth and define the contours.

IV. Discussion

The mass distribution for the SF of ^{259}Lr has been added to the schematic of mass-yield distributions for SF of trans-Bk isotopes shown in Fig. 1. The mass distributions of ^{258}No and ^{260}Rf have been measured, making it possible to compare the mass distribution of ^{259}Lr to those of its neighboring isotones. The system used to measure the ^{258}No and ^{260}Rf mass distributions[17] and the system used in this study to measure the mass distribution of ^{259}Lr have essentially the same resolution. The mass distribution of the ^{252}Cf standard measured in each system has a peak-to-valley ratio of about 5 to 1. The number of coincident SF fragments measured for ^{259}Lr was 442, compared to 382 and 300 for ^{258}No and ^{260}Rf, respectively. The transition from asymmetric to symmetric mass division in the Fm isotopes occurs in ^{257}Fm (N=157). A similar transition occurs in the No

isotopes before ^{258}No (N=156), presumably in ^{256}No which appears to be the transition nucleus.[18] If this trend continues to higher Z, we expect the transition to occur at N<156 for the Lr and Rf isotopes. Although the ^{259}Lr and ^{260}Rf mass distributions appear to be somewhat broader than the narrowly symmetric mass distributions for the heavier Fm, Md, and No isotopes, they still seem to be predominantly symmetric, as expected. The effect of adding one and two protons to ^{258}No while keeping the number of neutrons constant at N=156, can be seen in the smooth transition to the broader mass distribution of ^{260}Rf. The mass distribution broadens from a FWHM of about 15 mass numbers for ^{258}No to about 36 mass numbers for ^{260}Rf. A possible explanation for this broadening may be obtained by comparing the TKE distributions of these three isotones.

The most probable pre-neutron emission TKE for ^{259}Lr is 215.5±2.6 MeV with a FWHM of 40 MeV. This value is somewhat high on the linear plot of TKEs from the SF of other acitinides (Fig. 2) and is significantly higher than the values of 204 MeV and 200 MeV measured[17] for ^{258}No and ^{260}Rf, respectively. Unlike the ^{258}No TKE distribution which may have a very low intensity (5%) higher energy component around 232 MeV, the ^{259}Lr and ^{260}Rf TKE distributions appear to consist of only one low-energy component. The TKE distributions of several Fm, Md, No isotopes have been decomposed[2,19] into two Gaussian distributions, one centered at around 200 MeV and the other around 235 MeV (Fig. 3). These two components have been attributed to "bimodal" symmetric fission[19] with one symmetric mode leading to nearly spherical fragments with unusually high TKE release and the other to elongated fragments with lower TKE. The possibility for "multimodal" symmetric fission has been raised[16] because of the extremely large variance of TKE values for symmetric mass division.

The TKE distribution for ^{259}Fm (Fig. 3) consists of a single high energy component centered around 240 MeV[22] and has been attributed to the fact that

symmetric mass division in that isotope results in the formation of fragments near the doubly magic Z=50, N=82 configuration. These fragments are postulated to be nearly spherical and have higher Coulomb repulsion resulting in the high TKE release. The corresponding mass distribution is also very narrow with a FWHM of about 11 mass numbers. The addition of a proton brings about a competition between the two different modes of symmetric fission discussed above and results in two-component TKE distributions for the Md isotopes. The TKE distribution of ^{258}No, with one more proton and a few less neutrons, shows that the predominant mode is symmetric mass division into considerably deformed fragments. The lower energy component makes up about 95% of the total TKE distribution, and the mass distribution has become somewhat broader. The addition of a single proton in ^{259}Lr to give ^{260}Rf seems to result in a TKE distribution made up solely of the low energy component. The ^{259}Lr is likely to split into one Z=50 fragment and one Z=53 fragment, each with too few neutrons to be spherical. This tendency to form these deformed fragments results in a broader mass distribution and a negligible component of high-energy, mass-symmetric fission in ^{259}Lr. The contour plot (Fig. 9) shows a large variance in TKE for symmetric mass division. This is indicative of the multimodal type of fission, with shapes ranging from near-spherical with TKEs approaching the Q value, to deformed shapes with low TKE.

With the addition of another proton, we observe a still broader mass distribution for ^{260}Rf and a single component TKE distribution with a most probable TKE (200 MeV) that is somewhat low on the linear plot (Fig. 2). These properties indicate a type of fission similar to the liquid-drop type fission observed in the preactinide region and in the higher energy fission of the lighter actinides.[18] The current understanding of the spontaneous process seems to explain the trends in the kinetic-energy and mass distributions from the SF of the trans-Bk isotopes so far observed. However, the exact role of shell effects and liquid-drop forces in the parent nuclide

and resulting fragments is still not completely understood. A complete theoretical description of the fission process is a tremendous challenge that has not yet been realized. The goal of a comprehensive theoretical model that reproduces the observed trends, and explains the sudden transitions resulting from the addition of a single nucleon, requires still greater advances by theorists and experimenters alike.

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TABLE I. Properties of the measured (post-neutron-emission) and calculated initial (pre-neutron-emission) fragment kinetic-energy distributions for ^{259}Lr and the ^{252}Cf standard measured in the same system. Energies are given in MeV, based on the SKW calibration method (Ref. 10) with the Weissenberger constants (Ref. 11).

	259Lr		252Cf	
	Pre-n	Post-n	Pre-n	Post-n
Total kinetic energy				
Average	214.2	210.3	184.3	181.6
Most probable[a]	215.5	212.3	185.6	183.3
σ	17.0	17.0	16.0	15.7
FWHM[b]	39.9	40.0	37.6	36.9
Heavy fragment energy				
Average	100.0	99.0	79.6	78.6
Most probable[a]	101.4	102.0	79.6	78.7
σ	11.6	10.5	11.4	11.3
FWHM[b]	27.2	24.6	26.8	26.6
Light fragment energy				
Average	114.1	111.9	104.8	102.9
Most probable[a]	115.1	113.2	106.1	104.2
σ	7.7	7.5	9.7	9.6
FWHM[b]	18.1	17.6	22.8	22.6

[a]Standard deviation of the most probable values from the Gaussian fits is about 1.1%.

[b]Full-width-at-half-maximum, calculated from 2.35σ for Gaussian fit to the top half of the peak.

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FIGURE CAPTIONS

1. Schematic of mass-yield distributions, normalized to 200% fission fragment yield, for SF of trans-Bk isotopes (from Ref. 18).
2. $\overline{TKE}$ vs. $Z^2/A^{1/3}$. The solid line is linear fit of Viola;[20] dashed line is from Unik *et al.* [21] All of the $\overline{TKE}$ values have been normalized to the calibration parameters of Weissenberger *et al.* [11]
3. Schematic of TKE distributions for SF of some heavy trans-Es isotopes (from Ref. 16).
4. Alpha-energy spectrum for ^{259}Lr decay measured in the top detectors of the last five detector stations for a 46-particle μAh bombardment of ^{248}Cm with ^{15}N ions. The continuum in the low energy part of the spectrum is due to the decay of the A=8 isotopes produced by interactions of the beam with the beryllium target backing.
5. Decay curve of the ^{259}Lr alpha-peak at 8.439 MeV. The times indicated are the times since the end of collection of the samples. The average count rates during the time intervals are indicated by the symbols and the center curve is the most probable fit to the data. The upper and lower curves are the limits which encompass 68% of the probability in a Poisson distribution centered on the number of counts expected during the interval (from the most probable fit).
6. Decay curve of the ^{259}Lr SF coincidences. The significance of the curves is the same as in Fig. 5.
7. Gaussian fit to the pre-neutron-emission TKE distribution from the SF of ^{259}Lr. The data are in groupings of 10 MeV.

8. Post-neutron-emission (closed circles) and pre-neutron emmission (open triangles) mass-yield curves for ^{259}Lr (442 events). The pre-neutron-emission mass-yield curve for ^{259}Lr was derived from the SF coincidence data using a $\bar{\nu}(M)$ function similar to that used by Balagna *et al.*[1] for ^{257}Fm with $\bar{\nu}_T = 4.5$. The data are in groupings of four mass numbers. The curves are added only to serve as visual aids.

9. Contour diagram for ^{259}Lr fission yield as a function of pre-neutron-emission TKE and mass fraction. The contours indicate equal numbers of events based on data groupings of 10 MeV x 0.02 units of mass fraction. Contours labeled 1 through 5 represent 10 through 50 events, respectively.

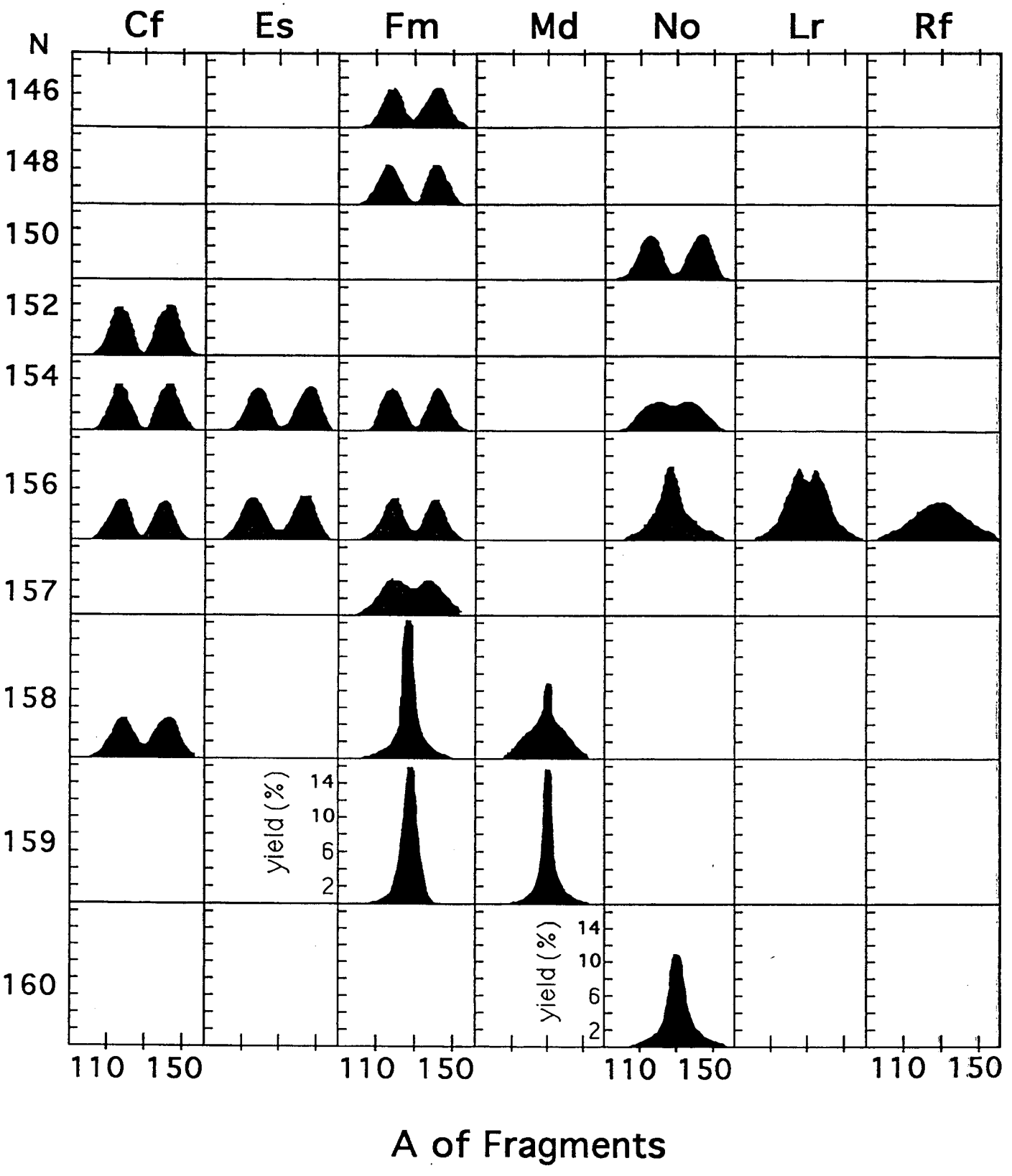
N
Cf
Es
Fm
Md
No
Lr
Rf
146
148
150
152
154
156
157
158
159
160
yield (%)
14
10
6
2
110 150
A of Fragments

Fig. 1

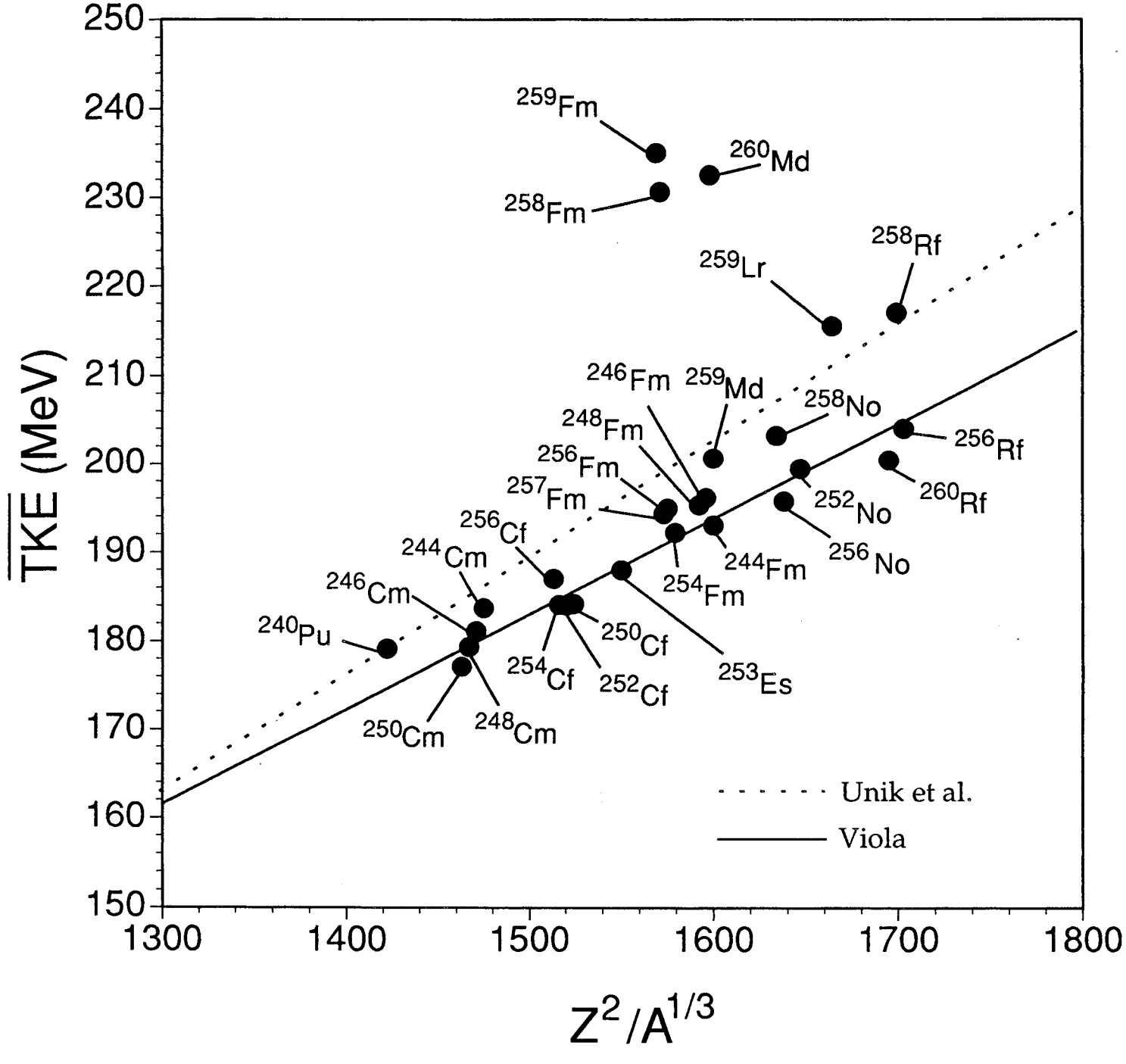
250
240
230
220
210
200
190
180
170
160
150
1300
1400
1500
1600
1700
1800
$\overline{TKE}$ (MeV)
$Z^2/A^{1/3}$
^{259}Fm
^{260}Md
^{258}Fm
^{258}Rf
^{259}Lr
^{246}Fm
^{259}Md
^{258}No
^{248}Fm
^{256}Rf
^{256}Fm
^{257}Fm
^{252}No
^{260}Rf
^{256}Cf
^{244}Cm
^{244}Fm
^{256}No
^{246}Cm
^{254}Fm
^{240}Pu
^{250}Cf
^{254}Cf
^{252}Cf
^{253}Es
^{250}Cm
^{248}Cm
Unik et al.
Viola

Fig. 2

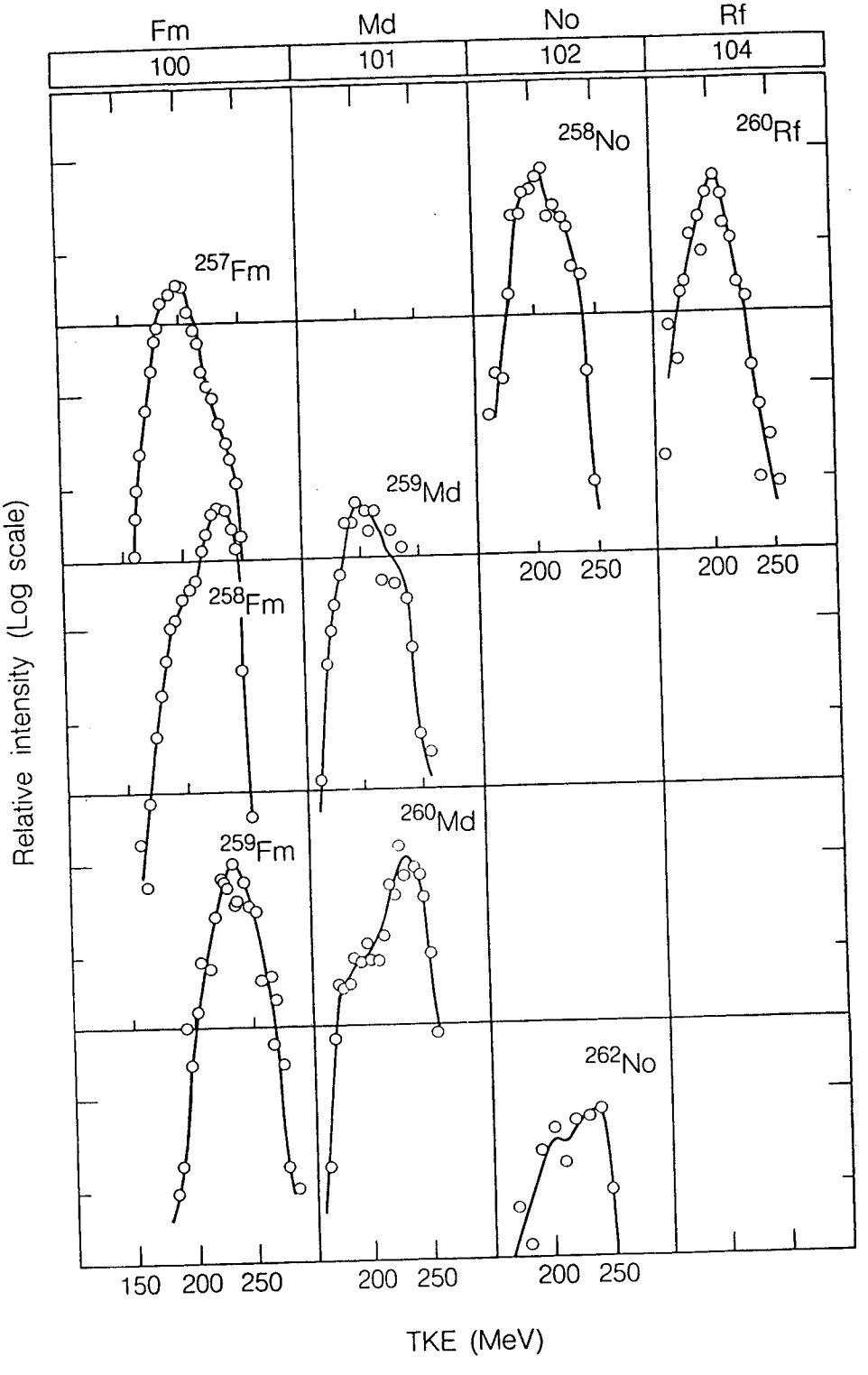

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Fig. 3

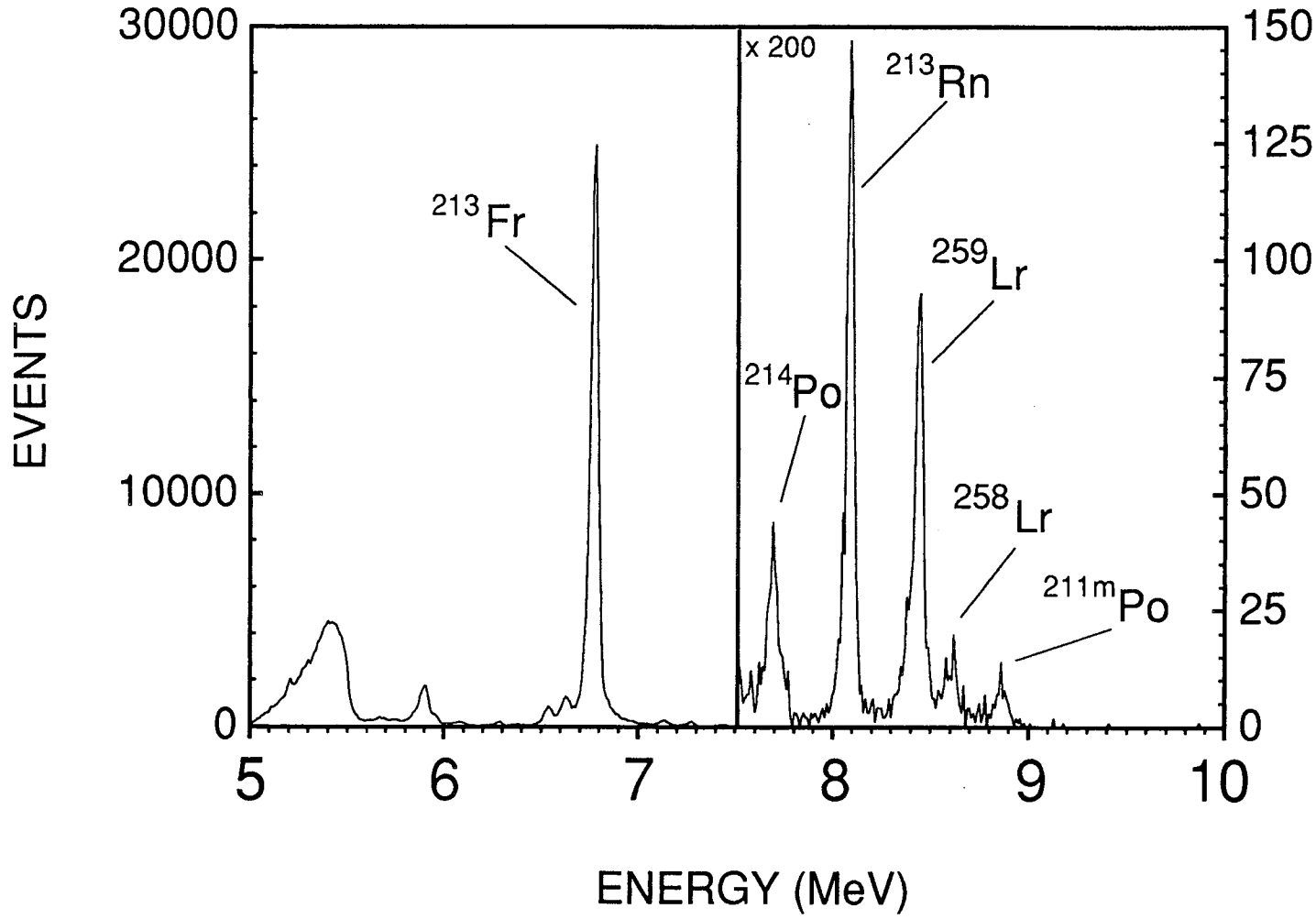
30000
20000
10000
0
EVENTS
150
125
100
75
50
25
0
x 200
^{213}Fr
^{214}Po
^{213}Rn
^{259}Lr
^{258}Lr
^{211m}Po
5
6
7
8
9
10
ENERGY (MeV)

Fig. 4

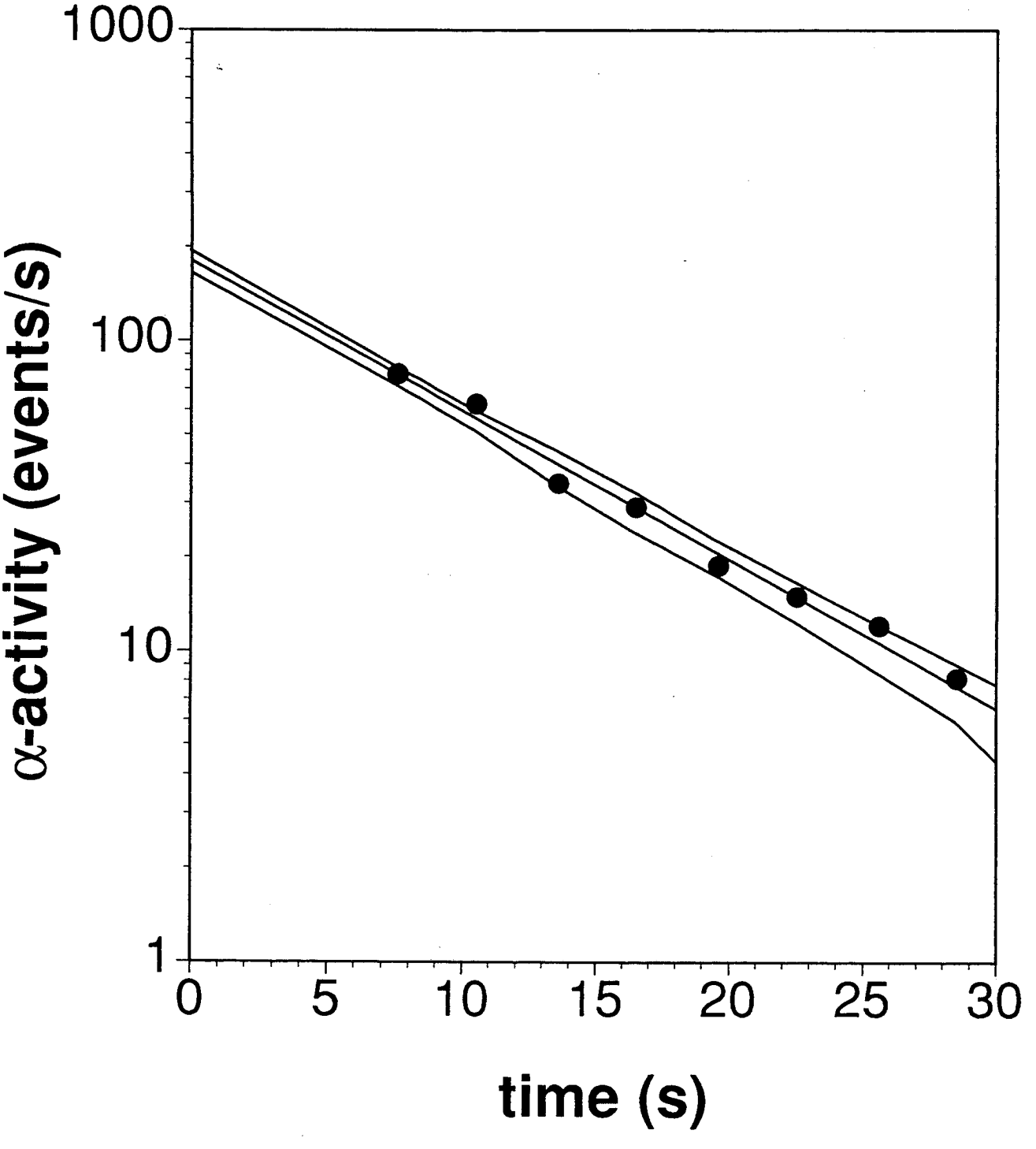

Fig. 5

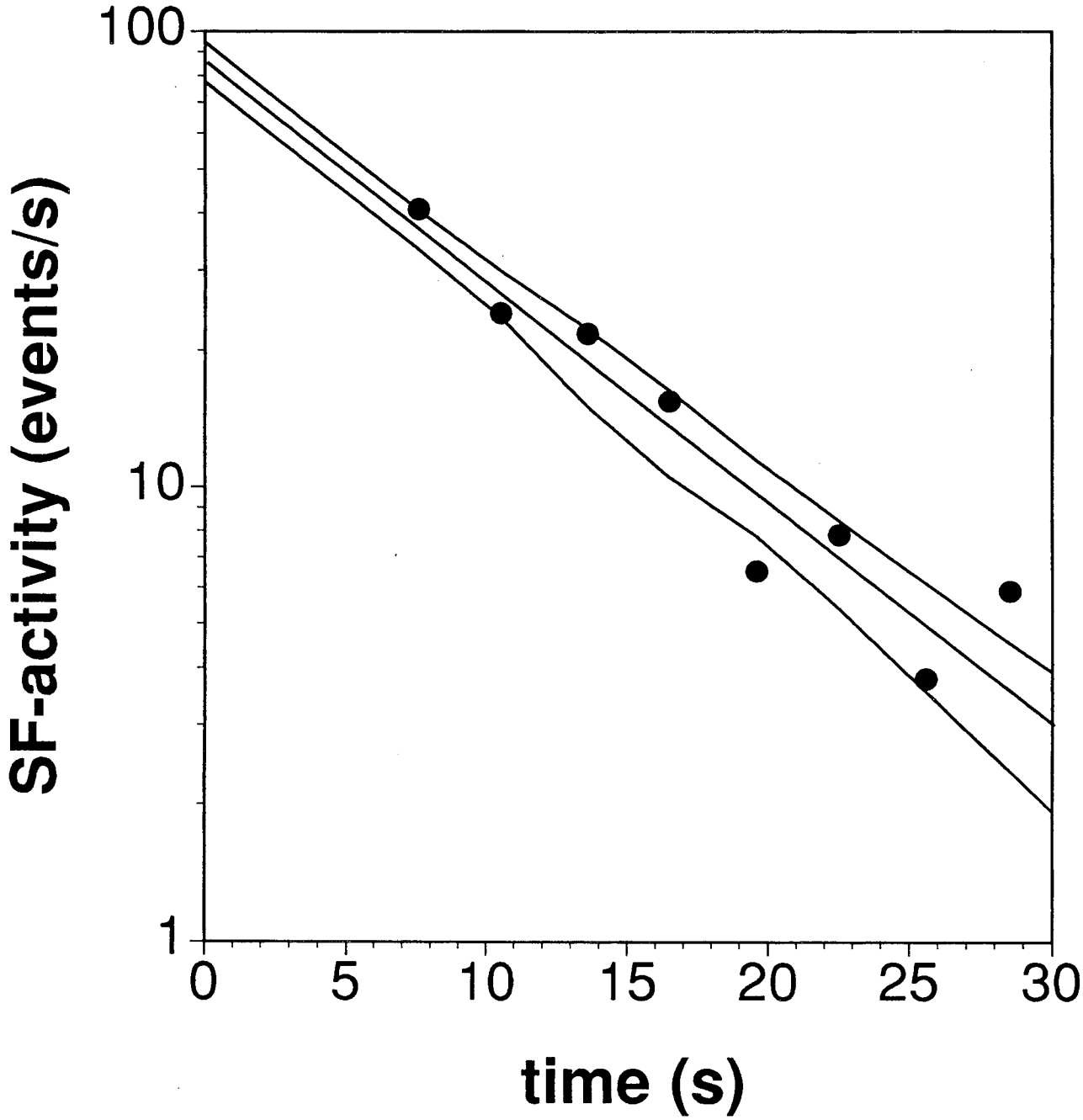

Fig. 6

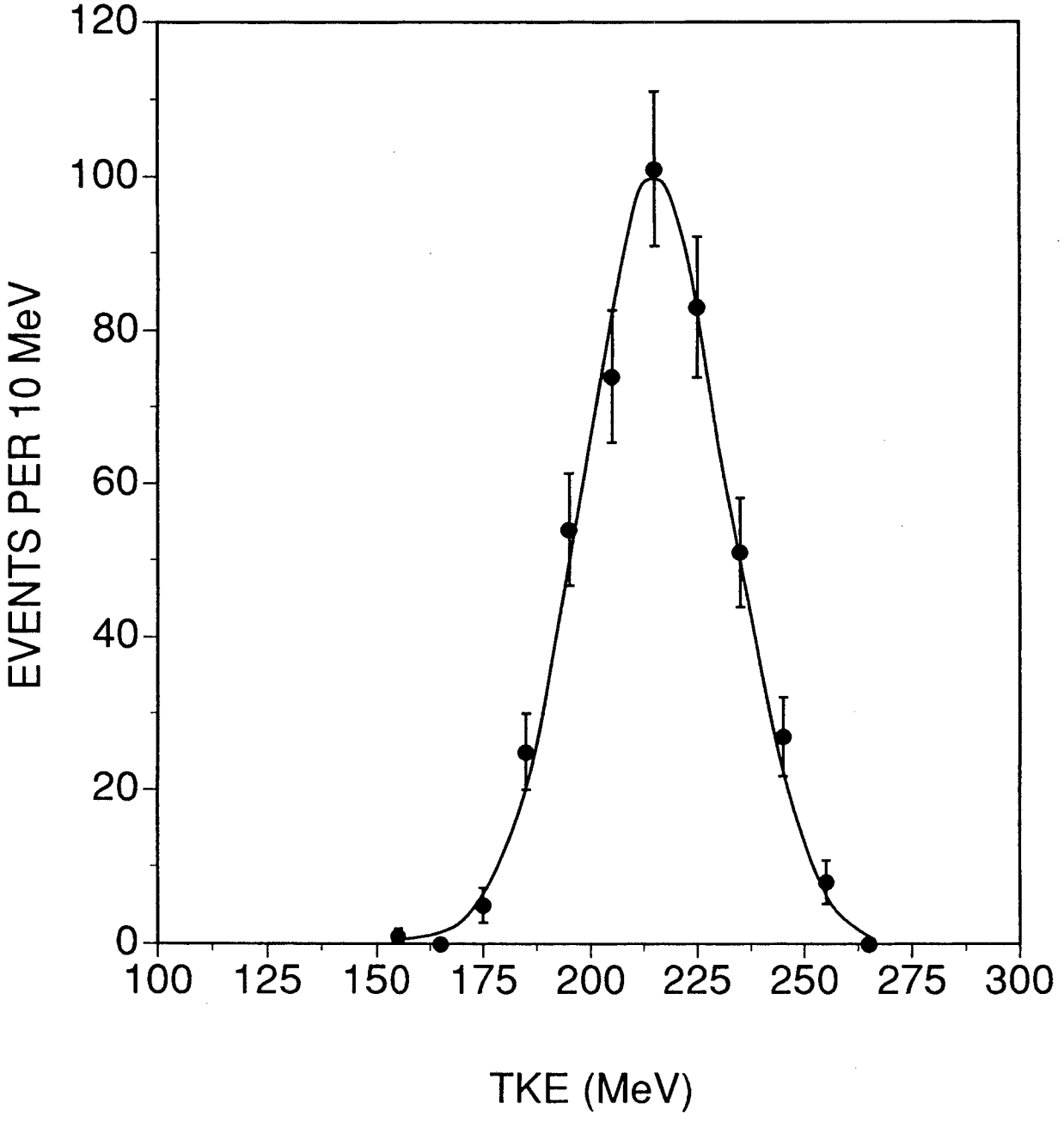
120
100
80
60
40
20
0
EVENTS PER 10 MeV
100
125
150
175
200
225
250
275
300
TKE (MeV)

Fig. 7

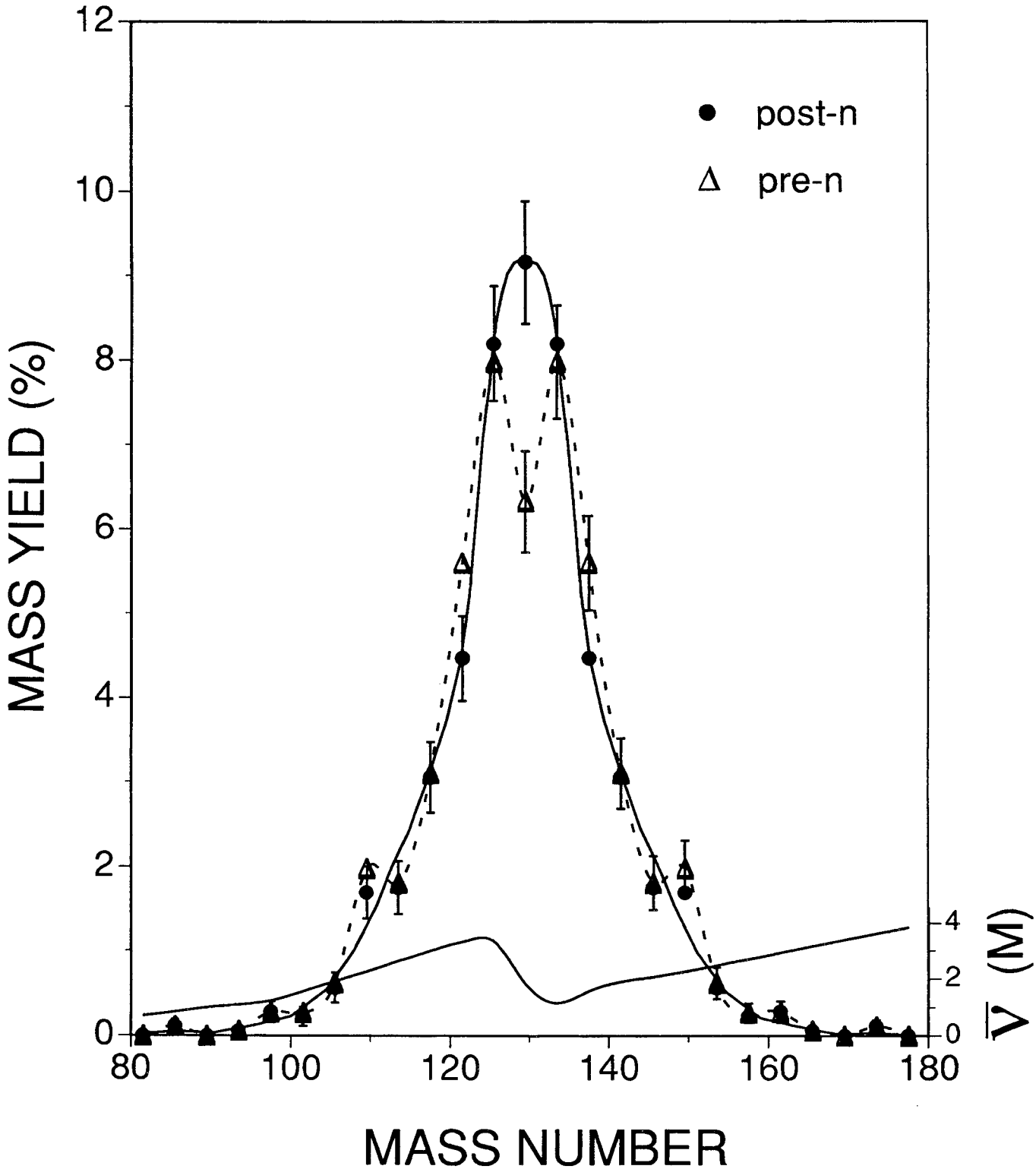
post-n
pre-n
MASS YIELD (%)
MASS NUMBER
$\bar{\nu}$ (M)
0
2
4
6
8
10
12
80
100
120
140
160
180

Fig. 8

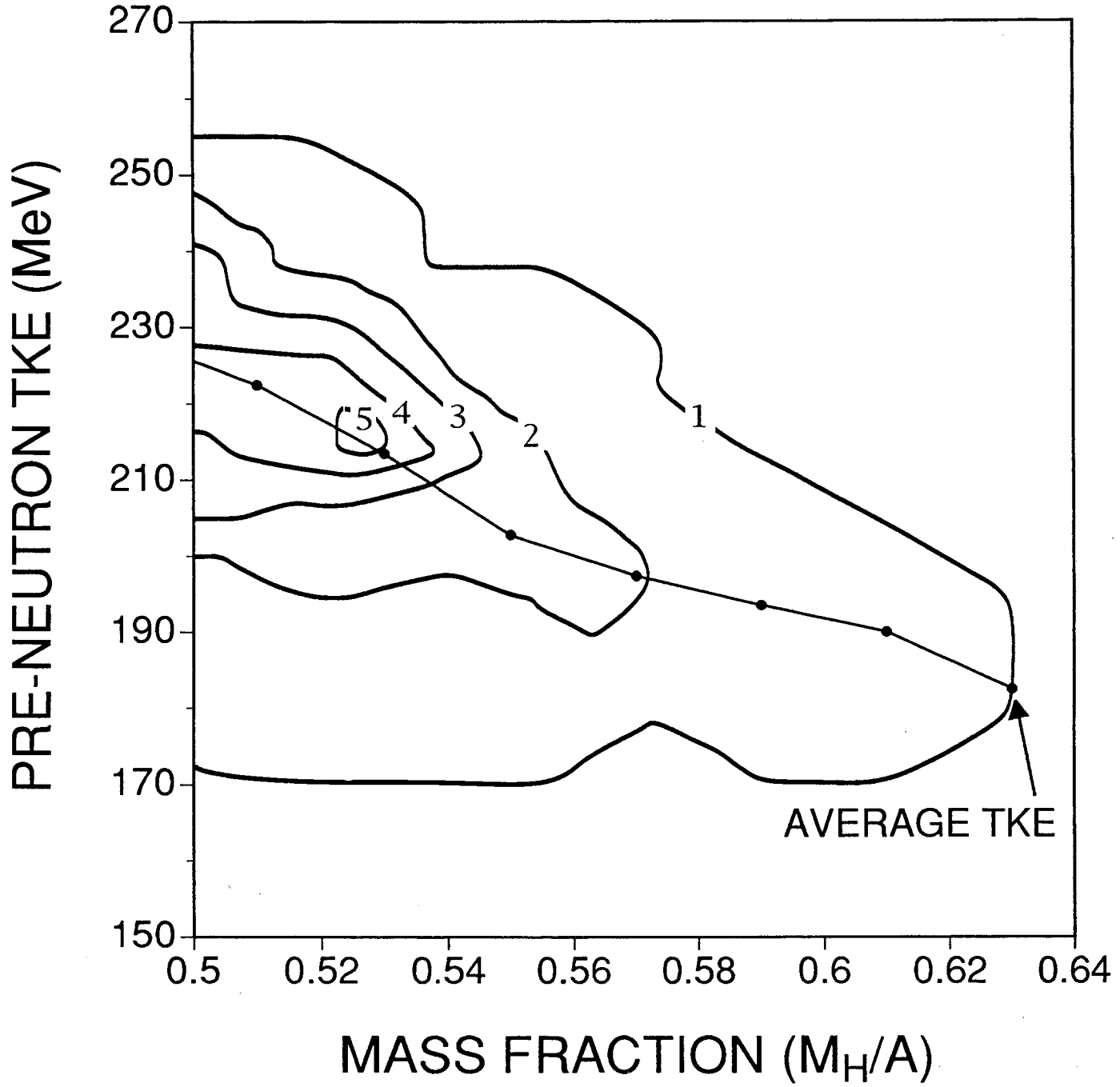
270
250
230
210
190
170
150
PRE-NEUTRON TKE (MeV)
0.5
0.52
0.54
0.56
0.58
0.6
0.62
0.64
MASS FRACTION (M_H/A)
1
2
3
4
5
AVERAGE TKE

Fig. 9

LAWRENCE BERKELEY LABORATORY
UNIVERSITY OF CALIFORNIA
TECHNICAL INFORMATION DEPARTMENT
BERKELEY, CALIFORNIA 94720