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Spontaneous Fission Properties of $^{262}\text{Rf}$

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Spontaneous Fission Properties of $^{262}\text{Rf}$


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Spontaneous fission properties of $^{262}_{104}$Rf


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We have measured the mass and kinetic-energy distributions of fragments from the spontaneous fission (SF) of $^{262}_{104}$Rf. The $^{262}_{104}$Rf was produced via the $^{244}$Pu ($^{22}$Ne, 4$n$) reaction with a production cross section of ~0.7 nb using 114.4-MeV projectiles. The kinetic energies and times of the coincident fission fragments were measured using our rotating wheel system. From these data the half-life, 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±2 MeV. The mass distribution is symmetric with a full width at half maximum of about 22 mass numbers. These results are consistent with trends observed for other trans-berkelium spontaneously fissioning isotopes. We determined the half-life to be 2.1±0.2 s by measuring its spontaneous fission decay. We also attempted to observe the alpha decay of $^{262}_{104}$Rf by searching for alpha decay correlated in time with SF from the alpha daughter, 1.2-ms $^{258}$No. We observed no such decays and have set an upper limit of 0.8% (68% confidence level) on the alpha decay branch of $^{262}_{104}$Rf.

I. INTRODUCTION

The spherical proton shell at Z=100 and neutron shell at N=152 have been shown to increase the stability of the nucleus against spontaneous fission (SF) [1]. However, the predicted deformed proton shell at Z=108 and neutron shell at N=162 have been the center of some debate. Macroscopic-microscopic calculations using these deformed shells provide two opposing results: one that predicts decreased stability toward spontaneous fission and one that predicts increased stability. An increase in stability is predicted by Sobiczewski [2] using calculations that include large deformation spaces. A decrease in stability is predicted by Möller et al. [3], due to the destabilizing effect of a deep new fission valley which leads to compact fission fragment shapes. A resolution of this difference in predicted half-lives is important because it can influence the future
direction of heavy element research. Short spontaneous fission half-lives prevent effective chemical and nuclear study, as well as positive identification, of the heaviest nuclei (Z\geq106). Previously, no known nuclei were near enough to these shells to show their influence on the stability against spontaneous fission.

The isotopes $^{265}$Sg and $^{266}$Sg (Z=106, N=159,160) have been reported [4] to have alpha-decay half-lives of 2-30 s and 10-30 s, respectively, with SF branches of 50% or less. These half-lives were inferred by using the phenomenological formula of Viola and Seaborg [5] and the measured alpha energy. The value for $^{266}$Sg is closer to the half-life estimated by Smolańczuk et al. [6] for increased stability (~1 m) than to the half-life estimated by Möller et al. [7] for decreased stability (~100 \mu s).

In these experiments [4], the SF half-life of $^{262}$Rf was measured to be $1.2^{+1.0}_{-0.5}$ s, based on the observation of six events in coincidence with the alpha events from $^{266}$Sg. This was the cause of some concern because this is quite different than the previously accepted value of around 50 ms [8]. It was our goal to produce $^{262}$Rf via the reaction $^{244}$Pu(22Ne,4n) in order to observe the reported 1.2-s activity and verify its half-life and assignment.

In 1981, Hoffman et al. [9] produced a 1.5-s SF activity in the reaction of $^{248}$Cm with $^{18}$O projectiles. Based on the half-life and the measured SF properties, "the most-likely assignment" for this 1.5-s activity was $^{259}$Fm. In 1982 and 1985, Somerville et al. [10] observed 1.3-s and 47-ms activities using the same reaction. The 47-ms activity was tentatively assigned to the decay of $^{262}$Rf and the 1.3-s activity was left unassigned. Although the 1.5-s and 1.3-s activities observed were probably indeed mostly $^{259}$Fm (produced by a $^{11}$Be transfer reaction), it is likely $^{262}$Rf also could have been present from the $^{248}$Cm($^{18}$O,4n) reaction and could not be distinguished due to its similarity in half-life to $^{259}$Fm.

A possible explanation for the observation of a 1.2-s SF activity is that there could be two activities for $^{262}$Rf, one being due to spontaneous fission from the ground state and the other due to spontaneous fission from an isomeric state, resulting in two half-lives
of 1.5 s and 50 ms. Such spontaneous fission from K-isomeric states has been predicted by Baran and Lojewski [11] to be possible for heavy nuclei. Therefore, the 1.2-s value by LLNL/Dubna and the 47-ms value by Somerville could both be correct, with the ~1.5-s activities observed by Hoffman et al. and Somerville et al. being a mixture of $^{259}$Fm and $^{262}$Rf.

To produce $^{262}_{104}$Rf, we used the $^{244}$Pu($^{22}$Ne,4n) reaction instead of the $^{248}$Cm($^{18}$O,4n) reaction because the production cross section for $^{256}$Fm and $^{259}$Fm from the former reaction should be much smaller [12], thus reducing the amount of interfering background from spontaneous fission activity. Our results for fission properties should then be indicative of the properties of $^{262}_{104}$Rf only. We also attempted to make a positive assignment of $^{262}_{104}$Rf by observing alpha correlations with $^{258}$No. Using Audi's recent atomic mass compilation [13] (which doesn't take into account the effects of the N=162 shell) and the alpha-decay systematics of Hatsukawa et al. [14], $^{262}_{104}$Rf should decay by alpha-emission with $E_\alpha=8.45\text{ MeV}$. However, when the effects of the N=162 shell [15] are included, $E_\alpha=8.26\text{ MeV}$ is predicted. In either case, we could make a positive identification of $^{262}_{104}$Rf and determine the alpha-branch by observing an alpha particle with energy between approximately 8.1 MeV and 8.6 MeV ($^{262}_{104}$Rf) followed within a few milliseconds by a spontaneous fission event from 1.2-ms $^{258}$No, the alpha daughter of $^{262}_{104}$Rf.

II. EXPERIMENTAL TECHNIQUES

The isotope $^{262}_{104}$Rf was produced at the Lawrence Berkeley Laboratory 88-Inch Cyclotron via the $^{244}$Pu($^{22}$Ne,4n) reaction. The target contained 765 $\mu$g/cm$^2$ $^{244}$Pu (98% isotopic purity) as the oxide deposited by the molecular plating method [16] in a 0.6-cm diameter circle on Be foil. The maximum cross section for the 4n reaction was calculated to be 1.5 nb at 112.7 MeV by using the evaporation code JORPLE [17]. The beam
current was 2.0 to 2.5 eμA. The beam passed through a 1.8-mg/cm² HAVAR entrance window, 0.3 mg/cm² N₂ cooling gas, and 2.59 mg/cm² Be target backing before entering the target material. Total energy losses were 22.6 MeV. The beam energy was chosen so as to result in a ²²Ne⁶⁺ energy of 114.4 MeV (laboratory system) in the center of the target. Beam energies of 109.8 MeV, 119.0 MeV, and 123.6 MeV in target were also used.

The reaction products recoil into the target chamber and are thermalized in helium at a pressure of 1.3 bar. The products are then attached to KCl aerosol contained in the helium gas and are swept out of the reaction chamber through a 1.4-mm or 1.6-mm i.d. teflon capillary tube to the vacuum chamber of our MG rotating wheel system [18] 7 m away. There are 80 collection sites along the periphery of the 20-inch diameter fiberglass wheel. Each collection site consists of a steel ring with a 0.63-mm diameter hole covered with 50±10 μg/cm² polypropylene foil. The transport and deposition efficiency was approximately 75%. The wheel was stepped at 1.5-s or 2.0-s intervals to position the foils between six pairs of passivated ion-implanted planar silicon (PIPS) detectors (100 mm² active area) which measure the kinetic energy of alpha particles and spontaneous fission fragments. The source-to-detector distance was about 2.0 mm, resulting in an efficiency in a given detector of 30% for alpha particles and 60% for fission fragments. The alpha particle energy resolution (FWHM) was about 40 keV in the top detectors and 60 keV in the bottom detectors; the latter being larger because of energy degradation in the polypropylene foil. The wheel is replaced with another wheel with clean foils every thirty minutes to minimize the build-up of long-lived activities.

Off-line alpha-energy calibrations were made by measuring the known alpha groups from ²¹²Bi and ²¹²Po in equilibrium with a ²¹²Pb source. On-line alpha-energy calibrations were made using the 7.27 MeV and 8.88 MeV alpha peaks of ²¹¹Po. Sources of ²⁵²Cf on 50±10 μg/cm² polypropylene foils were used for the energy calibration for the spontaneous fission fragments using the calibration method of Schmitt, Kiker, and Williams (SKW) [19] with constants determined by Weissenberger et al. [20]
Pulses from $\alpha$-particles between 5 and 10 MeV and fission fragments up to 200 MeV were digitized and stored in list mode, which stores time of event, channel number, and detector number. The timing requirement for coincident fission fragments in off-line sorting was about 2 $\mu$s. When searching for the alpha-decay of $^{252}$Rf, fission events within 10 ms of an alpha event with appropriate energy (7.5 to 9 MeV) were considered possible signatures of $\alpha$-SF coincidences between $\alpha$-decay of 2.1-s $^{252}$Rf and its 1.2-ms $^{258}$No daughter which decays by SF.

III. RESULTS

The $^{244}$Pu($^{22}$Ne,4n) reaction was run at 114.4 MeV at an average beam current of 2.0-2.5 $\mu$A for approximately 104 hours, at 109.8 MeV for 3.5 hours, at 119.0 MeV for 12 hours, and at 123.6 MeV for 12.5 hours. The production cross section was calculated to be $\sim$0.7 nb at 114.4 MeV. The production cross-sections at 109.8 MeV, 119.0 MeV, and 123.6 MeV were found to be $\sim$0.6 nb (based on 8 SF events), $\sim$0.5 nb (23 events), and $\sim$0.1 nb (6 events), respectively. The shape and magnitude of the resulting excitation function are consistent with those expected for the $^{244}$Pu($^{22}$Ne,4n) reaction, and therefore this activity has been assigned to the decay of $^{252}$Rf.

Due to the small number of detected SF events (200) at 114.4 MeV, the half-life for $^{252}$Rf was determined by performing a two-component decay curve fit to the SF activity using the maximum likelihood decay by the simplex method [MLDS] code [21]. From the fit, presented in Figure 1, the half-life is $2.1\pm0.2$ s. The stated error limits indicate the interval of equal-likelihood chances corresponding to a confidence level of 68%. Long-lived background SF activity was determined to be less than 2% of the total SF activity by counting the wheels with the foils off-line after the experiments.

The kinetic energies of 200 pairs of coincident fission fragments were measured in detector stations 1 through 4 or 1 through 3 (corresponding to six seconds since the end
of collection for stepping times of 1.5 s and 2 s, respectively). The pre-neutron TKE distribution is shown in Figure 2. The measured post-neutron-emission fragment kinetic energies and derived masses were corrected to pre-neutron-emission values using a saw-toothed \( \bar{v}(M) \) distribution similar to that measured for \(^{252}\text{Cf} \) [22] and \(^{256}\text{Fm} \) [23] and used by Balagna et al. [24] for \(^{257}\text{Fm} \). The average number of neutrons emitted per fission, \( \bar{v}_T \), was normalized to 4.4, a value estimated for \(^{262}\text{Rf} \) from a plot of \( \bar{v}_T \) vs mass number [1]. The best Gaussian fit to the TKE distribution gives a most probable pre-neutron-emission TKE of 215±2 MeV with a full width at half maximum (FWHM) of 50 MeV. The pre-neutron-emission kinetic-energy distributions for the high- and low-energy fragments from SF of \(^{262}\text{Rf} \) are shown in Figure 3. A summary of the kinetic energy measurements for \(^{262}\text{Rf} \) and the \(^{252}\text{Cf} \) calibration standard measured in the same system is given in Table 1.

The pre-neutron-emission mass-yield distribution is shown in Figure 4. The mass-yield data are expressed as yield (%) per mass number with the fragment yield normalized to 200%. The distribution is symmetric and can be fit with a Lorentzian distribution, but not with a single Gaussian. The FWHM is 22 mass numbers.

The contour plot in Figure 5(c) shows the post-neutron-emission TKE 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. Contours labeled 1 through 6 represent 6 equal increments of 4 through 24 events, respectively.

IV. DISCUSSION

Our value of 2.1±0.2 s for the half-life of \(^{262}\text{Rf} \) is within the error limits of the recently reported value of 1.2\(^{+1.0}_{-0.5} \) s [4], but we believe our value is much more accurate because it is based on 200 SF events while their value was based on only six SF events.
The values of $1.3\pm0.1$ s and $1.5\pm0.2$ s obtained from $^{248}$Cm + $^{18}$O by Somerville et al. [10] and by Hoffman et al. [9], as stated earlier, are probably due mostly to $^{259}$Fm ($t_{1/2} = 1.5$ s) with some possible contribution from the 2.1-s $^{262}$Rf. Therefore, the 2.1-s and 47-ms activities for $^{262}$Rf could both be correct, one for the ground-state decay and one for an isomeric-state decay.

Baran and Lojewski [11] have performed calculations for the spontaneous fission half-lives of nuclei from K-isomeric states. They find that SF from 2-quasiparticle excited states can occur as well as SF from the ground state. It has been shown [26] that the even-even nuclei $^{250}$Fm and $^{254}$No have 2-quasiparticle isomeric states. However, no SF has been observed from the excited states because the half-life for gamma emission to the ground state is much shorter than the partial half-life for SF from the isomeric state. Baran and Lojewski state that when the lowest 2-quasiparticle configuration has a particularly high spin projection (such as $K^\pi=9^+$), gamma emission would be hindered. In such cases, particularly for the heavier nuclei ($Z\geq104$), spontaneous fission from the isomeric state can then become observable. Therefore, the 47-ms activity could be fission from the K-isomeric state of $^{262}$Rf while the 2.1-s activity is fission from the ground state. This would agree with the similar observation [27] of a shorter isomeric half-life in $^{256}$Fm, the only case where spontaneous fission from a K-isomer in the first well of the potential energy surface has been measured. The partial SF half-life of the $K^\pi=7^+$ isomeric level in $^{256}$Fm is $\sim0.8$ ms [27], while the partial SF half-life of the ground state is $\sim2.9$ hr [8].

The SF half-life for $^{262}$Rf and all the partial SF half-lives for even-even nuclides known as of mid-1995 are plotted vs. neutron number in Figure 6. As can be seen in the figure, the trend extrapolated from lighter even-even Rf ($Z=104$) isotopes would suggest a SF partial half-life of only tens of milliseconds. The observed 2.1-s half-life, then, is evidence for stabilization due to the $Z=108$ and $N=162$ deformed shells, just as the longer half-lives in the region around $Z=100$ and $N=152$ ($^{250}$Cf, $^{252}$Fm, and $^{254}$No) show the stabilizing effect of the $N=152$ subshell.
The mass-yield distribution for $^{262}_{104}\text{Rf}$ is shown in Figure 7 together with those known for other trans-berkelium isotopes. It shows a symmetric mass distribution which could not be fit with a single Gaussian, but is well represented by a Lorentzian. Up until 1971, only asymmetric mass division had been observed for spontaneous fission, resulting in a "double-humped" distribution. Then it was discovered [24] that $^{257}\text{Fm}$ had an enhanced yield of symmetric division. Since then, the isotopes $^{258}\text{Fm}$ [28], $^{259}\text{Fm}$ [9,29], and $^{260}\text{Md}$ [28], among others, have been shown to have very symmetric distributions. The explanation for this is that these nuclei are approaching a mass $(^{264}\text{Fm})$ that can fission into two shell-stabilized doubly magic $^{132}\text{Sn}$ fragments, resulting in a greatly enhanced symmetric yield. The mass-yield distribution of $^{256}_{104}\text{Rf}$ is somewhat asymmetric. The mass-yield distributions of $^{258}_{104}\text{Rf}$ and $^{260}_{104}\text{Rf}$ are broadly symmetric, which was attributed to the return of "liquid-drop" type fission and the disappearance of the second barrier in the fission process [30]. However, if this were the case, the distribution of $^{262}_{104}\text{Rf}$ should not be nearly as narrow as the mass distribution we have observed which seems to indicate the influence of shell effects.

The TKE distribution for $^{262}_{104}\text{Rf}$ is shown in Figure 8 together with those known for other trans-einsteinium isotopes. It appears that $^{262}_{104}\text{Rf}$ has only a single component for its distribution, unlike those for $^{258}\text{Fm}$, $^{259,260}\text{Md}$, and $^{258,262}\text{No}$, which have been decomposed into two Gaussian distributions [28,31], one centered around 200 MeV and the other centered around 235 MeV. This so-called "bimodal" fission [32,33] results from two fission paths, one leading to symmetric compact spherical shapes which results in a high TKE, and the other leading to symmetric elongated shapes which have a lower TKE. There is also the possibility of "multimodal" symmetric fission [34,35], when a variety of fission channels or pathways are possible for a fissioning nucleus on the same potential energy surface. The small number of events for $^{262}_{104}\text{Rf}$ prevent us from conclusively determining if it is monomodal, bimodal, or multimodal fission.

The contour plot for $^{262}_{104}\text{Rf}$ together with those for $^{256}\text{No}$ and $^{259}\text{Lr}$ are shown in Figure 5, giving a "3-D" picture of the relationship of mass fraction and pre-neutron TKE.
It can be seen that the yield is greatest for symmetric division and that, in all cases, the TKE increases as the division becomes more and more symmetric due to the fact that some symmetric fragments with near spherical shapes may be formed, but there is a large variance at symmetry. As can be seen, there is no evidence for the very high-TKE symmetric fission as observed for the bimodal isotopes $^{258}\text{Fm}$, $^{259,260}\text{Md}$, and $^{258,262}\text{No}$. However, because of the large variances of the TKE values for symmetric mass division, a variety of different modes resulting in deformed as well as spherical shapes possibly could be involved, indicating "multimodal" fission.

The most probable pre-neutron TKE is plotted in Figure 9 as a function of the Coulomb parameter, $Z^2/A^{1/3}$. All nuclei follow a linear trend except the heavy Fm, Md, and No isotopes. These "abnormally" high TKEs can be explained by noting that these isotopes are the ones that have a sharply symmetric mass division into nearly doubly magic fragments. Therefore, the resulting compact spherical fragments will have higher TKEs than the elongated deformed fragments of other nuclei. The value of 215 MeV plotted for $^{262}\text{Rf}$ is shown to follow the linear fit by Unik [37], but is high relative to the fit by Viola [36].

During our experiment, we searched for $\alpha$-decay of $^{262}\text{Rf}$ to 1.2-ms $^{258}\text{No}$ (SF), but observed no such time-correlated $\alpha$-SF events and have assigned an upper limit of 0.8% (68% confidence level) for an alpha-decay branch of $^{262}\text{Rf}$. This corresponds to a partial alpha half-life of greater than 260 s. Audi's mass compilation [13] results in a calculated $E_\alpha$ of 8.45 MeV and a resulting decay branch of 18% [14], much higher than our present limit. However, this mass does not include any effect of the N=162 shell. If a mass is used that takes this shell into account [15], an $E_\alpha$ of 8.26 MeV is obtained, and a partial half-life of 43 s is expected [14], resulting in an alpha branch of 4.9%. Our result, therefore, is not only a strong indication of the existence of shells at $Z=108$ and $N=162$, but also shows that they have an even greater effect than currently suggested. More data on SF and alpha decay for nuclides with $Z\geq 104$ will be needed to better quantify the effect of these deformed subshells.
ACKNOWLEDGMENTS

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REFERENCES


TABLE CAPTION

1. Properties of the measured (post-neutron-emission) and calculated initial (pre-neutron emission) fragment kinetic-energy distributions for $^{262}_{104}$Rf and the $^{252}$Cf standard measured in the same system. Energies are given in MeV, based on the SKW calibration method [19] with the Weissenberger constants [20].
<table>
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<th>(^{262}\text{Rf})</th>
<th>(^{252}\text{Cf})</th>
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<td></td>
<td>Pre-n</td>
<td>Post-n</td>
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<tr>
<td><strong>Total kinetic energy</strong></td>
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<td></td>
</tr>
<tr>
<td>Average</td>
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<td>208.3</td>
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<tr>
<td>Most probable(^a)</td>
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<td>211.5</td>
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<td>(\sigma)</td>
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<tr>
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<tr>
<td>FWHM(^b)</td>
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<td>22.6</td>
</tr>
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</table>

\(^a\)Standard deviation of the most probable values from the Gaussian fits is about 0.9%.

\(^b\)Full width at half maximum, calculated from 2.35\(\sigma\) for Gaussian fit to the top half of the peak.
FIGURE CAPTIONS

1. Decay curve of the $^{262}_{104}$Rf SF coincidences. 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 encompass 68% of the probability in a Poisson distribution centered on the number of counts expected during the interval, obtained from the most probable fit by the simplex method [21].

2. Gaussian fit to the pre-neutron-emission TKE distribution from the SF of $^{262}_{104}$Rf. The data are in groupings of 10 MeV.

3. Gaussian fits to pre-neutron-emission distributions for high and low kinetic-energy fragments from SF of $^{262}_{104}$Rf.

4. Pre-neutron-emission mass-yield distribution for $^{262}_{104}$Rf (200 events). The pre-neutron-emission TKE was derived from the SF coincidence data using a $\bar{v}(M)$ function similar to that used by Balagna et al. [24] for $^{257}$Fm, with $\bar{v}_r=4.4$. The data are in groupings of 5 mass numbers. The curve is a Lorentzian fit. The bars indicate 1σ error limits. Also shown (open circles, dotted line) is the provisional mass-yield curve to which no neutron correction was applied.

5. Contour plots of pre-neutron-emission TKE vs. mass fraction. The connected points represent average TKE as a function of mass fraction. a) $^{256}$No (346 SFs). The contours indicate equal numbers of events based on data groupings 20 MeV X 0.04 units of mass fraction. Contours labeled 1 through 6 represent 10 through 60 events, respectively. b) $^{259}$Lr (442 SFs). The contours indicate equal numbers of events based on data groupings 20 MeV X 0.02 units of mass fraction. Contours labeled 1 through 5 represent 10 through 50 events, respectively. c) $^{262}_{104}$Rf (200 SFs). 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 6 represent 4 through 24 events, respectively. (From Ref. [25])

6. Logarithms of partial SF half-lives of even-even nuclei plotted vs. neutron number. Arrows are used to indicate lower limits. (From Ref. [25])

7. Schematic representation of all known mass-yield distributions (normalized to 200% fission fragment yield) for SF of trans-Bk isotopes. (From Ref. [25] and this work)

8. TKE distributions for SF of some trans-Es isotopes. (From Ref. [25] and this work)

9. Average or most probable TKE vs. $Z^2/A^{1/3}$. The solid line is the linear fit of Viola et al. [36] and the dashed line is the linear fit of Unik et al. [37]. The superscripts "h" and "l" represent the high and low energy components of the bimodal nuclei. (From Ref. [25])