A NEW SPONTANEOUS FISSION MODE FOR ^{252}Cf: HYPERDEFORMATION, CLUSTER RADIOACTIVITY, NEW LEVELS

J. H. Hamilton[1,2], G. M. Ter-Akopian[1,3], Yu. Ts. Oganessian[3], A. V. Daniel[1,3], J. Kormicki[1,*], G. S. Popeko[3], A. V. Ramayya[1], Q.-H. Lu[1], K. Butler-Moore[1**], W.-C. Ma[1†], S. Ćwiok[4,5], W. Nazarewicz[6], W. Greiner[1,2,7], A. Sandulescu[1,2,7,8], J. K. Deng[1‡], D. Shi[1], J. Kliman[9], M. Morhác[9], J. D. Cole[10], R. Aryaeinejad[10], S. Zhu[1], R. Babu[1], N. R. Johnson[11], I. Lee[11+], F. K. McGowan[11], J. X. Saladin[5]

[1]Physics Department, Vanderbilt University, Nashville, TN 37235 USA.
[2]Institute for Theoretical Physics, J.W. Goethe University, D-660054 Frankfurt, Germany.
[3]Joint Institute for Nuclear Research, Dubna, 141989 Russia.
[4]Warsaw University of Technology, PL-00662 Warsaw, Poland.
[5]Physics Department, University of Pittsburgh, Pittsburgh, PA 15260 USA.
[6]Physics Department, University of Tennessee, Knoxville, TN 37996 USA.
[7]Joint Institute for Heavy Ion Research, Oak Ridge, TN 37831 USA.
[8]Institute for Atomic Physics, Bucharest, P.O. Box MG-6, Romania
[9]Institute of Physics SASc, Bratislava, Slovak Republic.
[10]Idaho National Engineering Laboratory, Idaho Falls, ID 83415 USA.
[11]Oak Ridge National Laboratory, Oak Ridge, TN 37831 USA.

Abstract

Direct measurements of yields and neutron multiplicities were made for Sr-Nd, Zr-Ce, Mo-Ba, Ru-Xe, and Pd-Te from γ-ray coincidence studies in spontaneous fission of ^{252}Cf. Strong enhancement of the 7-10 ν emission channels is seen in the Mo-Ba data. Unfolding the Mo-Ba data revealed a new fission mode associated with the enhanced ν yields with much lower total kinetic energy going by ^{108}Mo-^{144}Ba, ^{107}Mo-^{145}Ba, and/or ^{106}Mo-^{146}Ba . Analysis indicates one or more of ^{144,145,146}Ba are hyperdeformed with 3:1 axis ratio. Theoretical calculations predict a third minimum in the PES for ^{252}Cf with $\beta_2 \sim 0.9$ and $\beta_3 \sim 0.7$. Zero neutron emission channels , a new form of cluster radioactivity, are seen in 8 and 7 correlated pairs in SF of ^{252}Cf and ^{242}Pu, respectively, with the odd-odd zero neutron channel yields strongly enhanced as predicted for cluster radioactivity. New level structures and isotopes include new octupole deformations, identical bands and other structures.

A NEW SPONTANEOUS FISSION MODE AND HYPERDEFORMATION

Recently we reported(1) the first direct measurements of yields and neutron multiplicities connected with these yields of correlated fragment pairs of Zr-Ce and Mo-Ba in the spontaneous fission (SF) of ^{252}Cf. The Mo-Ba neutron multiplicities extended from 0 to 10 neutron emission. It was noted(1) that the 0 and 8-10 neutron multiplicities considerably enhanced compared to those for gross (total) neutron multiplicities(2). The

enhancement at high neutron multiplicity could be an indication of a second fission mode. Two or more fission modes for disintegration of the same nucleus theoretically predicted[3-6] are now known for a number of heavy nuclei[7,8].

To help develop an understanding of the enhancements observed at large neutron multiplicities for the Mo-Ba pairs, we have extracted more complete sets of experimental yields and neutron multiplicities in the SF of ^{252}Cf including the odd-A nuclei for the Zr-Ce (40/58) and Mo-Ba (42/56) correlated pairs and for the next three strongest pairs of Sr-Nd (38/60), Ru-Xe (44/54), and Pd-Te (46/52)[9]. From these data we find that there are two fission modes in the breakup into Mo-Ba: first, the normal mode with large total kinetic energy (TKE) and broad mass distribution of primary fragments, and second a mode in which the TKE is much lower and the mass distribution is limited to one or more of three pairs, ^{108}Mo-^{144}Ba, ^{107}Mo-^{145}Ba, ^{106}Mo-^{146}Ba where at scission at least one or all of $^{144,145,146}Ba$ are hyperdeformed![9]

The experimental arrangement and data analysis procedures were described in our earlier paper[1]. A ^{252}Cf source was placed at the center of the 20 Compton-suppressed Spectrometer System at the Holifield Heavy Ion Research Facility. The neutron multiplicity distributions extracted for all five pairs are shown in Fig. 1[9]. The curves in Fig. 1 are Gaussian fits to the peaks of the distributions. Single Gaussian curves fit the full range of all the distributions except that of the Mo-Ba correlated pairs. Note that only the Mo-Ba pairs show a strong enhancement for the 7-10 neutron emission yields. We emphasize again that these data are the first direct measurements of such yields and neutron multiplicities.

The yields of correlated fragment pairs and neutron multiplicities shown in Fig. 1 originate as a result of the de-excitation processes of primary fission fragments. They carry information about the mass and excitation energy distributions of the primary fission fragments of corresponding fixed charge splits $Y(A_L, E_L^*, A_H, E_H^* | Z_L, Z_H)$. Yields of the fission fragment pairs $Y_i(A_L', A_H')$, created after the neutron evaporation, are directly connected with the yields of the primary fission fragment pairs $Y_j(A_L, A_H)$, with the excitation energy distribution of each primary fission fragment $F(E^*, A)$, and with the probability $P_n(E^*, A)$ of evaporation of n neutrons from the primary fragment where $n = A_j - A_i'$. Here $A_{L,H}'$ denote the masses of the fragments obtained after the completion of the de-excitation process of primary fragments. The extensive Mo-Ba data allowed an unfolding of the data to extract the distribution $Y(A, E_L^*, A_H, E_H^* | Z_L, Z_H)$.

First, a least square best fit of the calculated yields $Y_i^{calc}((A_L', A_H')$ to the pattern of the yields $Y_i(A_L', A_H')$ was searched by assuming Gaussian forms for the mass and excitation-energy-distributions of primary fission fragments. De-excitations of the primary fission fragments were calculated by employing the statistical code GNASH[10]. If one assumes that only a single Gaussian distribution approximates the primary fragment mass probability and that the excitation energy distribution of each mass fragment is in the neighborhood of a single Gaussian curve, no satisfactory fit can be obtained for the Mo-Ba data. However, a good fit to the data is obtained when we assume that <u>two distinct fission modes</u> contribute to the formation of the primary Mo-Ba fission fragments. In fact, good fits to the data of Table 1 were obtained by searching the parameters <TKE>, σ_{TKE}, $\bar{A}_H \sigma_{A_H}$ and $\bar{E}^*_{H_j}$ for the first fission mode assuming that only one Mo-Ba primary fragment pair contributes to the second fission mode. Very reasonable results were obtained by assuming that the single primary fragment pair responsible for the second mode is ^{108}Mo-^{144}Ba, ^{107}Mo-^{145}Ba, or ^{146}Mo-^{146}Ba. For each of these primary fragment paris, essentially the same value of <TKE> = 153 ± 3 was found for the second fission mode as a result of the unfolding procedure. For the first fission mode <TKE> = 189 ± 1 MeV followed independently of which of the above three pairs contribute to the second mode. Other parameters of the first mode also did not depend on the choice of the pair contributing to the second mode.

The ^{106}Mo-^{146}Ba pair give the best fit to the 7-10 neutron multiplicities (see Fig. 2). However, the experimental data do not allow us to say unambiguously which of the above three fragment pairs or their combinations provide for the existence of the second mode. However, the very fact of the manifestation of two distinct fission modes in the case of the Mo-Ba split of ^{252}Cf is established unambiguously. Mode 1 looks like a familiar fission mode of ^{252}Cf in the respect that its principal characteristics (<TKE>, σ_{TKE}, $\bar{A}_H$, $\bar{A}_L$) and excitation energies of the primary fragments are close to those that were known before[11]. The Mode 2 appears quite peculiar because of its low TKE value and low Coulomb barrier. The low TKE value suggests an enormously large elongation of the ^{252}Cf nucleus at its scission with a very high deformation of the fragments that emerge after scission.

From their large excitation energies, the barium fragments are those that are highly deformed when the Mode 2 occurs. From their excitation energies, one can estimate the ratios of the axes at scission: a/b = 2.8, 3.0 and 3.2 for ^{144}Ba, ^{145}Ba and ^{146}Ba, respectively. So, it is very probable that at scission, in Mode 2, the barium fragment or fragments are hyperdeformed with axis ratio of mostly likely 3:1! In the case when the pair ^{106}Mo-^{146}Ba emerges at Mode 2, ^{106}Mo has about the same excitation as is the case for the first mode, and thus a "normal" deformation. The excitation energy of the Mo fragment increases considerably for the ^{107}Mo-^{145}Ba pair and becomes comparable with that of the Ba fragment if the ^{108}Mo-^{144}Ba pair occurs in the Mode 2.

It is pŏssible that some reasonable modifications of the hypotheses underling our unfolding procedure could alter somewhat the numerical results. However, we believe that the yields and multiplicity data of Mo-Ba necessarily lead to the conclusion that there are two distinct modes occurring in the Mo-Ba fission of the ^{252}Cf nuclei, and that apparently Mode 2 is not present in the other fragment pairs as seen in Fig. 1. Quite independent of the details, one can make the following general observations about the fragments: (1) in the second mode the Ba fragments are left in an unusually highly excited state from which 5-8 neutrons are evaporated, (2) the fragments have unusually low TKE and so unusually low Coulomb energy at scission which means the charge centers are much further apart, features consistent with hyperdeformation in the Ba fragment or fragments.

Very recently Ćwiok et al.[12] considered the question of hyperdeformation and clustering in actinide nuclei. In that work systematic calculations of potential energy surfaces of the even-even Rn, Ra, Th and U isotopes were performed by using the shell-correction approach with the axially-deformed average Woods-Saxon potential. Their calculations yielded third minima in the PES, characterized by large elongations, $\beta_2 \sim 0.9$ and significant reflection asymmetry, $0.35 < \beta_3 \leq 0.65$, in addition to those of the ground states ($\beta_2 \sim 0.25$) and fission isomers ($\beta_2 \sim 0.6$) in several actinide nuclei. Rutz et al.[13] have recently investigated ^{240}Pu, ^{232}Th and ^{226}Ra in a relativistic mean-field model and likewise found a third minimum for ^{232}Rh at a hyperdeformed shape. Ćwiok et al.[12] suggested that the mass distribution of fission fragments should be greatly influenced by the structure of the HD minimum and the third saddle point. They also note that the observation of HD states in the actinides constitute an important confirmation of the shell structure of the nuclei. The unusual stability of these HD states is attributed to strong shell effects that are present in the average nuclear potential at the 3:1 shape.

In the same way[12] calculations were carried out for ^{252}Cf. The PES for ^{252}Cf is shown in Fig. 3 where β_2 extends between the second minimum and the outer barrier. There is a well-developed third minimum at $\beta_2 \sim 0.9$ and $\beta_3 \sim 0.7$ with the paths to scission shown. The static path for HD_{II} is shown in Fig. 4 along with the strongly reflection asymmetric shape of ^{252}Cf in the third minimum. In the upper right-hand corner is the calculated nuclear shape just beyond point B where R is ^{146}Ba. It would be fascinating to see directly the HD minimum in ^{252}Cf. It is possible that the HD, extracted for one or more ^{144,145,146}Ba fragments comes through the decay of this third minimum.

In conclusion, the observed coexistence of two fission modes in the spontaneous fission of ^{252}Cf involves a new type of bi-model fission. For the Mo-Ba division of ^{252}Cf, one and the same fragments, ^{144}Ba, ^{145}Ba and/or ^{146}Ba, appear either with a "standard" or enormously high excitation energy in the first or second fission modes, respectively. This implies that ^{144}Ba, ^{145}Ba and/or ^{146}Ba are found in two states which are remarkable for their very different deformations at scission. Their partners, ^{107,106}Mo, have approximately the same deformation at both fission modes while ^{108}Mo has a deformation approaching that of ^{144}Ba. The first "normal" fission mode bears features typical of the bulk of fission events of ^{252}Cf, whereas the second abnormal mode revealed here for the first time provides evidence for a hyperdeformed state or states in ^{144,145,146}Ba with $\approx$3:1 axis ratio. A third deep HD minimum in the PES for ^{252}Cf could be the origin of the HD Ba-Mo split.

ZERO NEUTRON EMISSION – A FORM OF CLUSTER RADIOACTIVITY

The experimental discoveries of new spontaneous decays with emission of clusters like ^{14}C, ^{20}O, ^{23}F, ^{24}Ne, ^{28}Mg and $^{32\text{-}34}$Se (cluster radioactivity) have confirmed the theoretical predictions based on the idea that there are cold rearrangements of a large number of nucleons from one ground state of the A-nucleon system to two ground states of the A_1 and $A_2(A = A_1 + A_2)$ nucleon systems[14]. These observed new superasymmetric decay modes, intermediate between alpha decay and symmetric fission, can be related to the spherical double magic nature of $^{208}_{82}Pb_{126}$ and its neighbors which causes a deep valley in the mass and charge asymmetry potential.

Cold fragmentation in spontaneous fission or in thermal-neutron induced fission, i.e. where the fragments with maximum kinetic energies close to the corresponding Q values or Q plus the binding energy of the neutron is difficult to observe because of the large background of the hot fission fragments. Fission fragment spectrometers which separated masses and charge with much higher resolving power but low efficiency allowed searches for only the highest TKE for fragments close to the ground state (refs. in 9). With a (practically 4π) twin ionization chamber masses and charges for were detected for cold events at all fragmentations[15,16]. It has been shown[17] that the above experimental data are in complete agreement with a description of cold fission as a type of cluster radioactivity. Unfortunately, in all the above experiments the final nuclei with their corresponding energy spectra are not measured directly, although recently A and Z, but not level populations, were observed. Consequently, a simple interpretation of cold fission as a decay process[17] met strong opposition.

In our SF work, four zero neutron emission channels were observed in $^{104}_{42}$Mo-$^{148}_{56}$Ba, $^{106}_{42}$Mo-$^{146}_{56}$Ba, $^{108}_{42}$Mo-$^{144}_{56}$ and $^{104}_{40}$Zr-$^{148}_{58}$Ce, by setting a gate on a known transition in each partner and looking at the gamma-rays in coincidence with this double gate and by gating one gamma-ray in one partner (Z_1) and looking at the gamma-rays associated with the partners (Z_2 = 98 - Z_1)[18]. The excited energy levels observed in these partner nuclei are shown in Fig. 4[18]. The single neutron binding energies are given below each nucleus. Since zero neutrons are emitted, the fragments must be left after fission with the order of or less than the neutron binding energies which go from 4.7 to 7.7 MeV. The maximum excited state energies observed in these nuclei go from 2304 up to 4683 keV[19]. Only a small fraction of the large Q-value goes into the excitation of the low-lying excited states. Practically the whole Q-values go into TKE's of the fragments. So, some cold fission can be completely separated from the normal fission with elongated fragments. Thus, the spontaneous emission of large clusters is completely analogous to the spontaneous emission of smaller clusters such as ^{14}C, ^{20}O, and ^{32}Si in cluster radioactivity[14]. Now, however, one sees fine structure in both fragments--not just in one fragment as previously.

From previous experimental studies of cold fission, we know that the TKE for odd-odd fragmentation have yields much closer to the corresponding Q-value than for the even-even fragmentation[20]. For even-Z-even-Z the odd-A-odd-A cluster radioactivity model predicts the yields will be greater than the even-even yields. Since our first report[18] of the zero neutron channel as a new form of cluster radioactivity, we have identified two new zero neutron channels, ^{105}Mo-^{147}Ba and ^{107}Mo-^{145}Ba. Indeed, these channels have yields about three to four times larger than the even-even Mo-Ba zero neutron channels. These new results strongly support the cluster radioactivity model. In recent Gammasphere work, the zero neutron channels ^{142}Xe-^{100}Zr, ^{140}Xe-^{102}Zr, ^{138}Xe-^{104}Zr, ^{140}Te-^{102}Mo, ^{138}Te-^{104}Mo, ^{136}Te-^{106}Mo, and ^{132}Sn-^{110}Ru have been observed in SF of ^{242}Pu.[21] Zero neutron channels have been observed in the ^{248}Cm work at Eurogam[22]. Calculations based on the cluster radioactivity model also predict the relative spin populations in cluster radioactivity of ^{252}Cf which are in good agreement with the experimental data.

NEW ISOTOPES AND LEVEL STRUCTURES

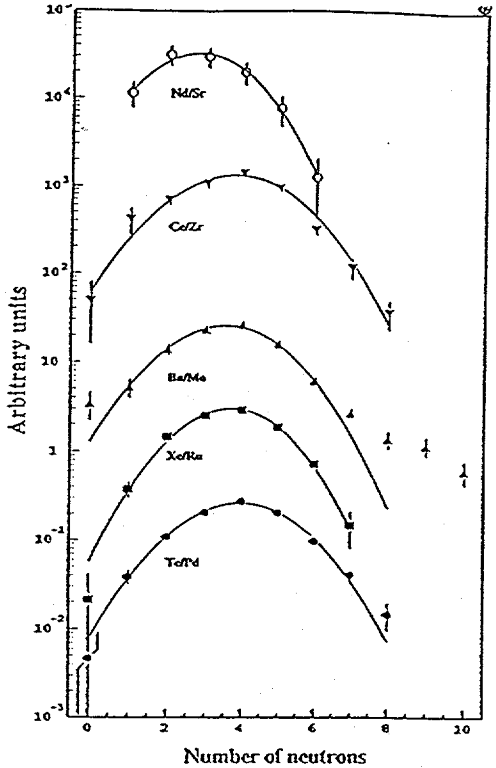
The neutron-rich nuclei populated in SF are a rich source of new insights into nuclear structure.[23] New high spin states to 20^+ and 19^- and new isotopes like ^{152}Ce and ^{160}Sm are observed.[23] Some of the new data are shown in Fig. 5. These include new examples of octupole deformation in ^{142,143}Ba and ^{144}Ce as predicted by theory, new γ-type vibrational bands in Ru and Pd nuclei, and new identical ground bands in $^{144,146}Ba$ (and octupole bands as well), $^{152-156}Nd$, $^{98,100}Sr$, and $^{108,110}Ru$.

ACKNOWLEDGEMENT

The support of the Alexander von Humboldt Foundation for one of us (JHH) is much appreciated. Two of us (G.M. T.-A and A.V. D.) wish to express appreciation for the hospitality and financial support received during the their stays at Vanderbilt University and Oak Ridge National Lab. Work at Vanderbilt University and Idaho National Engineering Lab. is supported in part by the U.S. Dept. of Energy under grant and contract No. DE-FG05-88ER40407 and DE-AC07-76ID01570, respectively. Work at the Joint Institute for Nuclear Research is supported in part by grant No. 94-02-05584-a of the Russian Federal Foundation of Basic Sciences. Work at the Institute of Physics of SASc is supported in part by the Grant Agency of the SASc under grant GA-SAV517/1993. Oak Ridge National Laboratory is managed by Martin Marietta Energy Systems, Inc., under contract No. DE-AC05-84OR21400 for the U.S. DOE. The Joint Institute for Heavy Ion Research has as member institutions the University of Tennessee, Vanderbilt University, and Oak Ridge National Lab.; it is supported by the members and by U.S. DOE through contract No. DE-FG05-87ER40361 with the U. of Tennessee.
[*]Also UNISOR, ORISE, Oak Ridge. Currently: [**]Idaho National Engineering Lab.; [†]Mississippi State University; [‡]Tsinghua University, Beijing; [+]Lawrence Berkeley Lab.

1. G. M. Ter-Akopian, et al., Phys. Rev. Lett. **73** (1994) 1477.
2. J. Wild, et al., Phys. Rev. **C41** (1990) 640.
3. V. V. Pashkevich, Nucl. Phys. **A161** (1971) 275; **A477** (1988) 1.
4. U. Brosa, S. Grossman, and A. Müller, Z. Nat. **41a** (1986) 1341.
5. P. Möller, J. R. Nix, and W. J. Swiatecki, Nucl. Phys. **A492** (1989) 349.
6. U. Brosa, S. Grossman, and A. Müller, Phys. Rep. **197** (1990) 167.
7. M. G. Itkis, et al., Z. Phys. **A320** (1985) 433.
8. E. K. Hulet, et al., Phys. Rev. Lett. **56** (1986) 313.
9. G. M. Ter-Akopian, et al., Phys. Rev. Lett. (1995), submitted.
10. A. C. Wall, At. Data Nucl. Data Tables **39** (1988) 1.
11. P. G. Yound, et al., *Workshop on Computation and Analysis of Nuclear Data Relevant to Nuclear Energy and Safety* (Trieste, Italy, 1992).
12. S. Ćwiok, et al., Phys. Lett. **B322**, 304 (1994).
13. K. Rutz, J. A. Maruhn, P.-G. Reinhard, and W. Greiner (1995), to be published.
14. A. Sandulescu and W. Greiner, Rep. Prog. Phys. **55** (1992) 1423.
15. C. Signarbieux, et al., J. Physique Lett. **42** (1981) L437.
16. J. Trochon, et al., Rad. Eff. **92** (1986) 327.
17. K. Depta, et al., J. Phys. G: Nucl. Phys. **14**(1988) 891; A. Sandulescu and W. Greiner, *ibid.* **15** (1989) 1815; A. Floescu, et al., *ibid.* **19** (1993) 669.
18. J. H. Hamilton, et al., J. Phys. G: Nucl. Part. Phys. Lett. **20** (1994) L85.
19. S. J. Zhu, et al., (1994), to be published.
20. G. Simon, These pour le grae de Docteur d'Etat, Universite de Paris-Sud, (1990).
21. Y. K. Dardenne, et al., (1995), to be published.
22. J. L. Durell, Proc. Physics from Large γ-Ray Detector Arrays (Berkeley, CA), Vol. 1, (1994) p. 107 .
23. J. H. Hamilton, et al., in *Progress in Particle and Nuclear Physics*, A. Faessler, ed., Pergamon Press (1995).

Fig. 1.



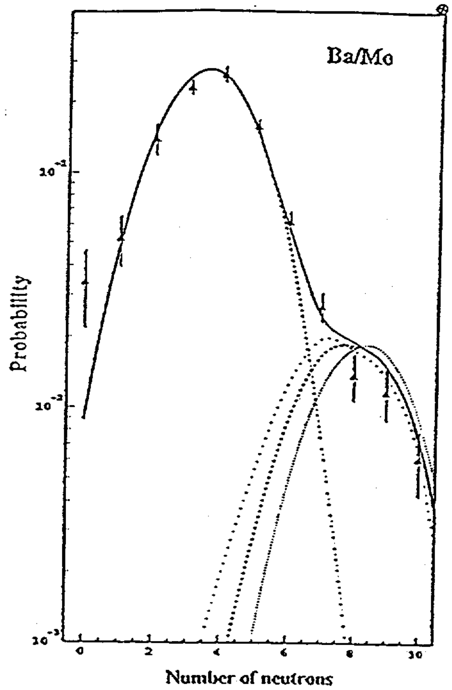


Fig. 2. Two Gaussian fits to the Mo-Ba neutron multiplicity data. The three curves shown under the 7-10 neutron multiplicity data are the fits when ^{106}Mo-^{146}Ba (farthest left curve), ^{107}Mo-^{145}Ba (middle curve) or ^{108}Mo-^{144}Ba (right curve) pair is the single mode responsible for the high multiplicity data.

Fig. 3.

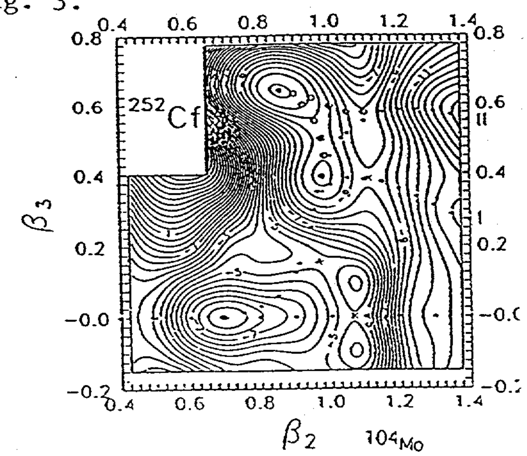
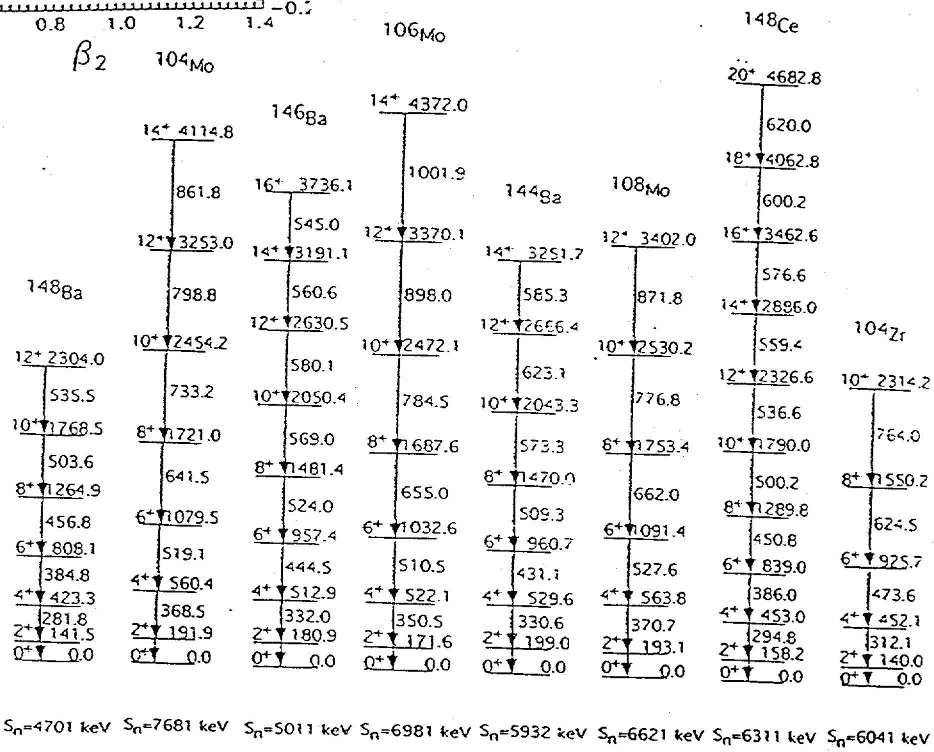


Fig. 4.



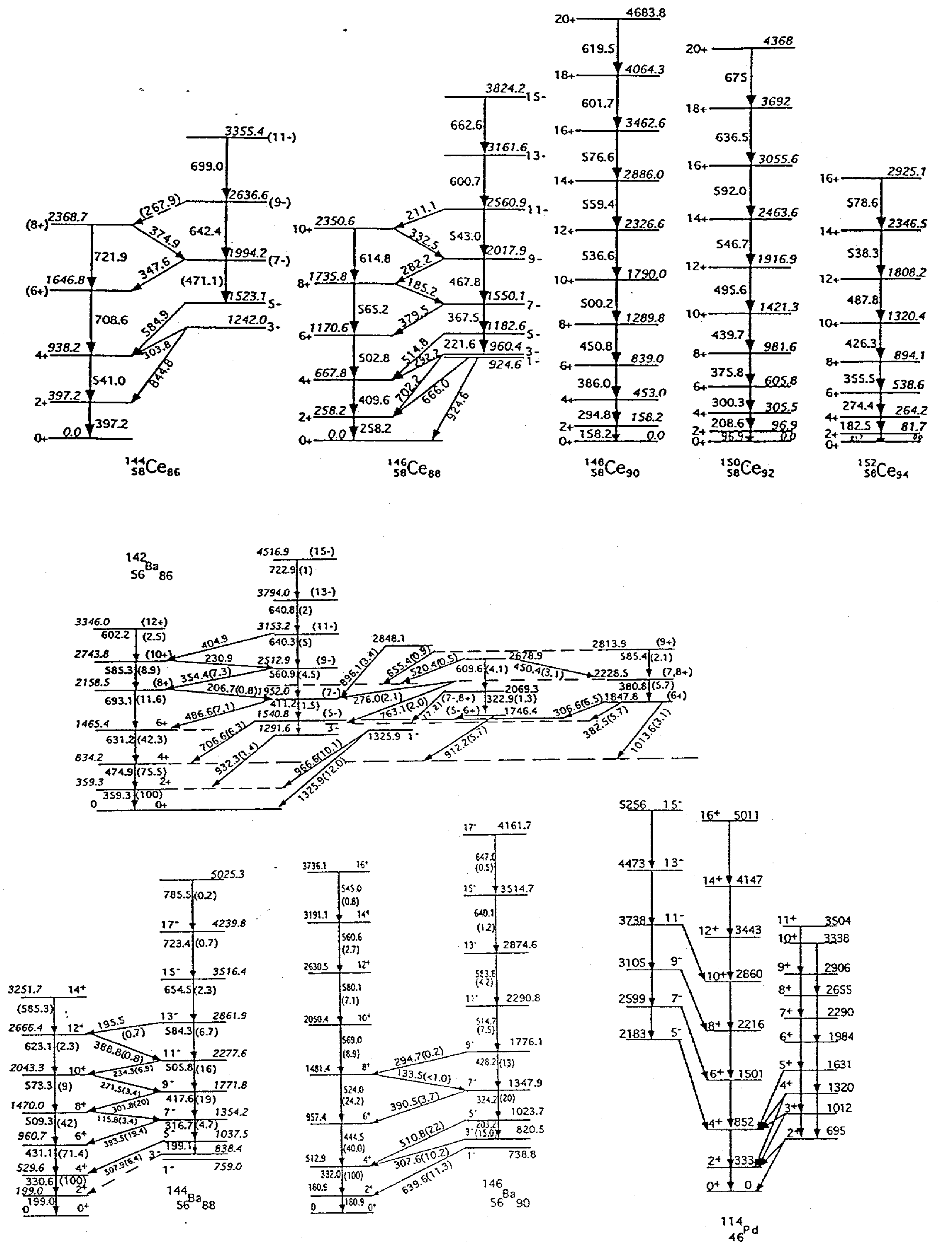


Fig. 5. Examples of new high spin levels to 20^{+} and 19^{-}, new γ-type vibrational bands (2^{+}-11^{+}), and new isotope ^{152}Ce.