Mapping low-energy fission by beta-delayed fission: from neutron-deficient to neutron-rich nuclei



LASD INT Prorgram, 14 October 2013

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Outlook

- Brief (experimental) review on low-energy fission
- Low-energy fission in "new" regions of the Nuclear Chart
- Beta Delayed Fission (β DF) what it is and why?
- β DF ^{194,196}At, ²⁰²Fr at ISOLDE (CERN)
- Further plans

Outlook

•Many nuclear properties change far from stability line (e.g. disappearance of traditional magic numbers; appearance of new shell gaps; halos, skins...

•What happens to fission far from stability, e.g. on the extremely proton-rich or neutron-rich side (relevant for r-process)?

•Not simple to answer, as to fission these nuclei at low excitation energy ($E^* \sim B_f$) is a very challenging task as none of them fissions from g.s.

Fission Barrier Calculations for the r-process nuclei

Full symbols – experimental data Lines – calculations (LDM,TF, ETFSI)



• Good agreement between $B_{f,cal}$ and $B_{f,exp}$ for nuclei close to stability

- Large disagreement far of stability (both on n-def. and n-rich sides)
- Need measured fission data far of stability to 'tune' fission models

A Detour:

What can one learn with a rate of 1 fission/h?

OR

What can one learn from ~100-1000 fission events?



Bimodal Fission

E. K. Hulet et, Phys. Rev. C40, 770 (1989)



Evidence for bimodal fission: strong deviation of TKE from a single Gaussian Detailed fission fragments energy measurements are "A MUST"

Experimental information on low-energy fission Nuclei with measured charge/mass split (RIPL-2 + GSI)



K.-H. Schmidt et al.

Experimental information on low-energy fission Nuclei with measured charge/mass split (RIPL-2 + GSI) Region of our interest II: beta-**Heavy Actini** symmetric; delayed fission of nuclei with aneous Region of our interest I: beta-A~230, N/Z~1.6: Fr, Pa. Ac ²⁵⁸Fm delayed fission of A~180-200 N/Z~1.22-1.3: TI,Bi, At, Fr Lr No 8 **ISOLDE(CERN)** 230 j 0 0 000 ²⁵⁶Fm ²⁰⁹Ra Cm Am Pa 236[] Ro Ac ²²⁷Ra Z=82 ²¹³At ¹⁸⁰Hg - particle induced N/Z=1.25 x - e.m. –induced E*~11 MeV 187**|r** ¹⁹⁶Au 126 Pre-actinides, light Ir-Th N/Z~1.4-1.5: predominantly symmetric, e.g. FRS(GSI)

Beta-Delayed Fission

Discovery: ^{232,234}Am (1966, Dubna)

βDF branch

N_{βDF} N_β



•Two step process: β decay followed by fission

•Low-energy fission (E*~3-12 MeV, limited by Q_{EC}) e.g. ¹⁸⁰TI: Q_{EC} =10.4 MeV, $B_{f,calc}$ =9.8 MeV

•Relatively low angular momentum of the state e.g. ¹⁸⁰TI: I=4 or 5 (some cases: up to 10)

Three regions to search for βDF

Necessary conditions for β DF to occur:

- $Q_{FC}(Parent) \sim B_f(Daughter)$ [$Q_{FC} B_f > -2 MeV$]
- Beta-branching ratio b_β>0



P. Moller

Beta-Delayed Fission Discovery: ^{232,234}Am (1966, Dubna)



$P_{\beta DF}$ Probability: Extraction of fission barriers?!

S_β!?

$$P_{\beta df} = \frac{\int_{0}^{Q_{\beta}} F(Q_{\beta} - E) S_{\beta}(E) \frac{\Gamma_{f}(E)}{\Gamma_{f}(E) + \Gamma_{\gamma}(E)} dE}{\int_{0}^{Q_{\beta}} F(Q_{\beta} - E) S_{\beta}(E) dE}$$
 Need to know (next slide)

$$\begin{split} & \Gamma_{f} \\ \hline \Gamma_{tot} = \frac{\Gamma_{f}}{\Gamma_{f+}\Gamma_{\gamma}} & \text{-ratio of the fission and total widths of excited levels in daughter} \\ & \Gamma_{r} = \frac{\Phi_{f+}\Gamma_{\gamma}}{2\pi\rho} & (\Gamma_{n} \text{ is not important for neutron-deficient nuclei}) \\ & \Gamma_{\gamma} = \frac{\Phi_{f+}\Gamma_{\gamma}}{2\pi\rho} & (\Gamma_{r}) + \frac{\Phi_{f+}\Phi_{f+}}{2\pi\rho} & (\Gamma_{r}) + \frac{\Phi_{f+}\Phi_{f+}}{2\pi\rho} & (\Gamma_{r}) + \frac{\Phi_{f+}\Phi_{f+}}{2\pi\rho} & (\Gamma_{r}) + \frac{\Phi_{f+}\Phi_{f+}}{2\pi\rho} & (\Gamma_{r}) + \frac{\Phi_{r}}{2\pi\rho} & (\Gamma$$

Measurement of $P_{\beta DF}$ allows to deduce Fission Barrier B_f

e.g. H.V. Klapdor et al., Z.Phys.A292, 1979,249; D. Habs et. al. Z.Phys. A285 (1978), 53

P_{BDF} Probability: Extraction of fission barriers?!

PHYSICAL REVIEW C 86, 024308 (2012)



FIG. 3. (Color online) (a) Fission-barrier heights of ¹⁸⁰Hg in four variants A–D. Four β -strength functions were used: calculated

Clearly – model-dependent (but we tried many parametrizations) Conclusion: "experimental barriers" are always lower than calculated

15

10

 E^* (MeV)

10

10

 $|Q_{\rm EC}|$

 $|Q_{EC}|$

Mass Separator ISOLDE (CERN)



Detection system for βDF studies at ISOLDE



A.Andreyev et al. PRL 105 (2010)

Mass distribution of fission fragments from bDF of ¹⁸⁰Tl

ASYMMETRIC energy split! Thus asymmetric mass split: $M_H=100(4)$ and $M_L=80(4)$



The most probable fission fragments are ¹⁰⁰Ru (N=56,Z=44) and ⁸⁰Kr (N=44,Z=36)

New Type of Asymmetric Fission in Proton-Rich Nuclei



¹⁸⁰Hg: More surprises? How does ¹⁸⁰Hg fission at *higher* excitation energies?



Supported by Reimei Foundation (JAEA)

Two types of asymmetry: what's the difference?

PHYSICAL REVIEW C 86, 024610 (2012)

Contrasting fission potential-energy structure of actinides and mercury isotopes

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Conclusions: The mechanism of asymmetric fission must be very different in the lighter proton-rich mercury isotopes compared to the actinide region and is apparently unrelated to fragment shell structure. Isotopes lighter than ¹⁹²Hg have the saddle point shielded from a deep symmetric valley by a significant ridge. The ridge vanishes for the heavier Hg isotopes, for which we would expect a qualitatively different asymmetry of the fragments.



'Brownian Metropolis Shape Motion'

based on J. Randrup and P. Moller, PRL 106, 132503 (2011)

Phys. Rev. C 85, 024306 (2012)

Calculated fission yields of neutron-deficient mercury isotopes

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The recent unexpected discovery of asymmetric fission of ¹⁸⁰Hg following the electron-capture decay of ¹⁸⁰Tl has led to intense interest in experimentally mapping the fission-yield properties over more extended regions of the nuclear chart and compound-system energies. We present here a first calculation of fission-fragment yields for neutron-deficient Hg isotopes, using the recently developed Brownian Metropolis shape motion treatment. The results for ¹⁸⁰Hg are in approximate agreement with the experimental data. For ¹⁷⁴Hg the symmetric yield increases strongly with decreasing energy, an unusual feature, which would be interesting to verify experimentally. PACS numbers: 25.85.-w, 24.10.Lx, 24.75.+i



FIG. 4. (Color online) Minima, saddles, major valleys, and ridges in the 5D potential-energy surface of ¹⁸⁰Hg (see text). At the last plotted point on the fission barrier, $(Q_2/b)^{(1/2)} \approx 11$, the asymmetry of the shape is $A_{\rm H}/A_{\rm L} = 108/72$.





'Improved Scission-Point Model'

PHYSICAL REVIEW C 86, 044315 (2012)

Mass distributions for induced fission of different Hg isotopes

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With the improved scission-point model mass distributions are calculated for induced fission of different Hg isotopes with even mass numbers A = 180, 184, 188, 192, 196, and 198. The calculated mass distribution and mean total kinetic energy of fission fragments are in good agreement with the existing experimental data. The asymmetric mass distribution of fission fragments of ¹⁸⁰Hg observed in the recent experiment is explained. The change in the shape of the mass distribution from asymmetric to more symmetric is revealed with increasing A of the fissioning ^AHg nucleus, and reactions are proposed to verify this prediction experimentally.

- Inter-fragment distance is not fixed and calculated.
 values of ~0.5-1 fm result (Wilkins fixed at 1.4 fm)
- •Mass symmetry/asymmetry doesn't change as a function of E* (up to E*~60 MeV) good for future experiments



'Self-consistent Scission-Point Model'

PHYSICAL REVIEW C 86, 064601 (2012)

Role of deformed shell effects on the mass asymmetry in nuclear fission of mercury isotopes

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$$\begin{split} E_{\text{av}}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d) \\ &= E_{\text{tot}} - E_{\text{HFB}}(Z_1, N_1, \beta_1) - E_{\text{HFB}}(Z_2, N_2, \beta_2) \\ &- E_{\text{nucl}}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d) - E_{\text{Coul}}(Z_{1,2}, N_{1,2}, \beta_{1,2}, d). \end{split}$$



FIG. 4. (Color online) Total nuclear density for the most energetically favorable scission configuration in ¹⁸⁰Hg fission, extracted from a self-consistent HFB calculation. In the lower part of the figure, two



FIG. 2. (Color online) Minimum absolute available energy at scission calculated for all possible fragmentations in (a) 180 Hg and (b) 198 Hg fission at 10 MeV and in (c) the thermal *n*-induced fission of 235 U.

'Mean-field HFB+Gogny D1S'

PHYSICAL REVIEW C 86, 024601 (2012)

Fission modes of mercury isotopes

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FIG. 2. (Color online) PES for ¹⁸⁰Hg (top) and ¹⁹⁸Hg (bottom) in the plane of collective coordinates $Q_{20} - Q_{30}$ in HFB-SkM^{*}. The aEF fission pathway corresponding to asymmetric elongated fragments is marked. The difference between contour lines is 4 MeV. The effects due to triaxiality, known to impact inner fission barriers in the actinides, are negligible here.

FIG. 3. (Color online) PES in HFB-D1S for ¹⁸⁰Hg (top) and ¹⁹⁸Hg (bottom) in the (Q_{20}, Q_{30}) plane in the pre-scission region of aEF valley. The symmetric limit corresponds to $Q_{30} = 0$. The aEF valley and density profiles for pre-scission configurations are indicated. The difference between contour lines is 0.5 MeV. Note different Q_{30} -scales in ¹⁸⁰Hg and ¹⁹⁸Hg plots.

β**DF of** ¹⁷⁸**TI @ISOLDE** V. Liberati et al (PRC, 2013, in print)



From Asymmetry to Symmetry



Fission of Proton-rich nuclei with A~180-200

Courtesy P. Moller (LANL) and J. Randrup (LBNL), 5th ASRC workshop on Fission, Tokai 2012

CERN-ISOLDE

JAEA tandem



IS534 (ISOLDE) , 9-14 May 2012: Mass Distributions Measurements of ^{194,196}Po via βDF of ^{194,196}At



IS534, 9-14 May 2012: Mass Distributions Measurements of $^{194,196}\text{Po}$ via βDF of $^{194,196}\text{At}$



Clear difference in energy (thus, mass) distribution between 2-peaked fission of ¹⁸⁰Hg and a broad distribution in ^{194,196}Po

May and June 2012: Mass Distributions Measurements via β DF of ^{194,196}At and ^{200,202}Fr



Mapping beta-delayed fission: from neutron-deficient to neutron-rich nuclei

Reviews of Modern Physics, 85, 1541 (2013)

Colloquium: Beta-delayed fission of atomic nuclei

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This Colloquium reviews the studies of exotic type of low-energy nuclear fission, the β -delayed fission (β DF). Emphasis is made on the new data from very neutron-deficient nuclei in the lead region, previously scarcely studied as far as fission is concerned. These

Known Beta-delayed fission nuclei

Isotope	$T_{1/2}$	$Q_{EC} - B_f$	Production ^{&} ,	$P_{\beta DF}$	Observables*	References
	7	[MeV]	Separation,			
			Detection			
β^+ /EC –delayed fission in the neutron-deficient isotopes						
178 Tl	252(20) ms	1.82	SR, IS, WM	${f 1.5(6) imes 10^{-3}}$	Z,A,T,KE,TKE,MD,GF	(Liberati <i>et al.</i> , 2013)
180 Tl	$1.09(1) \ s$	0.63	SR, IS, WM	${f 3.2(2) imes 10^{-5}}$	Z,A,T,KE,TKE,MD,GF	(Elseviers $et al., 2013$)
	$0.97^{+0.09}_{-0.08} { m s}$		FE,NS,MF	$\sim 3 \times 10^{-(7\pm 1)}$	T,EXF	(Lazarev et al., 1987, 1992)
$^{186m1,m2}{ m Bi}$	$9.8(4), 14.8(8) \text{ ms}^{\#}$	2.09	FE,RS,Si/Ge	$7.6\times10^{-2,e}$	T,EXF,KE,GF	(Lane <i>et al.</i> , 2013)
$^{188m1,m2}{ m Bi}$	$\sim 0.3 \ { m s}^c$	0.51	FE,NS,MF	$3.4 \times 10^{-4, a, c}$	T,EXF	(Lazarev et al., 1992)
	$265(10), 60(3) \text{ ms}^{\#}$		FE,RS,Si/Ge	$(0.16 - 0.48) \times 10^{-2,f}$	T,EXF,KE,GF	(Lane <i>et al.</i> , 2013)
192m1,m2 At	$88(6), 11.5(6) \text{ ms}^{\#}$	2.09	FE,RS,Si/Ge	$(7-35) \times 10^{-2}$	T,EXF,KE,GF	(Andreyev et al., 2013)
194m1,m2 At	$310(8), 253(10) \text{ ms}^{\#}$	-0.04	FE,RS,Si/Ge SR.IS.WM	$\sim (0.8 - 1.6) \times 10^{-2}$	T,EXF,KE,GF Z.A.T.KE.TKE MD.GF	(Andreyev et al., 2013) (Andreyev et al., 2012)
196 At	$0.23^{+0.05}$ s	-1.19	FE NS ME	$8.8 \times 10^{-4, a}$	T EXF	(Lazarev et al. 1992)
110	0.20-0.03	1.10	SB IS WM	0.0 / 10	Z A T KE TKE MD GE	(Andrevev et al 2012)
200 Fr	$49(4) \text{ ms}^{\#}$	0.82	SR IS WM		Z A T KE TKE MD GE	(Andrevev et al., 2012)
202m1, m2 Fr	$0.30(5), 0.29(5), s^{\#}$	-1.17	SR IS WM		Z A T KE TKE MD GF	(Andrevev $et al. 2011$)
²²⁸ Np	61.4(14) s	-0.87	FE.BC.MG	$2.0(9) \times 10^{-4}$	Z.T.KE.TKE.MD.GF	(Kreek et al., 1994a)
1.	60(5) s		FE.NS.MF	(.) /	T.EXF	(Kuznetsov $et al., 1966$)
232 Am	$1.31(4) \min$	1.65	FE.RC.MG	$6.9(10) imes 10^{-4}$	Z.T.KE.TKE.MD.GF	(Hall <i>et al.</i> , 1990a)
	55(7) s		FE.NS.Si	$(1.3^{+4}) \times 10^{-2}$	T.KE	(Habs et al., 1978)
	1.40(25) min		FE.NS.MF	6.96×10^{-2}	T.EXF	(Kuznetsov <i>et al.</i> , 1967)
234 Am	$2.32(8) \min$	0.29	FE,RC,MG	$6.6(18) imes 10^{-5}$	Z,T,KE,TKE,MD,GF	(Hall <i>et al.</i> , 1989a, 1990b)
	2.6(2) min		FE,NS,MF	$\sim 6.95 \times 10^{-5}$	T,EXF	(Kuznetsov et al., 1967)
238 Bk	144(5) s	-0.15	FE,RC,MG	$4.8(20) imes 10^{-4}$	Z,T,KE,TKE,MD,GF	(Kreek <i>et al.</i> , 1994b)
$^{240}\mathbf{Bk}$	$4.2(8) \min$	-1.99	FE,NS,MF	$(1.3^{+1.8}_{-0.7}) imes 10^{-5}$	Т	(Galeriu, 1983)
	5(2) min		FE,NS,MF	$1 \times 10^{-5, b}$	Т	(Gangrsky et al., 1980)
242 Es	11(3) s	-0.94	FE,RC,MG	${f 0.6(2) imes 10^{-2}}$	Z,T,KE,TKE,MD	(Shaughnessy et al., 2000)
	5 - 25 s		FE,RS,Si	$1.4(8) \times 10^{-2}$	T,KE	(Hingmann et al., 1984)
	17.8(16) s		FE,RS,Si	$(1.3^{+1.2}_{-0.7}) \times 10^{-2}$	T,KE	(Antalic <i>et al.</i> , 2010)
$^{244}\mathbf{Es}$	38(11) s	-2.24	FE,RC,MG	$1.2(4) imes 10^{-4}$	Z,T,KE,TKE,MD	(Shaughnessy et al., 2002)
			FE,NS,MF	$1 \times 10^{-4, b}$	Т	(Gangrsky et al., 1980)
^{246}Es	$7.7(5) \min$	-3.47	FE,RC,MG	$({f 3.7}^{+8.5}_{-3.0}) imes {f 10}^{-5}$	Z,T,KE	(Shaughnessy et al., 2001)
	8 min		FE,NS,MF	$3 \times 10^{-5, b}$	Т	(Gangrsky et al., 1980)
248 Es	$23(3) \min$	-4.26	FE,RC,MG	$3.5(18) imes 10^{-6}$	Z,T,KE	(Shaughnessy et al., 2001)
			FE,NS,MF	$3 \times 10^{-7, b}$	Т	(Gangrsky et al., 1980)
$^{246m1,m2}\mathrm{Md}$	0.9(2), 4.4(8) s	0.14	FE,RS,Si	$> 1 \times 10^{-1}$	T,KE	(Antalic <i>et al.</i> , 2010)
	$1.0(4) \ s^c$		FE,RS,Si	$\sim\!0.65\times10^{-1}$	T,KE	(Ninov et al., 1996)
^{250}Md	$52(6) s^{\#}$	-2.64	FE,NS,MF	$2 \times 10^{-4, b}$	Т	(Gangrsky et al., 1980)
β^- -delayed fission in the neutron-rich isotopes						
²²⁸ Ac	$6.15(2) h^{\#}$	-4.45	LLP, RC, MF/Ge	$5(2) \times 10^{-12}$		(Yanbing $et al., 2006$)
230 Ac	$122(3) \text{ s}^{\#}$	-2.73	TR, RC, MF/Ge	$1.19(40) \times 10^{-8}$		(Shuanggui et al., 2001)
256m Es	7.6 h [#]	-3.23	TR, RC, Si/Ge	2×10^{-5}	T,KE	(Hall <i>et al.</i> , 1989b)
^{234gs} Pa	$6.70(5) h^{\#}$	-2.55	NI,NS,MF	$3 \times 10^{-12, d}$	Т	(Gangrsky et al., 1978)
234m Pa	$1.159(11) \min^{\#}$		LLP,RC,MF	$10^{-12, d}$	Т	(Gangrsky et al., 1978)
²³⁶ Pa	$9.1(1) \min^{\#}$	-2.02	SR,RC,MF/Ge	$\sim 10^{-9}$	Т	(Batist <i>et al.</i> , 1977)
200			FE/GI,NS,MF	$10^{-9, d}/3 \times 10^{-10, d}$	Т	(Gangrsky et al., 1978)
²³⁸ Pa	$2.3(1) \min^{\#}$	-2.14	NI,NS,MF	$6 \times 10^{-7}, 1 \times 10^{-8, d}$	Т	(Gangrsky et al., 1978)
			NI,RC,MF	$< 2.6 \times 10^{-8}$		(Baas-May <i>et al.</i> , 1985)

Mapping 'Terra Incognita' in Low-Energy Fission



A. N. Andreyev, M. Huyse, P. Van Duppen, "Beta-delayed Fission in atomic nuclei", Reviews of Modern Physics, 85, 1541 (2013)