Entry Distribution, Fission Barrier, and Formation Mechanism of $^{254}_{102}$No

P. Reiter,[1,2] T. L. Khoo,[1] T. Lauritsen,[1] C. J. Lister,[1] D. Seweryniak,[1] A. A. Sonzogni,[1] I. Ahmad,[1] N. Amzal,[3] P. Bhattacharyya,[4] P. A. Butler,[3] M. P. Carpenter,[1] A. J. Chewter,[3] J. A. Cizewski,[1,5] C. N. Davids,[1] K. Y. Ding,[5] N. Fotiades,[5] J. P. Greene,[1] P. T. Greenlees,[3] A. Heinz,[1] W. F. Henning,[1] R.-D. Herzberg,[3] R. V. F. Janssens,[1] G. D. Jones,[3] H. Kankaanpää,[7] F. G. Kondev,[1] W. Korten,[6] M. Leino,[7] S. Siem,[1,8] J. Uusitalo,[1] K. Vetter,[9] and I. Wiedenhöver[1]

[1]*Argonne National Laboratory, Argonne, Illinois 60439*
[2]*Ludwig-Maximilians-Universität, Am Coulombwall 1, D-85748 Garching, Germany*
[3]*University of Liverpool, Liverpool L69 7ZE, England*
[4]*Purdue University, West Lafayette, Indiana 47097*
[5]*Rutgers University, New Brunswick, New Jersey 08903*
[6]*DAPNIA/SPhN, CEA Saclay, F-91191 Gif-sur-Yvette Cedex, France*
[7]*University of Jyväskylä, Jyväskylä, Finland*
[8]*University of Oslo, Oslo, Norway*
[9]*Lawrence Berkeley National Laboratory, Berkeley, California 94720*

(Received 3 January 2000)

The entry distribution in angular momentum and excitation energy for the formation of ^{254}No has been measured after the ^{208}Pb(^{48}Ca, $2n$) reaction at 215 and 219 MeV. This nucleus is populated up to spin $22\hbar$ and excitation energy $\gtrsim 6$ MeV above the yrast line, with the half-maximum points of the energy distributions at ~ 5 MeV for spins between $12\hbar$ and $22\hbar$. This suggests that the fission barrier is $\gtrsim 5$ MeV and that the shell-correction energy persists to high spin.

PACS numbers: 27.90.+b, 21.10.Hw, 24.60.Dr, 24.75.+i

The heaviest nuclei, with $Z > 100$, are at the limit of Coulomb instability. They would be unstable against spontaneous fission but for a large shell-correction energy, which leads to additional binding and creates a sizable fission barrier of up to 8 MeV [1–3]. The existence of these very heavy elements is a striking manifestation of shell structure in nuclei, and arises from the identical mechanism responsible for the proposed [4] stability of an "island" of superheavy elements around $Z = 114$, $N = 184$. The detection of a series of elements with Z up to 112 [5] and recent reports [6,7] of the detection of elements 114 and 116, 118 provide support for the stability of superheavy elements.

Shell-stabilized nuclei could be different from ordinary nuclei, where the binding is largely derived from the liquid-drop energy. However, there is little experimental information on the properties of shell-stabilized nuclei. Their high-spin behavior, e.g., the variation with spin of the moment of inertia and of the fission barrier, would provide information about the angular momentum dependence of the shell energy, which is not only interesting in its own right but also provides a new test of theories that calculate the properties of superheavy elements. Since the heaviest nuclei are only weakly bound in the ground state, it is interesting to determine the limiting spin and excitation energy that they can sustain. The limits of stability in spin and excitation energy are governed by the fission barrier. Knowledge of the barrier is also essential for understanding the production mechanism of superheavy nuclei. The synthesis process may be viewed in two steps: (i) formation of a compound nucleus and (ii) survival of the latter against fission as it cools by emitting neutrons and gamma rays. The competition against fission in step (ii) is largely governed by the fission barrier. At high angular momentum, the barrier depends sensitively on how the shell-correction energy varies with spin, since the barrier is predominantly generated by the shell correction. Hence, information on the barrier is not only important for understanding the formation of superheavy nuclei but also provides a measure of the robustness of the shell correction against rotation.

In lighter actinide nuclei, the fission barrier parameters are most directly obtained in nucleon-transfer [8] or neutron-capture reactions [9] from the variation of the fission probability as a function of excitation energy E^*. Since no suitable target exists, this method is not applicable to the heaviest elements. We propose a new method that may be used to deduce the fission barrier $B_f(I)$ and also its variation with spin I. Below the neutron threshold, where fission and γ decay are the only open channels, the fission probability $P_f(E^*)$ is given by $P_f(E^*) = \frac{\Gamma_f}{\Gamma_f+\Gamma_\gamma}$, where Γ_f and Γ_γ are the fission and γ decay widths. Equivalently, one can also use the probability for γ emission, $P_\gamma(E^*) = 1 - P_f(E^*)$. $P_\gamma(E^*)$ can be obtained from the so-called entry distribution, which is the initial population cross-section distribution, $\sigma(I, E^*) = \sigma_n(I, E^*) \cdot P_\gamma(I, E^*)$, from which γ decay starts towards the ground state. The cross section $\sigma_n(I, E^*)$ to populate a state at (I, E^*) after emission of neutrons can be estimated from statistical model calculations. However, in order to be independent of such a calculation in this work, we note that successful γ competition yields a point (I, E^*) in the entry distribution, which indicates that the energy of

0031-9007/00/84(16)/3542(4)$15.00 © 2000 The American Physical Society

the saddle point, $E_s(I) = E_{\mathrm{yrast}}(I) + B_f(I)$, is located above this point. This follows from the fact that the gamma probability rapidly decreases towards zero above the barrier, since the time scale for fission is much faster than that for γ emission.

In this Letter, we present results on the entry distributions of $^{254}_{102}$No, which is an example of a shell-stabilized nucleus [1,2]. We have previously identified [10] the ground-state band (gsb) of ^{254}No up to spin 14, and have deduced a deformation parameter of $\beta = 0.27(2)$. A subsequent measurement at Jyväskylä [11] confirmed these results and extended the gsb up to spin 16.

The reaction ^{208}Pb(^{48}Ca, $2n$)^{254}No was used to populate states in ^{254}No. Gammasphere [12], a multidetector array comprising 101 Ge detectors, surrounded by bismuth germanium oxide (BGO) Compton suppressors, was used to measure not only the γ rays with high resolution but also the γ-ray multiplicity and sum energy. The γ rays from ^{254}No nuclei were extracted from a background due to fission, which was $>10^4$ times more intense, by requiring coincidences with evaporation residues. The latter were unambiguously identified with the Argonne Fragment Mass Analyzer (FMA) [13], through measurements of (i) the mass/charge ratio [in the focal-plane detector, a parallel-plate avalanche counter (PPAC)], (ii) the time of flight between the PPAC and a double-sided Si strip detector (DSSD) located 40 cm downstream, (iii) the time of flight between the target and PPAC, and (iv) the implant energy in the DSSD. These conditions provided clean γ-ray spectra of ^{254}No, as verified by the recoil-decay tagging method, which correlated an α decay with a nobelium implant in the same pixel of the DSSD. Further details of the analysis methods are described in Ref. [10].

To minimize deterioration of the ^{208}Pb targets ($\approx$0.5 mg/cm^2), they were mounted on a rotating wheel and the beam was wobbled vertically ±2.5 mm across the target with a magnetic steerer (with no loss of the FMA mass resolution). Beams with energies of 215 and 219 MeV and intensities of 9 to 12 pnA were provided by ATLAS, the Argonne superconducting linear accelerator. The compound nucleus (CN) excitation energies (at mid target) were $E_{\mathrm{CN}} \approx 19.3$ and 22.7 MeV, respectively. Our beam energies are positioned around the maximum, located at $E_{\mathrm{CN}} \approx 21.7$ MeV [11,14], of a narrow excitation function for the production of ^{254}No.

The γ spectra obtained at 215 MeV, previously published in [10], and at 219 MeV are compared in Fig. 1. Transitions from higher-spin members of the gsb are clearly enhanced at the higher bombarding energy and the gsb could be extended up to spin 20^+ [Fig. 1(b)]. The assignment of the $20^+ \rightarrow 18^+$ transition is tentative; it is based on a smooth continuation of the gsb (see inset showing the moment of inertia), and is also supported by some $\gamma\gamma$ coincidences. The relative transition intensities, given in the insets, show that the population has saturated by spin 8 at the higher beam energy.

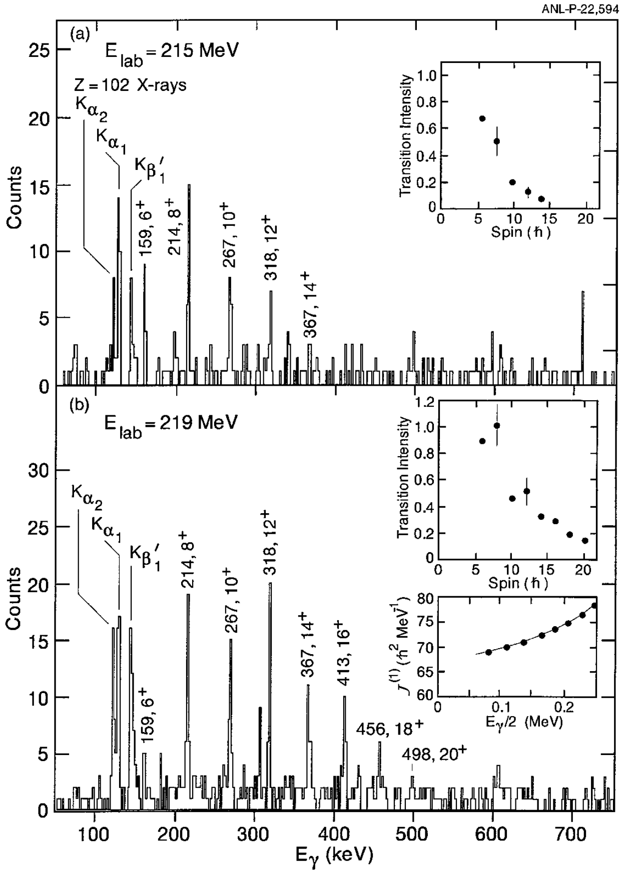

FIG. 1. ^{254}No γ spectra, obtained by requiring recoil-γ coincidences in Gammasphere and the FMA focal-plane detectors, at beam energies of (a) 215 MeV and (b) 219 MeV. The gsb transitions are labeled by their energies (in keV) and initial spins. Note the large increase in high-spin population at the higher beam energy, which is also seen in the insets that show relative intensities (with some typical statistical errors). The $2^+ \rightarrow 0^+$ and $4^+ \rightarrow 2^+$ transitions, which were almost completely converted, are defined to have unit intensity. The equal intensities (within errors) of transitions from the 6^+ and 8^+ levels in inset (b) suggest that the population has saturated by spin 8. In inset (a), the relative intensities of the detected transitions were chosen such that they extrapolate to unity by spin 4. A second inset in (b) shows the moment of inertia $\mathcal{J}^{(1)}$ vs $E_\gamma/2$.

In order to determine the initial angular momentum and excitation energy of the ^{254}No residues, we measured the number of detector modules that fired and the total energy emitted by γ radiation. These quantities could be measured because Gammasphere is an efficient $\approx 4\pi$ γ detector, with a granularity of 101 detector modules and a sum-energy efficiency of 72% (for 898-keV photons). (A module consists of a Ge crystal and its surrounding BGO detectors.) Source measurements were performed to deduce the sum-energy and multiplicity response functions of the array by using an event mixing technique [15]. Based on the measured response functions, a two-dimensional Monte Carlo unfolding procedure [16,17] transformed the two-dimensional distribution of detector multiplicity vs γ sum energy into a distribution of γ multiplicity vs excitation energy. (To correct for the effect of the trigger requirement of two Compton suppressed Ge events, the efficiency dependence on multiplicity was taken into account.)

The initial spin of the evaporation residue is deduced from the γ multiplicity by using the expression $I = \Delta I(N_\gamma + N_e - N_{\mathrm{stat}}) + \Delta I_{\mathrm{stat}} \cdot N_{\mathrm{stat}}$, where N_γ and N_e are the γ and conversion-electron multiplicities, while N_{stat} is the number of statistical γ rays. The following values were adopted: $\Delta I = 2$ (appropriate for a well-deformed nucleus) and $\Delta I_{\mathrm{stat}} = 0.25$, $N_{\mathrm{stat}} = 3$ (appropriate for moderate spins and excitation energies). For high-Z nuclei, the internal conversion coefficients can be very large for low-energy transitions. From the measured properties of the gsb (see Fig. 1 and Ref. [10]), the multiplicity of the conversion electrons are deduced: $N_e = 2.9^{+1}_{-0.5}$ and $3.6^{+0.5}_{-0.2}$ at $E_{\mathrm{Lab}} = 215$ and 219 MeV, respectively; the uncertainties represent systematic errors.

The entry distributions, which represent the starting point for γ decay and formation of ^{254}No, are shown in Fig. 2 for the two beam energies. The one-dimensional spin and excitation energy distributions are also given. It is evident that a small increase in beam energy leads to noticeably higher initial spins and excitation energies. The entry distribution at the lower beam energy reveals that it is the maximum allowable energy $E^*_{\max}$ after neutron emission that imposes a $16\hbar$ limit on the angular momentum and not the fission barrier. ($E^*_{\max} = E_{\mathrm{CN}} - S_{n1} - S_{n2}$; E_{CN} is the compound-nucleus excitation energy, and S_{n1}, S_{n2} are the neutron separation energies.) At the higher beam energy, states up to spin $22\hbar$ and $E^* = 8.5$ MeV (up to 6 MeV above the yrast line) are populated in the entry distribution, showing that the nucleus clearly can survive against fission up to these limits. The intensities of the gsb transitions are consistent with the relative cross sections given by the entry-spin distribution, e.g., the gsb is populated up to spin 20. At the higher beam energy, the entry distribution no longer extends to $E^*_{\max}$, perhaps an indication of fission competition.

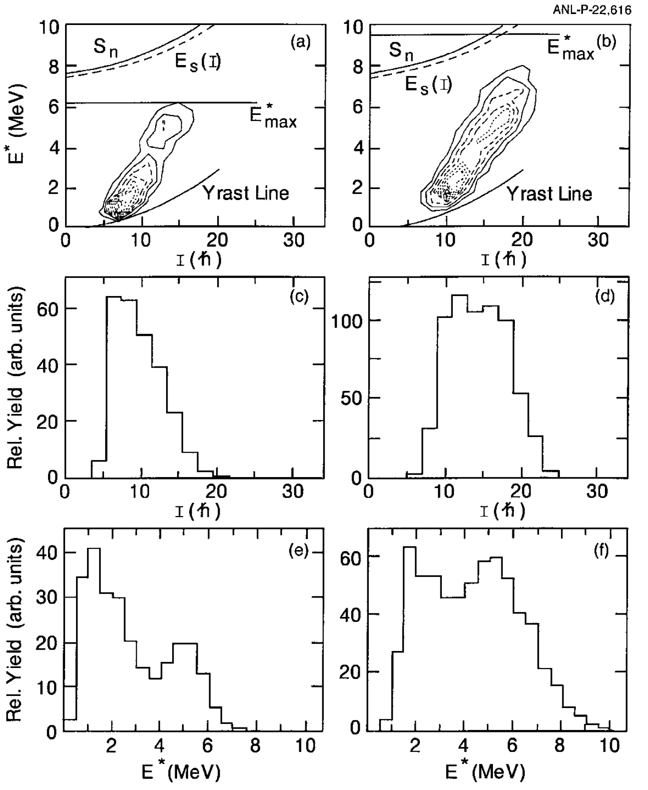

FIG. 2. Contour plots [(a) and (b)] of the entry distributions in spin and excitation energy and their projections at $E_{\mathrm{Lab}} = 215$ (left panels) and 219 (right panels) MeV. The measured yrast line, the neutron-separation energy S_n, a theoretical [19] saddle-point energy $E_s(I)$, and the maximum allowable energy, $E^*_{\max} = E_{\mathrm{CN}} - S_{n1} - S_{n2}$, in ^{254}No are indicated. The distributions in spin [(c) and (d)] and excitation energy [(e) and (f)] are also shown.

Gamma decay to the ground state originates from the entry distribution, implying successful γ competition over fission. As described earlier, the highest-energy point of the entry distribution for each spin lies below the saddle energy, $E_s(I) = E_{\mathrm{yrast}}(I) + B_f(I)$ (or within 0.5 MeV), so that a lower bound on $B_f(I)$ can be obtained. Only a lower bound on B_f can be deduced since the decreasing population with increasing excitation energy could, in principle, also be due to a reduced cross section after neutron emission. Energy distributions for individual spin bins, projected from the entry distributions in Figs. 2(a) and 2(b), show that the half-maximum points correspond to 5 MeV above the yrast line for $I \geq 12$. This suggests that, even at high spin, $B_f \geq 5$ MeV, a surprisingly large value for a nucleus as fissile as ^{254}No.

In fact, this value is comparable to our estimate [18] of the barrier for ^{238}U ($\sim$5.4 MeV at spin 20), where the dominant contribution is from the liquid-drop term, which decreases with spin. In contrast, in ^{254}No the dominant contribution to the barrier comes from the shell energy, which accounts for 6.6 MeV of the theoretical static barrier [19] of 7.6 MeV at zero spin (calculated as described in Ref. [3]). There are no calculations of the shell-correction energy at higher spin, but, if it were to remain constant with spin, the saddle-point energy, $E_s(I)$, would lie along the dashed line in Fig. 2. $B_f(I)$ is calculated [19] as $B_f(I) = B_f(0) + [B_{ld}(I) - B_{ld}(0)]$, where $B_{ld}(I)$ denotes the liquid-drop barrier at spin I. The slight decrease from the solid line denoting the neutron separation energy is due to the diminution of the liquid-drop term. The information on the barrier also gives a constraint on the shell energy at high spin: $E^{\exp}_{\mathrm{shell}} = B^{\exp}_f(I) - B_{ld}(I) \gtrsim 4$ MeV for $I = 12$–22. The measured entry distribution contradicts the hypothesis [20,21] that the shell-correction energy damps to $1/e$ of its zero spin value at spin 15. Such a damping would have chopped off a substantial upper portion of the entry distribution, e.g., $E^* < 4.8$ MeV at spin 20.

Our data provide new information important for understanding the synthesis of superheavy nuclei. Previously, the only constraints on theory have come from excitation function measurements of ground-state cross sections. The present results reveal that high partial waves contribute to the formation of evaporation residues, as predicted by Smolanczuk [22]. The fission barrier governs the survival of the compound nucleus, as it evaporates neutrons and γ rays in competition with fission decay. Fusion-evaporation calculations, similar to those of Ref. [23], suggest that a barrier of 5 MeV would lead to a much larger cross section for production of ^{254}No than is observed [11,14]. This suggests either that there is a hindrance in the formation of the compound nucleus or that the fission barrier damps rapidly with excitation energy.

An unexpected feature of our entry distribution is the sharp tilt angle with respect to the yrast line. This is due, at least in part, to the small excitation energy of the low-spin entry states, which appears to be a distinct component [see Figs. 2(a), 2(b), 2(e), and 2(f)]. This feature cannot be easily explained by a simple statistical model. At low excitation energy, the level density is small, so that states near the yrast line should have only a small population. The deviation of the entry distribution from the line representing $E^*_{\max}$ gives the energy removed by the two neutrons. Hence, evaporation residues with low partial waves appear to be associated with unusually energetic neutrons. On the other hand, at higher spin ($I \geq 14$) the high excitation energy above the yrast line is more normal. Hence, there is a hint of at least two mechanisms in the formation of superheavy nuclei: a normal statistical one responsible for high-spin formation and another one with emission of higher energy (perhaps preequilibrium) neutrons, which is important at lower spins. The possibility of preequilibrium neutron emission has been discussed by Armbruster [20]. However, other cases should be investigated before firm conclusions are drawn. It cannot be ruled out that the low sum energy events are due to losses from decay γ rays below a 0.28 s isomer [24], likely with $I^\pi = 8^-$. However, this explanation is not probable since a coincidence gate set on the low-energy portion of the entry distribution yielded the transition from the 8^+ gsb level, which, if populated by the isomer, would have decayed far away from the acceptance region of Gammasphere.

In summary, we have measured the entry distribution for a shell-stabilized nucleus. The limiting angular momentum and excitation energy are deduced for excited states in ^{254}No after the ^{208}Pb(^{48}Ca, $2n$) reaction. The data provide direct information on the fission barrier and on the shell-correction energy, based on a novel experimental technique to determine a lower bound of the barrier height. In the synthesis of very heavy nuclei, the entry distributions suggest that high partial waves contribute and that there may be more than one reaction mechanism.

The work is supported by the U.S. Department of Energy, under Contracts No. W-31-109-ENG-38 and No. DE-AC03-76SF00098, the U.S. National Science Foundation, the U.K. Engineering and Physical Sciences Research Council, and a NATO grant (for S. S.) through the Research Council of Norway. We acknowledge the efforts of the ATLAS crew in providing the required currents of ^{48}Ca, and of J. Falout, J. Fox, and B. Nardi for help with the target wheel. We thank B. B. Back, R. Smolanczuk, and A. Sobiczewski for valuable private communications.

[1] A. Sobiczewski, Phys. Part. Nuclei **25**, 119 (1994).
[2] P. Möller and J. R. Nix, J. Phys. G **20**, 1681 (1994).
[3] R. Smolanczuk *et al.*, Phys. Rev. C **52**, 1871 (1995).
[4] S. G. Nilsson *et al.*, Nucl. Phys. **A115**, 545 (1968).
[5] S. Hofmann, Rep. Prog. Phys. **61**, 639 (1998).
[6] Yu. Ts. Oganessian *et al.*, Nature (London) **400**, 242 (1999); Phys. Rev. Lett. **83**, 3154 (1999).
[7] V. Ninov *et al.*, Phys. Rev. Lett. **83**, 1104 (1999).
[8] B. B. Back *et al.*, Phys. Rev. C **9**, 1924 (1974).
[9] S. Bjørnholm and J. E. Lynn, Rev. Mod. Phys. **52**, 725 (1980).
[10] P. Reiter *et al.*, Phys. Rev. Lett. **82**, 509 (1999).
[11] M. Leino *et al.*, Eur. Phys. J. A **6**, 63 (1999).
[12] I. Y. Lee, Nucl. Phys. **A520**, 641c (1990).
[13] C. N. Davids *et al.*, Nucl. Instrum. Methods Phys. Res., Sect. B **70**, 358 (1992).
[14] H. W. Gäggeler *et al.*, Nucl. Phys. **A502**, 561c (1989).
[15] M. Jääskeläinen *et al.*, Nucl. Instrum. Methods Phys. Res. **204**, 385 (1983).
[16] Ph. Benet, Ph.D. thesis, L'Universite Louis Pasteur de Strasbourg, CRN/PN 88-29, 1988.
[17] T. Lauritsen *et al.*, Phys. Rev. Lett. **69**, 2479 (1992).
[18] $B_f(I)$ for ^{238}U was calculated using the Sierk model for the liquid-drop term and a constant shell-correction energy, given by the difference between the experimental and liquid-drop mass.
[19] R. Smolanczuk (private communication).
[20] P. Armbruster, in *Proceedings of the Robert A. Welch Foundation, 41st Conference on Chemical Research, The Transactinide Elements* (Robert A. Welch Foundation, Houston, TX, 1997), p. 231.
[21] K.-H. Schmidt *et al.*, in *Proceedings of the Symposium on Physics and Chemistry of Fission, Jülich* (IAEA, Vienna, 1980), p. 409.
[22] R. Smolanczuk, Phys. Rev. C **59**, 2634 (1999).
[23] A. A. Sonzogni *et al.*, Phys. Rev. C **58**, R1873 (1998).
[24] A. Ghiorso *et al.*, Phys. Rev. C **7**, 2032 (1973).