Cluster radioactivity of $^{294}_{118}$Og$_{176}$

Zachary Matheson,1 Samuel Giuliani,1 Witold Nazarewicz,1 Jhilam Sadhukhan,2,3 and Nicolas Schunck4

1 Department of Physics and Astronomy and FRIB Laboratory, Michigan State University, East Lansing, Michigan 48824, USA
2 Variable Energy Cyclotron Centre, Kolkata 700064
3 Homi Bhabha National Institute, Mumbai 400094, India
4 Nuclear and Chemical Science Division, Lawrence Livermore National Laboratory, Livermore, California 94551, USA

(Dated: December 18, 2018)

According to theory, cluster radioactivity becomes an important decay mode in superheavy nuclei.

In this work, we predict that the strongly-asymmetric fission, or cluster emission, is in fact the dominant fission channel for $^{294}_{118}$Og$_{176}$, which is currently the heaviest synthetic isotope known. Our theoretical approach incorporates important features of fission dynamics, including quantum tunneling and stochastic dynamics up to scission. We show that, despite appreciable differences in static fission properties such as fission barriers and spontaneous fission lifetimes, the prediction of cluster radioactivity in $^{294}_{118}$Og$_{176}$ is robust with respect to the details of calculations, including the choice of energy density functional, collective inertia, and the strength of the dissipation term.

Introduction – The region of superheavy nuclei ($Z \geq 104$) is one of the frontiers of modern nuclear physics. The heavy-ion fusion experiments have been able to push the boundaries of the nuclear chart all the way to $^{208}_{82}$Pb $\rightarrow ^{180}_{68}$Ra $\rightarrow ^{209}_{82}$Pb $+ ^{14}_{6}$C decay [29]. It is always a rare event with a small branching ratio [30]. From the theoretical point of view, half-life calculations based on semiempirical models predict cluster radioactivity to be the dominant decay channel of several superheavy nuclei [31–37]. Similar predictions have been obtained by more microscopic calculations using nuclear DFT framework [23, 38]. However, so far no determination of fission yields has been made that explicitly demonstrates the emergence of cluster radioactivity in superheavy elements, either theoretically or experimentally. In this paper, which extends the discussion of recent Ref. [23] to fission yields, we calculate spontaneous fission yields of $^{294}_{118}$Og$_{176}$ and explicitly predict for the first time that cluster emission is dramatically enhanced to the point that it becomes the primary spontaneous fission channel.

In addition to this prediction, our work is a tour de force demonstration of the state-of-the-art in microscopic theory of spontaneous fission. First, we briefly sketch the microscopic framework used to calculate fission fragment distributions. We then compute the spontaneous fission characteristics of $^{294}_{118}$Og$_{176}$. Finally, we study the robustness of our fission yield predictions with respect to different energy density functionals (EDFs), collective spaces, collective inertias, and dissipation.

Model – We calculate fission fragment distributions following the approach described in Ref.[22], which may be divided into two stages. In the first stage, we use the semiclassical WKB approximation to model spontaneous fission as quantum tunneling through a multidimensional potential energy surface (PES) characterized by $N$ collective coordinates $q \equiv (q_1, \ldots, q_N)$. In our implementation
of the WKB approximation, the most-probable tunneling path \( L(s) \) in the collective space is found via minimization of the collective action

\[
S(L) = \frac{1}{\hbar} \int_{s_{\text{in}}}^{s_{\text{out}}} \sqrt{2 \mathcal{M}(s)} (V(s) - E_0) \, ds,
\]

where \( s \) is the curvilinear coordinate along the path \( L \), \( \mathcal{M}(s) \) is the collective inertia \([39]\) and \( V(s) \) is the potential energy along \( L(s) \). \( E_0 \) stands for the collective ground-state energy. The dynamic programming method \([40]\) is employed to determine the path \( L(s) \). The calculation is repeated for different outer turning points, and each of these points is then assigned an exit probability \( P(s_{\text{out}}) = [1 + \exp (2S)]^{-1} \) \([41]\). In the second stage, fission trajectories begin from the outer turning line and then evolve along the PES according to the Langevin equations:

\[
\frac{dp_i}{dt} = -\frac{p_i p_k}{2} \frac{\partial}{\partial q_i} (\mathcal{M}^{-1})_{jk} - \frac{\partial V}{\partial q_i} - \eta_{ij} (\mathcal{M}^{-1})_{jk} p_k + g_{ij} \Gamma_j(t),
\]

\[
\frac{dq_i}{dt} = (\mathcal{M}^{-1})_{ij} p_j,
\]

where \( p_i \) is the collective momentum conjugate to \( q_i \). The dissipation tensor \( \eta_{ij} \) is related to the random force strength \( g_{ij} \) via the fluctuation-dissipation theorem, and \( \Gamma_j(t) \) is a Gaussian-distributed, time-dependent stochastic variable. All trajectories ending at a particular fragment configuration are weighted with the appropriate \( P(s_{\text{out}}) \). Particle number fluctuations in the neck at or near the scission line were accounted for by convoluting our Langevin yields with a Gaussian function of width \( \sigma_A = 6 \) for \( A \) and \( \sigma_Z = 4 \) for \( Z \).

The key ingredients in these calculations, \( V \) and \( \mathcal{M} \), are calculated self-consistently by solving the Hartree-Fock-Bogoliubov equations employing Skyrme and Gogny EDFs. To evaluate the robustness of our results with respect to different inputs, we perform calculations using three distinct EDFs: UNEDF1_HFB\([42]\), a Skyrme functional which was optimized to data for spherical and deformed nuclei, including fission isomers; SkM* \([43]\), another Skyrme functional designed for fission barriers and surface energy; and D1S \([44]\), a parametrization of the finite-range Gogny interaction fitted on fission barriers of actinides.

In self-consistent fission models, lowest multipole moments characterizing nuclear shape deformations are usually selected as collective coordinates. The remaining shape degrees of freedom are, in principle, decided through the energy minimization. In the present work, axial quadrupole moment \( Q_{20} \), triaxial quadrupole moment \( Q_{22} \), and axial octupole moment \( Q_{30} \) are considered as collective coordinates since the fission dynamics associated with fragment-yield distributions is mostly confined within this deformation space. Additionally, pairing correlations have a strong impact on the spontaneous fission half-lives calculated via action minimization \([19, 45, 46]\). It is taken into account through the coordinate \( \lambda_2 \) representing dynamic pairing fluctuations \([45]\). To obtain \( S(L) \), a dimensionless collective space is introduced as in Ref. \([45]\).

To balance computational speed with complexity, we used a different collective space for each functional. The most detailed calculation was carried out using UNEDF1_HFB in four dimensional space of \((Q_{20}, Q_{22}, Q_{30}, \lambda_2)\). Calculations were performed using the symmetry-unrestricted DFT solver HFODD \([47]\). To assure good convergence, we used the 1500 lowest single particle levels corresponding to 30 stretched harmonic oscillator shells.

The analysis of the four-dimensional (4D) PES showed that \( Q_{30} \) remains negligible through the first saddle point (up to at least \( Q_{20} = 100 \text{b} \)), and that \( Q_{22} \) and \( \lambda_2 \) are unimportant beyond the outer turning-point hyper-surface. This allowed us to simplify calculations with other functionals. The SkM* calculations were performed in a piecewise space \((Q_{20}, Q_{22}, \lambda_2)\) up to the fission isomer, \((Q_{20}, Q_{30}, \lambda_2)\) from fission isomer to outer turning points, and \((Q_{20}, Q_{30})\) beyond the outer turning-point line) with the same pairing properties as given in \([39, 45, 48]\), and the same HFODD basis as in UNEDF1_HFB calculations. In case of Gogny D1S calculations, a two-dimensional collective space described by coordinates \((Q_{20}, Q_{30})\) was used within the DFT solver HFBAxial \([49]\), where the stretched harmonic oscillator basis corresponding to 17 harmonic oscillator shells was optimized for each \((Q_{20}, Q_{30})\) value.

Several approximations are commonly used to compute the collective inertia, which describes the tendency of the nucleus to resist configuration changes. The most accurate prescription available till date is obtained from non-perturbative cranking approximation to the adiabatic time-dependent Hartree-Fock-Bogoliubov (ATDHFB) inertia \( (\mathcal{M}^A) \) \([50]\). On the other hand, perturbative expressions prioritize computational simplicity by sacrificing details of the level crossing dynamics, which results in a smoothed-out collective inertia. We also performed calculations using both the perturbative cranking ATDHFB inertia \( (\mathcal{M}^{\text{AP}}) \) \([50]\) and the perturbative GCM inertia \( (\mathcal{M}^{\text{GCM}}) \) \([15]\), which has the same structure as \( \mathcal{M}^{\text{AP}} \) but with an absolute magnitude quenched by a factor 1.5 \([51]\). We additionally did calculations using a constant inertia parameter \( \mathcal{M}^c \).

In this work, we set the dissipation strength \( \eta_{ij} \) as an adjustable parameter rather than using some of the common prescriptions \([52, 53]\), which have not yet been adapted to DFT inputs. We examined the sensitivity of our calculations to \( \eta_{ij} \) by varying it around the baseline value used in \([22]\): \( \eta_0 \equiv (\eta_{22}, \eta_{23}, \eta_{33}) \equiv (50 \hbar, 5 \hbar, 40 \hbar) \).

Results – We first compare in Fig. 1 the two-dimensional PESs in the \((Q_{20}, Q_{30})\) plane for the functionals UNEDF1_HFB, D1S, and SkM*, and D1S. These
we notice that the overall topology of the PES is roughly similar in all models, with a symmetric saddle point occurring around $Q_{20} \approx 40$ b, a second barrier beginning around $Q_{20} \approx 100 - 120$ b along the symmetric fission path, presence of local minima at large deformations, a deep valley that leads to an highly-asymmetric split, and the secondary less-asymmetric fission valley that emerges at large elongations.

But there are differences as well, such as the height of the first saddle point, the depth of the highly-asymmetric fission valley, and the height of the ridge separating two fission valleys. As a result, the outer turning points are pushed to larger elongations in D1S and SkM* as compared to UNEDF1_HFB. These differences in the PES topology strongly affect the predicted spontaneous fission half-lives $\tau_{SF}$, which in the case of UNEDF1_HFB, SkM* and D1S are $9.1 \times 10^{-9}$ s, $4.0 \times 10^{-5}$ s and $3.2 \times 10^{-2}$ s, respectively (see also [13, 17] for a detailed discussion of half-lives). These large variations of $\tau_{s}$ reflect the well-known sensitivity of spontaneous fission half-lives to changes in the quantities entering the collective action (1). The predictions of UNEDF1_HFB and, to a lesser degree, SkM* for $\tau_{s}$ are clearly incompatible with experiment, as $^{294}$Og is known to decay by $\alpha$-decay to a half-life of 0.58 ms [3]. However, as we demonstrate below, fission yields predicted in all three models are very similar.

Despite the strong variations in the predicted $\tau_{SF}$, we see in Fig. 2 that the predicted fission yields are in fact rather independent of the EDF choice. Namely, all three functionals predict a heavy fragment in the neighborhood of $^{208}$Pb_{126} and a light fragment near $^{86}$Kr_{50}. In addition functional SkM* predicts a small probability associated with lighter fragments around $^{126}$Xe_{72}.

The sensitivity to the different inputs are shown in Fig. 3 through one-dimensional projections on the fragment mass and charge. The top panels of Fig. 3 shows again that all three functionals predict highly-asymmetric fission with the heavy fragment centered at or around $^{208}$Pb_{126}. While the peaks corresponding to the D1S and SkM* functionals overlap quite well, the UNEDF1_HFB peak is broader and shifted slightly towards the more asymmetric splits. This may be related to the relative flatness of the UNEDF1_HFB PES compared to the others, which makes it more susceptible to large fluctuations. The secondary peak around $^{126}$Xe_{72} predicted by SkM*, associated with the more symmetric fission valley of Fig. 1(c) is clearly seen. For D1S and UNEDF1_HFB, the yield distributions do not show a tail at lower masses/charges. As discussed below, this can be associated with the collective inertia and the energy ridge (particularly pronounced for D1S), both effectively
separating the two fission valleys.

The bottom panels of Fig. 3 show the effect of varying the strength of the dissipation tensor. This parameter has a noticeable impact on the yields, particularly on the tails and the yields associated with the more-symmetric channel. The results corresponding to $\eta = \eta_0, 0.5\eta_0,$ and $2\eta_0$ are very close. This is consistent with findings of Refs. [56–58], which found that the yield distributions are not very sensitive to the precise value of dissipation tensor. In the no-dissipation variant ($\eta_{ij} = g_{ij} = 0$), the Langevin equations become deterministic and there is a one-to-one correspondence between outer turning points and scission points. In this variant, one predicts an increased contribution arising from more-symmetric fission valley. However, since this variant ignores near-scission fluctuations that are essential for the proper description of the width of the fission yields [57–59], we do not consider it to be realistic: we show it for reference only. In general, irrespective of the choice of energy density functional, we found a rather similar pattern of yield distributions with respect to the inertia tensor (except for $M^c$) and the dissipation strength.

Finally, to better understand the formation of the most-probable fission fragments, we studied the nucleon localization functions [58, 60] along the cluster-decay path. By comparing the $^{294}_{174}$Og\textsubscript{176} localizations with those belonging to $^{208}_{82}$Pb\textsubscript{126} and $^{86}_{36}$Kr\textsubscript{50}, we found that both the lead prefragment and the $N \approx 50$ neutrons belonging to krypton are well-localized early in the evolution, shortly beyond the outer turning line. This result highlights the importance of shell closures in determining the most probable fragment configuration in fissioning nuclei.

**Conclusion** – In this letter, we predict that the dominant spontaneous fission mode of $^{294}_{174}$Og\textsubscript{176} will be a highly-asymmetric cluster emission centered around the doubly-magic $^{208}_{82}$Pb\textsubscript{126} and magic $^{86}_{36}$Kr\textsubscript{50}. We have shown that this prediction is fairly robust with respect to the choice of input parameters, such as energy density functional, collective inertia, and dissipation tensor. In particular, we emphasize that differences in barrier heights predicted by different EDFs do not affect the calculated fission yields. We confirmed the implicit assumption of [22], that 4D calculations do not necessarily offer an improved description of the tunneling compared to a well-chosen 3D description, and we argue for a hierarchy of ingredients necessary for a Langevin description of low-energy fission. This work reinforces the conclusions of Ref. [23]: in future superheavy element searches, the range of expected fission fragments should take into account the possibility of cluster emission, which would lead to heavier fragments than those which appear in actinide fission.
ACKNOWLEDGEMENTS

This work was supported by the U.S. Department of Energy under Award Numbers DOE-DE-NA0002847 (NNSA, the Stewardship Science Academic Alliances program), DE-SC0013365 (Office of Science), and DE-SC0018083 (Office of Science, NUCLEI SciDAC-4 collaboration), as well as by the Office of Science, Office of Workforce Development for Teachers and Scientists, Office of Science Graduate Student Research (SCGSR) program. The SCGSR program is administered by the Oak Ridge Institute for Science and Education (ORISE) for the DOE. ORISE is managed by ORAU under contract number DE-SC0014664. This work was also partly performed under the auspices of the US Department of Energy by the Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Computing support came from the Lawrence Livermore National Laboratory (LLNL) Institutional Computing Grand Challenge program.

[1] Y. T. Oganessian et al., Phys. Rev. C 74, 044602 (2006).
[2] Y. T. Oganessian et al., Phys. Rev. Lett. 109, 162501 (2012).
[3] N. Brewer et al., Phys. Rev. C 98, 024317 (2018).
[4] S. Dmitriev, M. Itkis, and Y. Oganessian, Eur. Phys. J. WOC 131, 08001 (2016).
[5] Y. T. Oganessian and S. N. Dmitriev, Russ. Chem. Rev. 85, 901 (2016).
[6] S. Hofmann et al., Eur. Phys. J. A 52, 180 (2016).
[7] J. B. Roberto and K. P. Rykaczewski, Sep. Sci. Technol. 53, 1813 (2018).
[8] C. E. D¨ullmann and M. Block, Sci. Am. 318, 48 (2018).
[9] P. Jerabek, B. Schuetrumpf, P. Schwerdtfeger, and W. Nazarewicz, Phys. Rev. Lett. 120, 5 (2018).
[10] W. Nazarewicz, Nature Phys. 14, 537 (2018).
[11] S. A. Giuliani, Z. Matheson, W. Nazarewicz, E. Olsen, P.-G. Reinhard, J. Sadhukhan, B. Schuetrumpf, N. Schunck, and P. Schwerdtfeger, Rev. Mod. Phys. (2018).
[12] F. P. Heßberger, Euro. Phys. J. A 53, 75 (2017).
[13] A. Baran, M. Kowal, P. G. Reinhard, L. M. Robledo, A. Staszczak, and M. Warda, Nucl. Phys. A 944, 442 (2015).
[14] S. A. Giuliani, G. Martínez-Pinedo, and L. M. Robledo, Phys. Rev. C 97, 034323 (2018).
[15] N. Schunck and L. M. Robledo, Rep. Prog. Phys. 79, 116301 (2016).
[16] J. Erler, K. Langanke, H. P. Loens, G. Martínez-Pinedo, and P.-G. Reinhard, Phys. Rev. C 85, 025802 (2012).
[17] A. Staszczak, A. Baran, and W. Nazarewicz, Phys. Rev. C 87, 024320 (2013).
[18] S. A. Giuliani and L. M. Robledo, Phys. Rev. C 88, 054325 (2013).
[19] S. A. Giuliani, L. M. Robledo, and R. Rodríguez-Guzmán, Phys. Rev. C 90, 054311 (2014).
[20] J.-F. Lemaitre, S. Goriely, S. Hilaire, and N. Dubray, Phys. Rev. C 98, 024623 (2018).
[21] R. Rodríguez-Guzmán and L. M. Robledo, Phys. Rev. C 98, 034308 (2018).
[22] J. Sadhukhan, W. Nazarewicz, and N. Schunck, Phys. Rev. C 93, 011304 (2016).
[23] M. Warda, A. Zidleh, and L. M. Robledo, Phys. Rev. C 98, 041602 (2018).
[24] D. Regnier, N. Dubray, N. Schunck, and M. Verrière, Phys. Rev. C 93, 054611 (2016).
[25] D. Regnier, N. Dubray, and N. Schunck, (2018), arXiv:1810.08402 [nucl-th].
[26] A. Sandulescu, D. Poenaru, and W. Greiner, Sov. J. Part. Nuclei 11, 528 (1980).
[27] D. Poenaru, W. Greiner, K. Depta, M. Ivascu, D. Mazilu, and A. Sandulescu, At. Data Nucl. Data Tables 34, 423 (1986).
[28] G. Royer, R. K. Gupta, and V. Denisov, Nucl. Phys. A 632, 275 (1998).
[29] H. J. Rose and G. A. Jones, Nature 307, 245247 (1983).
[30] D. N. Poenaru and W. Greiner, in Clusters in Nuclei: Volume 1, edited by C. Beck (Springer Berlin Heidelberg, Berlin, Heidelberg, 2010) p. 1.
[31] D. N. Poenaru, R. A. Gherghescu, and W. Greiner, Phys. Rev. Lett. 107, 062503 (2011).
[32] D. N. Poenaru, R. A. Gherghescu, and W. Greiner, Phys. Rev. C 85, 034615 (2012).
[33] D. N. Poenaru, R. A. Gherghescu, and W. Greiner, J. Phys. Conf. Ser. 436, 012056 (2013).
[34] D. N. Poenaru, R. A. Gherghescu, W. Greiner, and N. S. Shakib, in Nuclear Physics: Present and Future (Springer International Publishing, 2015) p. 131.
[35] D. N. Poenaru and R. A. Gherghescu, Phys. Rev. C 97, 044621 (2018).
[36] K. P. Santhosh and C. Nithya, Phys. Rev. C 97, 064616 (2018).
[37] Y. L. Zhang and Y. Z. Wang, Phys. Rev. C 97, 014318 (2018).
[38] M. Warda and L. M. Robledo, Phys. Rev. C 84, 044608 (2011).
[39] J. Sadhukhan, K. Mazurek, A. Baran, J. Dobaczewski, W. Nazarewicz, and J. A. Sheikh, Phys. Rev. C 88, 064314 (2013).
[40] A. Baran, K. Pomorski, A. Łukasiak, and A. Sobiczewski, Nucl. Phys. A 361, 83 (1981).
[41] A. Baran, Phys. Lett. B 76, 8 (1978).
[42] N. Schunck, J. D. McDonnell, J. Sarich, S. M. Wild, and D. Higdon, J. Phys. G 42, 34024 (2015).
[43] J. Bartel, P. Quentin, M. Brack, C. Guet, and H.-B. Häkansson, Nucl. Phys. A 386, 79 (1982).
[44] J. Berger, M. Girod, and D. Gogny, Nucl. Phys. A 502, 85 (1989).
[45] J. Sadhukhan, J. Dobaczewski, W. Nazarewicz, J. A. Sheikh, and A. Baran, Phys. Rev. C 90, 061304 (2014).
[46] J. Zhao, B.-N. Lu, T. Nikšić, D. Vretenar, and S.-G. N. S. Shakib, in Nuclear Physics: Present and Future (Springer Berlin Heidelberg, Berlin, Heidelberg, 2010) p. 1.
[47] D. N. Poenaru and R. A. Gherghescu, Phys. Rev. C 97, 044621 (2018).
[48] K. P. Santhosh and C. Nithya, Phys. Rev. C 97, 064616 (2018).
[49] Y. L. Zhang and Y. Z. Wang, Phys. Rev. C 97, 014318 (2018).
[50] M. Warda and L. M. Robledo, Phys. Rev. C 84, 044608 (2011).
[51] S. A. Giuliani and L. M. Robledo, Phys. Lett. B 787, 134 (2018).
[52] M. D. Usang, F. A. Ivanyuk, C. Ishizuka, and S. Chiba, Phys. Rev. C 96, 064617 (2017).
[53] C. Ishizuka, M. D. Usang, F. A. Ivanyuk, J. A. Maruhn, K. Nishio, and S. Chiba, Phys. Rev. C 96, 064616 (2017).
[54] Interactive Chart of Nuclides, NuDat 2.7 https://www.nndc.bnl.gov/nudat2/.
[55] A. Bulgac, S. Jin, K. Roche, N. Schunck, and I. Stetcu, ArXiv e-prints (2018), arXiv:1806.00694 [nucl-th].
[56] J. Randrup, P. Möller, and A. J. Sierk, Phys. Rev. C 84, 034613 (2011).
[57] A. J. Sierk, Phys. Rev. C 96, 034603 (2017).
[58] J. Sadhukhan, C. Zhang, W. Nazarewicz, and N. Schunck, Phys. Rev. C 061301, 1 (2017).
[59] Y. Aritomo, S. Chiba, and F. Ivanyuk, Phys. Rev. C 90, 054609 (2014).
[60] C. L. Zhang, B. Schuetrumpf, and W. Nazarewicz, Phys. Rev. C 94, 1 (2016).