

# Synthesis of Heavy and Superheavy Neutron-Rich Nuclei in Multinucleon Transfer Reactions at Grazing Angle Using Uranium Beams

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## Abstract

Information on the heaviest elements has been obtained up to now via fusion-evaporation reactions. It is however well known that the only nuclei one can reach using fusion-evaporation reactions are neutron deficient and moreover in a very limited number due to the restricted number of beam-target combinations. An alternative to fusion-evaporation could be deep-inelastic collisions. Theoretical calculations predict large cross-sections for neutron-rich heavy elements production close to zero degrees and at grazing angles [1,2]. We recently performed an experiment at Argonne National Laboratory using a  $^{136}\text{Xe}$  beam on a  $^{238}\text{U}$  target. The preliminary results are promising. The goal of this proposal is to investigate deep inelastic reaction mechanisms in the heavy elements' region using the AGATA germanium array and DANTE detector coupled to the PRISMA separator. Using a  $^{238}\text{U}$  beam on a  $^{64}\text{Ni}$  target at grazing angles. Such an experiment would pave the way for future synthesis on the synthesis of new superheavy neutron-rich isotopes.

## 1 Motivation

One of the frontiers of modern nuclear physics is the production of undiscovered neutron-rich nuclei far from the  $\beta$ -stability line in the heavy-mass range. Although 3327 nuclides have

been discovered experimentally so far, they are still far from the theoretical prediction of about 8000-10000. On the neutron-rich side, in the north-eastern region of the nuclide map, lies the a vast and yet unexplored area. The synthesis of these unknown nuclide is important for the understanding of nucleosynthesis in nuclear astrophysics and to unravel the mystery of the origin of the heavy elements from iron to uranium in the universe.

Motivated by the attempt to populate neutron-rich isotopes around  $N = 126$ , experimental research on the Multinucleon Transfer (MNT) reaction of  $^{136}\text{Xe} + ^{198}\text{Pt}$  has been carried out at GANIL [3]. For neutron-rich species the MNT reaction has a great advantage in production cross sections over the projectile fragmentation reaction mechanism. At GSI, the deep inelastic MNT reaction of  $^{48}\text{Ca} + ^{248}\text{Cm}$  helped to identify five new neutron deficient isotopes with  $Z > 92$  [4–7].

During the last few years, we have been heavily involved in MNT reaction experiments using different beam-projectile combinations with promising results. At GANIL with VAMOS using the reaction  $^{238}\text{U} + ^{238}\text{U}$  (see Ph.D. thesis of A. Utegov) and at ANL with the gas-filled separator AGFA coupled to the GAMMSPHERE germanium detector array using  $^{136}\text{U}$  beam on  $^{238}\text{U}$  target and detecting the reaction products at 0 degree (see Ph.D. thesis of J. Bequet to be published in September 2025).

Regarding the best reaction to produce neutron-rich nuclei, the various models differ in their results concerning the optimal projectile-target combination. The Di-Nuclear System (DNS) model suggests the use of intermediate-heavy neutron-rich projectiles such as  $^{48}\text{Ca}$  combined with heavy targets such as Pu, Cm or Cf isotopes. Collisions of two very heavy nuclei are found to be unsuitable in this model because of very short interaction times that do not allow massive nucleon transfer. In comparison, the Langevin model suggests systems like  $^{238}\text{U} + ^{250}\text{Cf}$  or  $^{238}\text{U} + ^{248}\text{Cm}$  [8]. Experimental data confirms that MNT reactions with more neutrons in the colliding system shift the isotopic distribution of the MNT products towards the neutron-rich side of the chart of nuclides (Segré chart) [8]. Therefore, heavy neutron-rich beams such as  $^{136}\text{Xe}$ , or heavier beams such as  $^{238}\text{U}$ , appear to be more favorable.

As mentioned above, experimental and theoretical efforts have revealed that the distributions of transfer products in the reaction system with more neutrons tend to be on the neutron-rich side of the Segré chart. The transfer cross sections strongly depend on the incident energy in low-energy MNT reactions. The choice of beam energy is not an easy one. Near the Coulomb Barrier, the reaction is governed only by Quasi-elastic scattering mechanism, with the products emitted near the Grazing angle. Higher in incident energy, the Quasi-elastic is still present near the Grazing angle, but the deep-inelastic part of the mechanism will start to be present at other scattering angles [9]. At higher incident energy the neutron enrichment will be less important due to the enhanced evaporation, but the extra energy will allow more dumped collisions, thus enlarging the range of exit channels, allowing to exchange more nucleons [10, 11]. Exploring an optimal incident energy provides important guidance for the experiment planning.

This proposal has to be interpreted as a part of a more extended program aiming at the understanding of the different steps of MNT reactions, which is essential for their successful application in the synthesis of new isotopes, employing different projectile-target combinations at different angles and energies with the goal of:

- Producing known isotopes of interest and measuring their properties (masses, half-lives,

isomers, decays etc.)

- Production of new exotic isotopes and measurement of their properties.

Based on our past experience, it has become clear that the ideal experimental setup for observing new heavy and exotic MNT products would require a large angular acceptance, allowing MNT products to be produced with cross sections down to the picobarn scale. For this reason, PRISMA, with its detection systems coupled with AGATA is an ideal setup to perform such measurements. Successfully experiments at LNL has been already performed in the actinide’s region [12–14].

With our experiment, we hope to open up a new avenue to tackle these long-standing questions and to provide new experimental data that will be needed for comparison with calculations provided by different theoretical models [8, 15, 16]. In such an experiment nucleus in the region of octupole deformations could be produced in relative high statistics. In such an experiment profiting of Agata germanium array, we also address important question concerning the structure of nuclei around Pu, Am and Cm. We intend to study the collectivity of such a nuclei. How do collective properties evolve near the predicted octupole-deformed region and population patterns of heavy neutron-rich isotopes as a function of mass and charge transfer.

## 2 Proposed experiment

We propose to investigate the production cross sections of a variety of neutron-rich nuclei in the  $Z = 90-98$  range produced in multinucleon transfer reactions using a uranium beam at 7.2 MeV/A and a target of  $^{64}\text{Ni}$  0.3 mg thick. Theoretical calculations using the codes GRAZING (courtesy of M. Siciliano) and DIT [17] coupled with Gemini++ [18] (courtesy of J. Bequet and I. Stefan)<sup>1</sup>, agree that the production cross-section for neutron-rich nuclei could be of the order of several hundred nb to several mb (e.g.  $(^{240}\text{Pu})$  is estimated to be 1 mb). DIT is used to simulate the MNT mechanism and Gemini++ is used for the evaporation and fission of the quasi-projectiles and quasi-targets nuclei.

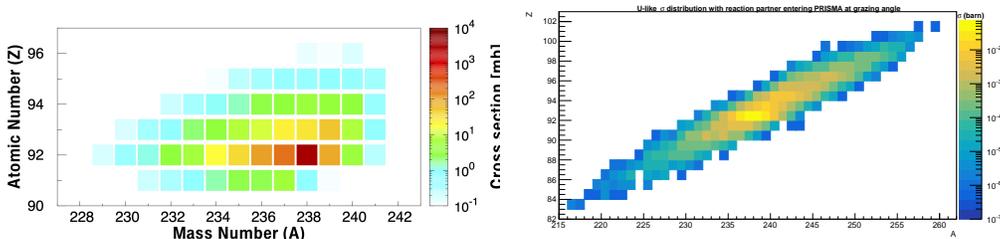


Figure 1: GRAZING (left) and DIT + Gemini++ (right) calculation for the  $^{238}\text{U}$  on  $^{64}\text{Ni}$ .

The isotopes of elements  $92 < Z < 98$  will be identified using the PRISMA spectrometer. The  $^{238}\text{U}$  beam will be delivered by the PIAVE-ALPI accelerator with an intensity of 1 pnA. The energy has been chosen to favor the largest exchange of nucleons between the beam and the target, to be in what is called the deep-inelastic regime. The target-like

reaction fragments in the Ni region will be identified with the magnetic mass spectrometer PRISMA placed around the grazing angle. We will, therefore, study the neutron rich heavy MNT products (U-like events) by gating on the binary partner  $^{64}\text{Ni}$  identified in PRISMA. The  $40 \times 60 \text{ mm}^2$  microchannel plate detector DANTE could be envisaged to be used in the target chamber, enabling the measurements of the Uranium like nuclei, allowing a kinematic coincidence between the different reaction products. The choice of the indirect reaction allows the heavy nuclei to have a narrower angular distribution, enabling the search for products at lower cross-sections.

The choice of inverse kinematics will allow the practical elimination of elastic scattered uranium from the detection system ( $< 18^\circ$ ), a great advantage from the detection point of view. Added to this, the fission fragments will not enter PRISMA.

In order to estimate the best angle at which we could measure the highest cross section for the neutron rich heavy multinucleon transfer nuclei, simulations using DIT coupled with Gemini++ code has been performed. In the spectrum 2 right sided is plotted the angular distribution of the light nuclei in function of their mass number, in the Z axis the rates/min are shown. Quasi-target nuclei associated with neutron-rich Uranium like events are produced with the best cross section at angles around 44-56 degree. The angular acceptance of PRISMA is shown with red lines. The corresponding cross section of the heavy nuclei we intend to study using AGATA germanium array is plotted in 2 left side.

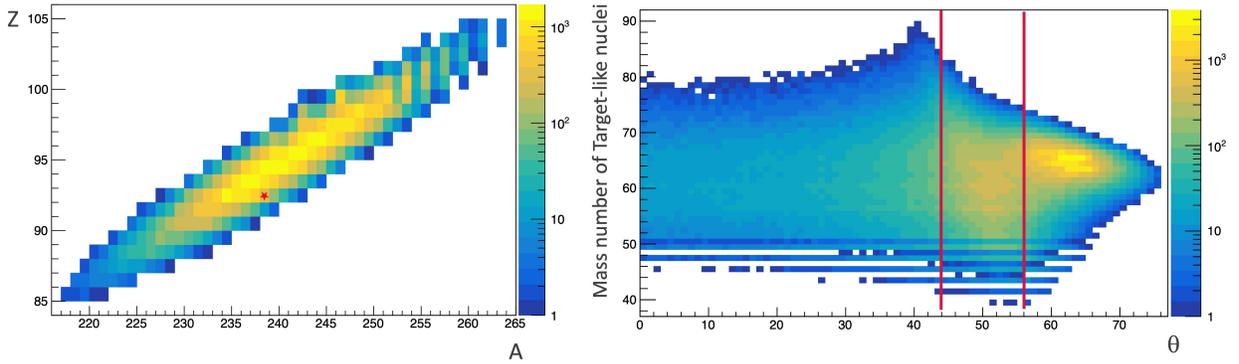


Figure 2: (Left) Rate/min of U-like events considering the Ni-like events entering in PRISMA (considering the angular acceptance of PRISMA). (Right) Angular distribution of U-like events: mass distribution of U-like events(Y axis) vs emission angle, in Rate/min (Z axis) vs Rate/min (Z axis). Simulation has been performed with DIT and Gemini++.

We plan to couple PRISMA with the Agata array where the rays from the excited states in both beam-like and target-like nuclei will be detected. The target-like products will leave the target with an energy of 6.3 MeV/A, will pass through PRISMA to eventually be analyzed in the focal plane detector system which is composed of a MWPPAC divided into 10 sections and an array of  $10 \times 4$  segmented  $\text{CH}_4$  gas-filled transverse-field multiparametric ionization chambers (IC). Each segment of the IC acts as DE section and provides a signal proportional to the energy loss of the passing fragment. The detection system provides all the necessary information for complete Z and A identification of the analyzed reaction product.

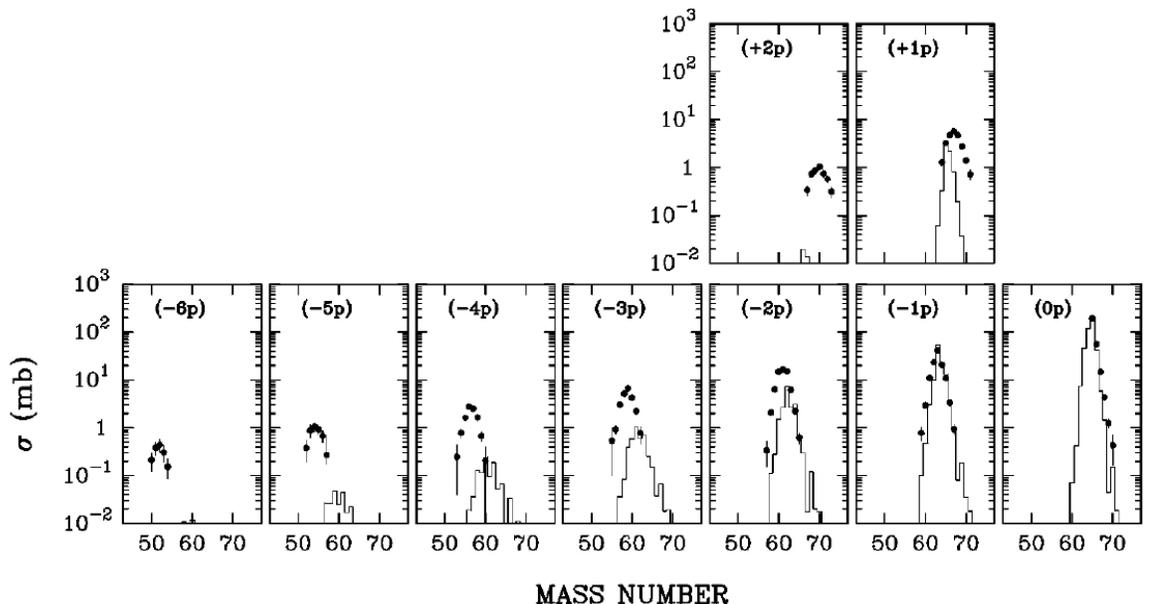


Figure 3: Experimental (points) and calculated (histogram) angle and Q-value integrated cross section for the indicated transfer products.

As demonstrated in earlier work by Corradi et al. [14], see Fig. 3, the  $^{238}\text{U} + ^{64}\text{Ni}$  reaction populates a wide variety of neutron-rich nuclei through proton-stripping and neutron-pickup channels. These include heavy reaction partners such as +1p (Np), +2p (Pu), +3p (Am), and +4p (Cm). With the current availability of AGATA coupled to PRISMA at Legnaro, we propose to perform high-resolution  $\gamma$ -ray spectroscopy of these nuclei with unprecedented precision. AGATA’s advanced  $\gamma$ -ray tracking, superior Doppler correction, and fine angular resolution are crucial for resolving weak and closely spaced transitions in heavy, fast-recoiling nuclei, particularly under inverse kinematics.

Importantly, this setup also enables the identification of long-lived fission isomers in the actinide region. These isomers provide unique insight into the structure of highly deformed nuclear states and the multi-humped shape of the fission barrier. Their long lifetimes—often in the microsecond range—indicate the presence of secondary minima associated with superdeformed or hyperdeformed configurations. Through  $\gamma$ -ray spectroscopy, we can probe their collective properties, decay modes, and tunneling dynamics, offering essential constraints for theoretical models of nuclear deformation and fission in heavy, neutron-rich systems.

In table 1 shows a few examples of nuclei expected to be produced in the MNT reaction  $^{238}\text{U} + ^{64}\text{Ni}$  with respect to their decay mode, half-life, production cross-section and counting rates, assuming a beam intensity of 1 pA and PRISMA angular acceptance.

To achieve the proposed goals of detailed spectroscopy of neutron-rich actinides, we request **10 days of beam time**. Based on DIT simulations in agreement with GRAZING, and after accounting for AGATA’s 15% photopeak efficiency and PRISMA efficiency of 2515%

Nucleus	Decay Mode	$T_{1/2}$	Cross-section (mb)	$\gamma$ -ray/min
$^{236}\text{U}$	$\alpha$	$2.34 \times 10^7$ y	0.69	772
$^{239}\text{U}$	$\beta^-$	23.45 min	0.74	82
$^{240}\text{Np}$	$\beta^-$	7.22 min	0.21	238
$^{241}\text{Np}$	$\beta^-$	13.9 min	0.91	101
$^{240}\text{Pu}$	$\alpha$	6561 y	0.9	1029

Table 1: Examples of production cross-sections and calculated counting rates expected at the target (given in particles/min) considering a beam intensity of 1 pnA.

percent, we expect for example in the case of  $^{240}\text{Pu}$  a  $\gamma$ -ray detection rates of approximately 39  $\gamma$ /min. These rates will enable the reconstruction of level schemes, identification of collective excitations, and lifetime measurements via Doppler-based techniques. The extended duration is essential not only for accumulating sufficient statistics on the weaker channels, but also to allow PRISMA settings to be optimized for mass and charge identification across different Z and A. Given AGATA’s temporary presence at Legnaro, this experiment represents a timely opportunity to collect high-resolution spectroscopic data on nuclei that are otherwise difficult to access.

### 3 Summary

With the current availability of AGATA coupled to PRISMA at Legnaro, together with new uranium beams, we have a unique opportunity to study multinucleon transfer (MNT) reactions as an effective pathway to access exotic heavy nuclei. This experiment will focus on two regions of the nuclear chart where experimental data remain scarce. The first is the region of neutron-rich superheavy nuclei, where theoretical models predict the next spherical shell closures beyond  $^{208}\text{Pb}$ , at proton numbers  $Z=114$ , 120, or 126 and neutron number  $N=184$ . While such nuclei are out of reach using fusion reactions with stable projectiles, they may become accessible through MNT reactions using  $^{238}\text{U}$  projectiles on actinide targets. The advantage of MNT lies in its ability to populate a wide range of isotopes in a single experimental setting, thanks to broad excitation functions and wide distributions in Z and A.

Before moving toward this unexplored territory, however, it is essential to understand the underlying reaction mechanisms in detail. In this context, we propose to use the AGATA PRISMA setup to study the production cross sections of neutron-rich nuclei in the  $Z=92\text{--}98$  region via the reaction  $^{238}\text{U} + ^{64}\text{Ni}$ . The results will be compared to predictions from state-of-the-art theoretical models. Compared to previous experimental configurations, AGATA PRISMA offers significantly improved resolution and selectivity, enabling detailed spectroscopic investigations of weakly populated and previously inaccessible nuclei.

Importantly, AGATA’s advanced timing and tracking capabilities allow for precise lifetime measurements using Doppler-based techniques, enabling the extraction of collective properties such as  $B(E2)$ ,  $B(E3)$ , and  $\rho^2(E0)$ . These measurements will provide critical insight into the structure and decay of fission isomers and will help constrain theoretical models of fission barriers in the actinide region. Given the temporary presence of AGATA

at Legnaro, this is a timely and scientifically valuable opportunity that should be exploited without delay.

A beam energy of 7.2 MeV/u with an intensity of 1 pnA and a target thickness of 0.3 mg/cm<sup>2</sup> are required to accumulate the needed number of counts in a requested irradiation time of 10 days beam on target.

## References

- [1] A. V. Karpov and V. V. Saiko. Modeling near-barrier collisions of heavy ions based on a langevin-type approach. *Phys. Rev. C*, 96:024618, Aug 2017.
- [2] V.I. Zagrebaev and W. Greiner. Cross sections for the production of superheavy nuclei. *Nuclear Physics A*, 944:257 – 307, 2015. Special Issue on Superheavy Elements.
- [3] Y.X. Watanabe, Y. Hirayama, N. Imai, H. Ishiyama, S.C. Jeong, H. Miyatake, E. Clement, G. de France, A. Navin, M. Rejmund, C. Schmitt, G. Pollarolo, L. Corradi, E. Fioretto, D. Montanari, S.H. Choi, Y.H. Kim, J.S. Song, M. Niikura, D. Suzuki, H. Nishibata, and J. Takatsu. Study of collisions of <sup>136</sup>Xe+<sup>198</sup>Pt for the kek isotope separator. *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, 317:752 – 755, 2013. XVIth International Conference on ElectroMagnetic Isotope Separators and Techniques Related to their Applications, December 2–7, 2012 at Matsue, Japan.
- [4] S. Heinz, H. M. Devaraja, O. Beliuskina, V. Comas, S. Hofmann, C. Hornung, G. Münzenberg, D. Ackermann, M. Gupta, R. A. Henderson, F. P. Heßberger, B. Kindler, B. Lommel, R. Mann, J. Maurer, K. J. Moody, K. Nishio, A. G. Popeko, D. A. Shaughnessy, M. A. Stoyer, and A. V. Yeremin. Synthesis of new transuranium isotopes in multinucleon transfer reactions using a velocity filter. *The European Physical Journal A*, 52(9):278, 2016.
- [5] H.M. Devaraja, S. Heinz, O. Beliuskina, V. Comas, S. Hofmann, C. Hornung, G. Münzenberg, K. Nishio, D. Ackermann, Y.K. Gambhir, M. Gupta, R.A. Henderson, F.P. Heßberger, J. Khuyagbaatar, B. Kindler, B. Lommel, K.J. Moody, J. Maurer, R. Mann, A.G. Popeko, D.A. Shaughnessy, M.A. Stoyer, and A.V. Yeremin. Observation of new neutron-deficient isotopes with z92 in multinucleon transfer reactions. *Physics Letters B*, 748:199 – 203, 2015.
- [6] E. Jäger E. Schimpf M. Schödel C. Mühle F. Klos A. Trler A. Yakushev A. Belov T. Belyakova M. Kaparkova V. Kukhtin E. Lamzin A. Semchenkov, W. Bröchle and S. Sytchevsky. The transactinide separator and chemistry apparatus (tasca) at gsi - optimization of ion-optical structures and magnet designs. *Nucl. Instr. and Meth. B*, 266:4153, 2008.
- [7] H.M. Devaraja, A.V. Yeremin, M.L. Chelnokov, V.I. Chepigin, S. Heinz, A.V. Isaev, I.N. Izosimov, Sh.A. Kalandarov, A.V. Karpov, D.E. Katrasev, A.A. Kuznetsova, O.N. Malyshev, R.S. Mukhin, A.G. Popeko, Yu.A. Popov, V.V. Saiko, B. Sailaubekov,

- E.A. Sokol, A.I. Svirikhin, M.S. Tezekbayeva, U.A. Abitayeva, E.K. Almanbetova, A.A. Almas, A.K. Azhibekov, M.A. Bychkov, O. Dorvaux, B. Gall, K. Hauschild, K. Kessaci, A. Lopez-Martens, E.V. Mardyban, K. Mendibayev, Zh.Ye. Nakypbek, and B.A. Urazbekov. Systematic studies to produce heavy above-target nuclides in multinucleon transfer reactions. *Physics Letters B*, 862:139353, 2025.
- [8] V. I. Zagrebaev, Yu. Ts. Oganessian, M. G. Itkis, and Walter Greiner. Superheavy nuclei and quasi-atoms produced in collisions of transuranium ions. *Phys. Rev. C*, 73:031602, Mar 2006.
- [9] J. Wilczyński. Nuclear molecules and nuclear friction. *Physics Letters B*, 47(6):484–486, 1973.
- [10] I. Stefan, B. Fornal, S. Leoni, F. Azaiez, C. Portail, J.C. Thomas, A.V. Karpov, D. Ackermann, P. Bednarczyk, Y. Blumenfeld, S. Calinescu, A. Chbihi, M. Ciemala, N. Cieplicka-Oryńczak, F.C.L. Crespi, S. Franchoo, F. Hammache, L.W. Iskra, B. Jacquot, R.V.F. Janssens, O. Kamalou, T. Lauritsen, M. Lewitowicz, L. Olivier, S.M. Lukyanov, M. Maccormick, A. Maj, P. Marini, I. Matea, M.A. Naumenko, F. de Oliveira Santos, C. Petrone, Yu.E. Penionzhkevich, F. Rotaru, H. Savajols, O. Sorlin, M. Stanoiu, B. Szpak, O.B. Tarasov, and D. Verney. Neutron-rich nuclei produced at zero degrees in damped collisions induced by a beam of 180 on a 238u target. *Physics Letters B*, 779:456–459, 2018.
- [11] W. U. Schröder and J. R. Huizenga. *Damped Nuclear Reactions*, pages 113–726. Springer US, Boston, MA, 1985.
- [12] A. Vogt, B. Birkenbach, P. Reiter, L. Corradi, T. Mijatović, D. Montanari, S. Szilner, D. Bazzacco, M. Bowry, A. Bracco, B. Bruyneel, F. C. L. Crespi, G. de Angelis, P. Désesquelles, J. Eberth, E. Farnea, E. Fioretto, A. Gadea, K. Geibel, A. Gengelbach, A. Giaz, A. Görgen, A. Gottardo, J. Grebosz, H. Hess, P. R. John, J. Jolie, D. S. Judson, A. Jungclaus, W. Korten, S. Leoni, S. Lunardi, R. Menegazzo, D. Mengoni, C. Michelagnoli, G. Montagnoli, D. Napoli, L. Pellegrini, G. Pollarolo, A. Pullia, B. Quintana, F. Radeck, F. Recchia, D. Rosso, E. Şahin, M. D. Salsac, F. Scarlassara, P.-A. Söderström, A. M. Stefanini, T. Steinbach, O. Stezowski, B. Szpak, Ch. Theisen, C. Ur, J. J. Valiente-Dobón, V. Vandone, and A. Wiens. Light and heavy transfer products in  $^{136}\text{Xe} + ^{238}\text{U}$  multinucleon transfer reactions. *Phys. Rev. C*, 92:024619, Aug 2015.
- [13] F. Galtarossa, L. Corradi, S. Szilner, E. Fioretto, G. Pollarolo, T. Mijatović, D. Montanari, D. Ackermann, D. Bourgin, S. Courtin, G. Fruet, A. Goasduff, J. Grebosz, F. Haas, D. Jelavić Malenica, S. C. Jeong, H. M. Jia, P. R. John, D. Mengoni, M. Milin, G. Montagnoli, F. Scarlassara, N. Skukan, N. Soić, A. M. Stefanini, E. Strano, V. Tokić, C. A. Ur, J. J. Valiente-Dobón, and Y. X. Watanabe. Mass correlation between light and heavy reaction products in multinucleon transfer  $^{197}\text{Au} + ^{130}\text{Te}$  collisions. *Phys. Rev. C*, 97:054606, May 2018.
- [14] L. Corradi, S. Szilner, G. Pollarolo, and et al. Multinucleon transfer processes in heavy-ion collisions. *Journal of Physics G: Nuclear and Particle Physics*, 36:113101, 2009.

- [15] H. L. Liu, F. R. Xu, P. M. Walker, and C. A. Bertulani. Effects of high-order deformation on high- $k$  isomers in superheavy nuclei. *Phys. Rev. C*, 83:011303, Jan 2011.
- [16] Kazuyuki Sekizawa and Kazuhiro Yabana. Time-dependent hartree-fock calculations for multinucleon transfer processes in  $^{40,48}\text{ca}+^{124}\text{sn}$ ,  $^{40}\text{ca}+^{208}\text{pb}$ , and  $^{58}\text{ni}+^{208}\text{pb}$  reactions. *Phys. Rev. C*, 88:014614, Jul 2013.
- [17] L. Tassan-Got and C. St ephan. Deep inelastic transfers: A way to dissipate energy and angular momentum for reactions in the fermi energy domain. *Nuclear Physics A*, 524:121, 1991.
- [18] R. J. Charity. Systematic description of evaporation spectra for light and heavy compound nuclei. *Phys. Rev. C*, 82:014610, Jul 2010.