## Physics Letters B 771 (2017) 119-124



Contents lists available at ScienceDirect

Physics Letters B

www.elsevier.com/locate/physletb

# Modeling multi-nucleon transfer in symmetric collisions of massive nuclei



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### ARTICLE INFO

Article history: Received 18 November 2016 Received in revised form 12 May 2017 Accepted 15 May 2017 Available online 18 May 2017 Editor: D.F. Geesaman

Keywords: Multi-nucleon transfer GRAZING predictions DNS model Improved Quantum Molecular Dynamics model <sup>204</sup>Hg + <sup>198</sup>Pt

## 1. Introduction

Multi-nucleon transfer reactions have long been used to make heavy nuclei for nuclear spectroscopic studies. Recently attention has been focused on the use of these reactions to study nuclei with N = 126 and to synthesize new neutron-rich heavy nuclei. (Regarding this last point, we note that all trans-uranium nuclei synthesized in hot fusion reactions are neutron-deficient relative to the line of beta stability.)

One of the motivations for the study of multi-nucleon transfer reactions in the heavy nuclei is the recent work of Zagrebaev and Greiner [1]. These authors have pointed out several promising examples of opportunities to make new neutron rich actinide nuclei using multi-nucleon transfer reactions such as  $^{238}U + ^{248}Cm$ . In these reactions, they postulate a mass transfer from the projectile to the target nucleus driven by shell effects in the multidimensional potential energy surface that governs the dynamics of

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#### ABSTRACT

Symmetric collisions of massive nuclei, such as  $^{238}U + ^{248}Cm$ , have been proposed as ways to make new n-rich heavy nuclei via multi-nucleon transfer (MNT) reactions. We have measured the yields of several projectile-like and target-like fragments from the reaction of 1360 MeV  $^{204}Hg + ^{198}Pt$ . We find that current models for this symmetric collision (GRAZING, DNS, ImQMD) significantly underestimate the yields of these transfer products, even for small transfers.

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such collisions at low excitation energies (so-called inverse quasifission). These reactions are difficult to study due to low beam intensities, low cross-sections (picobarn–nanobarn) and the difficulty of detecting the most neutron rich products, which are  $\beta$ -emitters.

Because of these limits on experimental verification of these exciting possibilities, people have recognized the utility and necessity of comparing measurements of multi-nucleon transfer reactions with appropriate models in simpler systems. A well-known model for predicting the cross sections for transfer products is GRAZING, a semi-classical model due to Pollarolo and Winther [2,3]. GRAZ-ING uses a semi-classical model of the reacting ions moving on classical trajectories with quantum calculations of the probability of excitation of collective states and nucleon transfer. This model describes few nucleon transfers [4] well. It has been employed to describe the production of projectile like fragments (PLFs) involving transfers of 4–5 nucleons in the asymmetric reaction of <sup>136</sup>Xe with <sup>238</sup>U, where the predictions of this model "agree well" with measurements [5]. (A modification of the GRAZING code to calculate the decay of primary products by fission and neutron emission has been published [6].)

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http://dx.doi.org/10.1016/j.physletb.2017.05.044



Fig. 1. The observed fragment yields for the  $E_{c.m.} = 619$  MeV  $^{204}$ Hg +  $^{198}$ Pt reaction compared to the predictions of the GRAZING model.

Barrett et al. [7] compared quantitatively the predictions of the GRAZING model and the calculations of Zagrebaev and Greiner to describe the yields of the PLFs and the target-like fragments (TLFs) for the reaction of  $E_{c.m.} = 450$  MeV <sup>136</sup>Xe with <sup>208</sup>Pb. They found that the GRAZING model worked well to describe transfers of  $\Delta Z = -1$  to +2, but failed to describe larger transfers while the model of Zagrebaev and Greiner reproduced the observed distributions for  $\Delta Z = -8$  to +4. Li et al. [8] reviewing the same data, came to similar conclusions about the GRAZING model.

Wen et al. [9] and Zhu et al. [10] have pointed out that significant improvements can be made by combining a dinuclear system (DNS) approach with the GRAZING model. (Zhu et al. [10] also predict, using the DNS model, the possibility of making n-rich nuclei with Z = 99-104 in the symmetric collisions of massive nuclei.) Feng [11] also shows the predictions of the DNS model for the data of Barrett et al. [7]. The DNS model treats the more central collisions neglected in the GRAZING model leading to significantly better predictions for large transfers in the  $E_{c.m.} = 450$  MeV  $^{136}Xe + ^{208}Pb$  reaction and the  $E_{c.m.} = 307.5$  MeV  $^{64}Ni$  with  $^{238}U$  reactions. The GRAZING model describes transfer in peripheral collisions where no capture has occurred while the DNS model describes transfer in more central collisions after capture. The two approaches are thus complementary.

Li et al. [8] did find excellent agreement between the data for the  $E_{c.m.} = 450$  MeV  $^{136}Xe + ^{208}Pb$  reaction and the predictions of the improved Quantum Molecular Dynamics model [12].

In this paper, we report the measurement of the yields of the TLFs and PLFs from the **symmetric** reaction of  $E_{c.m.} = 619$  MeV  $^{204}_{80}$  Hg with  $^{798}_{798}$  Pt. We find that the GRAZING/DNS model underestimates the yields of the smallest transfers (and the larger transfers) by an order of magnitude or more while the ImQMD model does a better (but not adequate) job of representing the data.

## 2. Experimental methods

This experiment was performed at the ATLAS facility of the Argonne National Laboratory. A beam of 1360 MeV  $^{204}$ Hg<sup>31+</sup> ions struck a thick (47 mg/cm<sup>2</sup>)  $^{198}$ Pt target (91.63%  $^{198}$ Pt). The beam stopped in the thick target and the center of target beam energy was 1257 MeV (E<sub>c.m.</sub> = 619 MeV). The effective target thickness was 4.8 mg/cm<sup>2</sup>. The irradiation lasted 1661 minutes with an average beam intensity of  $1.3 \times 10^9$  particles/s. Counting of the irradiated sample using a well-calibrated Ge detector (in the AT-LAS hot chemistry laboratory) commenced about 22 hours after the end of the irradiation and continued for about 3 days. (Nine sequential measurements of the target radioactivity were made.)



Fig. 2. The observed fragment yields for the  $E_{c.m.} = 619$  MeV  $^{204}$ Hg +  $^{198}$ Pt reaction compared to the predictions of the DNS model.

The analysis of these Ge  $\gamma$ -ray decay spectra was carried out using the FitzPeaks [13] software. The end of bombardment (EOB) activities of the nuclides were used to calculate absolute production cross sections, taking into account the variable beam intensities using standard equations for the growth and decay of radionuclides during irradiation [14]. These cross sections represent "cumulative" yields; i.e., they have not been corrected for the effects of precursor beta decay.

To correct for precursor beta decay, we have assumed that the beta-decay corrected independent yield cross sections for a given species,  $\sigma$  (Z, A), can be represented as a histogram that lies along a Gaussian curve.

$$\sigma(Z, A) = \sigma(A) \left[ 2\pi C_Z^2(A) \right]^{-1/2} \exp\left[ \frac{-(Z - Z_{mp})^2}{2C_Z^2(A)} \right]$$
(1)

where  $\sigma(A)$  is the total isobaric yield (the mass yield),  $C_Z(A)$  is the Gaussian width parameter for mass number A and  $Z_{mp}(A)$  is the most probable atomic number for that A. Given this assumption, the beta decay feeding correction factors for cumulative yield isobars can be calculated once the centroid and width of the Gaussian function are known.

One assumes that the value of  $\sigma(A)$  varies smoothly and slowly as a function of mass number and is roughly constant within any A range when determining  $C_Z(A)$ , and  $Z_{mp}(A)$ . The measured nuclidic formation cross sections are then placed in groups according to mass number. We assume the charge distributions of neighboring isobaric chains are similar and radionuclide yields from a limited mass region can be used to determine a single charge distribution curve for that mass region. One can then use the laws of radioactive decay to iteratively correct the measured cumulative formation cross sections for precursor decay. These "independent yield" cross sections are shown in Fig. 1.

Morrissey et al. [15] have examined the uncertainties associated with this method for correcting for  $\beta$ -decay precursors and find a systematic uncertainty of  $\sim \pm 30\%$  associated with this procedure. This error has been added in quadrature to the errors in the measured values.

## 3. Results and discussion

Measured TLF and PLF independent yield cross-sections for the interaction of  $E_{c.m.} = 619$  MeV  $^{204}$ Hg with  $^{198}$ Pt are shown in Figs. 1 and 2 along with the predictions of the GRAZING and DNS models. In all cases, with the exception of the Au fragments, the measured yields were compared to the most likely fragment from the GRAZING/DNS predictions (TLF or PLF). These predictions include the effect of neutron emission by the excited fragments.



Fig. 3. The theory evaluation factors for the  $E_{c.m.}=450$  MeV  $^{136}Xe+^{208}Pb$  reaction, for calculations performed with the GRAZING – DNS model.

With the exception of the yield of the <sup>204</sup>Hg fragments (which include other processes beside multi-nucleon transfer), the observed TLF and PLF fragment yields are orders of magnitude larger than the GRAZING/DNS predictions.

It should be further noted that the measured yield of <sup>199</sup>Au is substantially greater than the neighboring isotopes, <sup>200</sup>Au and <sup>198</sup>Au. This is probably due to the occurrence of the <sup>198</sup>Pt (n,  $\gamma$ ) reaction followed by the  $\beta$ -decay of <sup>199</sup>Pt to <sup>199</sup>Au. Since the decay measurements began 22 hours after the end of irradiation, the yield of 30.8 m <sup>199</sup>Pt was not detected directly.

In the GRAZING code, one requires some choices of parameters having to do with single particle level densities of the nuclei near the Fermi energy. These parameters were varied but with no substantial improvement in the fit to the data.

To make a quantitative comparison between the observations and the predictions of the GRAZING/DNS models, we introduce the use of theory evaluation factors [7]. For each data point, we define

$$tef_i = \log\left(\frac{\sigma_{theory}}{\sigma_{expt}}\right) \tag{2}$$

where  $\sigma_{theory}$  and  $\sigma_{expt}$  are the calculated and measured values of the transfer cross sections. Then, the average theory evaluation factor is given by

$$\overline{tef} = \frac{1}{N_d} \sum_{i=1}^{N_d} tef_i \tag{3}$$

where  $N_d$  is the number of data points. The variance of the average theory evaluation factor is given by

$$\sigma = \frac{1}{N_d} \left( \sum_i \left( tef_i - \overline{tef} \right)^2 \right)^{1/2} \tag{4}$$

Note that **tef** is a logarithmic quantity and theories that have **tef** values differing by 1 or 2 actually differ by orders of magnitude in their reliability.

To orient the reader as to what might be expected, we show, in Fig. 3, the **tef** values for (a) the PLFs and (b) the TLFs from the interaction of  $E_{c.m.} = 450$  MeV  $^{136}Xe + ^{208}Pb$  [7]. The theory calculations are made using the GRAZING + DNS models. In Fig. 3a, one



Fig. 4. The theory evaluation factors for the  $E_{c.m.}=619$  MeV  $^{204}\text{Hg}+^{198}\text{Pt}$  reaction using the GRAZING + DNS model.

observes the typical pattern where GRAZING + DNS correctly describes the small transfers for the PLFs ( $\Delta Z = 0, \pm 1$ ) but becomes progressively worse in describing the larger transfers. For the addition of protons to the target nucleus (Fig. 3b) one sees a similar trend although for the largest transfer ( $\Delta Z = +4$ ), GRAZING + DNS is not as bad as for the PLFs. In describing proton transfers to the doubly magic <sup>208</sup>Pb, one notes that GRAZING+DNS becomes progressively worse by one order of magnitude for each transferred proton.

In Fig. 4, one shows the **tef** values for the  $E_{c.m.} = 619$  MeV <sup>204</sup>Hg with <sup>198</sup>Pt reaction (this work). For this system, GRAZ-ING + DNS gives a poorer description of the data as the number of transferred protons ( $Z \ge 81$ ) increases. Compared to the <sup>136</sup>Xe + <sup>208</sup>Pb reaction (Fig. 3b) the GRAZING + DNS predictions for the TLFs in the <sup>204</sup>Hg + <sup>198</sup>Pt reaction are substantially worse.

As discussed above [8], the improved Quantum Molecular Dynamics model [12] (ImQMD) has been demonstrated to correctly predict the yields of multi-nucleon transfer products. In Figs. 5 and 6, we compare the predictions of this model [16] with our observations for the  $E_{c.m.} = 619$  MeV  $^{204}$ Hg +  $^{198}$ Pt reaction. In these calculations, 390000 simulation events were calculated for impact parameters of b = 1, 2, 3...13 fm. Secondary decay of the primary products was calculated using GEMINI [17]. The version of the ImQMD model used in the calculations was that described in [16]. For nuclei around Z = 82, the predicted cross sections from ImQMD simulations are significantly smaller than the data, which could be due to the lack of shell effects in the ImQMD simulations. Better agreement is observed for this model compared to GRAZING + DNS model indicating this model is a more appropriate model for symmetric multi-nucleon transfer reactions between massive nuclei and, by comparison, that GRAZING + DNS is not an appropriate model. Since there are well-documented cases [4,5] of the ability of the GRAZING model to correctly describe asymmetric collisions, additional examples of how well GRAZING (and the DNS model) describe symmetric collisions would be welcome.

## Acknowledgements

We thank Prof. F. S. Zhang and co-workers for making the DNS and ImQMD calculations cited in this paper and R.V. F. Janssens for helpful comments. This work was supported in part by the Director, Office of Energy Research, Division of Nuclear Physics of the Office of High Energy and Nuclear Physics of the US Department of Energy under Grant DE-SC0014380 (OSU), Grant Number



**Fig. 5.** The observed fragment yields (red circles) for the  $E_{c.m.} = 619$  MeV  $^{204}$ Hg +  $^{198}$ Pt reaction compared to the predictions (blue circles and line) of the ImQMD model. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)



Fig. 6. The theory evaluation factors for the  $E_{c.m.}=619$  MeV  $^{204}\rm Hg+{}^{198}\rm Pt$  reaction using the ImQMD model.

DE-FG02-94ER40834 (Maryland), Grant No. DE-FG02-94ER40848 (UML), Contract No. DE-AC02-98CH10886 (BNL) and Contract DE-AC02-06CH11357 (ANL), and the National Science Foundation under Award 1505043. One of us (TW) wants to thank the USDOE for support as a summer fellow in the Summer School on Fuel Cycle Chemistry (DE-NE0008478). This research used resources of the ANL's ATLAS facility, which is a DOE Office of Science User Facility.

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