Letter

Observing the ping-pong modality of the isospin degree of freedom in cluster emission from heavy-ion reactions

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Thermodynamic and chemical two-body correlations of the isotope-resolved clusters are measured in 86 Kr + 208 Pb reactions at 25 MeV/u. The yield and kinetic variables of the A = 3 isobars are analyzed in coincidence with the intermediate mass fragments of $6 \le A \le 11$. While the velocity spectra of both *t* and 3 He exhibit scaling behavior over the type of the intermediate mass fragments, the yield ratios of $t/{}^{3}$ He correlate reversely to the neutron-to-proton ratio N/Z of the latter, showing the ping-pong modality of the N/Z of the emitted particles. The commonality that the N/Z of the residues keeps the initial system value is extended to the cluster emission in heavy ion reactions. The comparison to transport model calculations to the data is discussed.

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Introduction. Heavy ion reaction (HIR) is a femtoscopic nonequilibrium process, where neutrons and protons transport differently [1]. Relying on transport model description on a variety of observables in HIRs, one can unveil the equation of state of nuclear matter (nEOS), which is essential to describe the explosive events of dense stellar objects where the heavy elements are created in the universe [2–5]. The large uncertainties of the nEOS come from its isospin vector part, namely the density dependent nuclear symmetry energy $E_{\text{sym}}(\rho)$ [6], a longstanding frontier bridging astrophysics and nuclear physics. The accurate constraint of $E_{\text{sym}}(\rho)$ becomes more indispensable than ever since the detection of the gravitational wave from GW170817 [7–9].

Despite large progress on constraining $E_{\text{sym}}(\rho)$ in terrestrial nuclear laboratories [10–19], the slope parameter of $E_{\text{sym}}(\rho)$ suffers 30% uncertainty and the symmetry energy at about $2\rho_0$ is even less constrained. To achieve accurate results, great efforts are demanded to improve the reliability of the theoretical models [20–24] and to accumulate more experimental data containing isospin sensitive probes [25,26].

The ongoing endeavors encounter unavoidably two fundamental questions arising from the thermodynamic and chemical complication of HIRs. One is the transport of a given degree of freedom (DOF) in a dynamic process, i.e., how does the DOF of isospin (IDOF) evolve in HIRs? The question stimulates enormous progress on isospin chronology applications [27–32]. The other is clustering, which makes it more complicated to probe $E_{\text{sym}}(\rho)$ using HIRs, since cluster formation influences the isospin transport and the yield of the final products [33].

To this end, it is essential to elucidate the two questions and their interplay in the specific HIRs before the $E_{\text{sym}}(\rho)$ can be stringently constrained. We are motivated to illustrate how the IDOF evolves with the presence of cluster emission in HIRs by introducing the temporal two-body correlation analysis.

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The motivation is novel, because the isospin observables from HIRs are so far mostly one-body quantity, which prevents to draw detailed and comprehensive picture on how the IDOF evolves with the presence of clustering. Among many existed observables, the ratio of A = 3 isobars, $t/{}^{3}$ He, has been identified to probe $E_{\text{sym}}(\rho)$ both theoretically [17,18,34–40] and experimentally [40–48].

In this letter, the thermodynamic and chemical correlations of the A = 3 isobars and the intermediate mass fragments (IMFs) are analyzed in 25 MeV/u⁸⁶Kr + ²⁰⁸Pb reactions. The anticorrelation of the ratio $t/{}^{3}$ He with the N/Z of the heavy fragments are observed, showing a novel ping-pong modality. Further, we report the commonality that the N/Z of the initial system undergoing cluster emission is kept in various nuclear processes. The comparison of the data to the transport model simulations is discussed.

Experimental setup. The experiment was performed with the Compact Spectrometer for Heavy IoN Experiment (CSHINE) [49,50], installed at the final focal plane of the Radioactive Ion Beam Line at Lanzhou (RIBLL-I) [51]. The 25 MeV/u⁸⁶Kr beam was delivered by the Heavy Ion Research Facility at Lanzhou (HIRFL) [52], bombarding a natural lead target with the thickness of 1 mg/cm². In the current configuration of CSHINE, light charged particles (LCPs) and IMFs are measured by four silicon-strip detector telescopes (SSDTs), while the coincident fission fragments (FFs) are measured by three parallel plate avalanche counters (PPACs), each covering a sensitive area of $240 \times 280 \text{ mm}^2$. Each SSDT consists of one single-sided silicon-strip detector (SSSSD) and one double-sided silicon strip detector (DSSSD), backed by a 3×3 CsI(Tl) crystal hodoscope with length of 50 mm. The sensitive area of each SSDT is 64×64 mm², and the granularity is $4 \times 4 \text{ mm}^2$ delivering about 1° angular resolution. The SSDTs cover the angular range of 10° -60° in laboratory. The energy resolution of the SSDT is better than 2%, and the isotopes up to Z = 6 can be identified. For the technical details of the CSHINE, one can refer to [49,50]. Since the event statistics of the four-body coincidence, i.e., with two charged particles in SSDTs and two FFs in PPACs, does not suffice, here, we analyze the two-body coincidence of two fragments in the SSDTs. Multihits and signal sharing are treated with care in the track recognition, the overall track recognition efficiency is about 90%. The detailed procedure of track reconstruction can be found in [53].

Figure 1 summarizes the data used in the current analysis. Panel (a) presents the scatter plot of $\Delta E_2 - E_{CsI}$ of the SSDT1. It is shown that all the isotopes can be identified clearly to Z = 6. Panel (b) presents the PID plots for all isotopes on $\Delta E_2 - E_{CsI}$ summed over SSDT 1 to 4 after the tracking procedure [53]. Panels (c) and (d) present scatter plots of longitudinal vs. transverse velocity distributions of triton and ³He stopped in the CsI(TI) hodoscope, respectively.

Results and Discussions. In order to observe the temporally correlated emissions of charged particles, we investigate the two-body events in the SSDTs, with one being the LCP, $F_L(Z_L, A_L)$, the other being a IMF, $F_H(Z_H, A_H)$, with $6 \le A_H \le 11$, where the subscripts L and H denote *light* and *heavy* to differentiate the two particles, respectively. Here, F_L is chosen as one of the mirror nuclei, triton, or ³He. Accord-



FIG. 1. The scatter plot of $\Delta E_2 - E_{CsI}$ of the SSDT1 (a). The PID plots for all isotopes on $\Delta E_2 - E_{CsI}$ summed over SSDT 1 to 4 (b). The scatter plots of longitudinal vs. transverse velocity distributions of triton and ³He are plotted in (c) and (d), respectively. The azimuthal efficiency is corrected.

ing to earlier studies using correlation function method, the heavy fragments are emitted with an averagely shorter time constant at this energy region [54]. Thus, it is reasonable to assume that the F_H is emitted earlier than F_L in our study. To circumvent the spectrum discontinuity caused by the gap between ΔE_1 and ΔE_2 , only the particles F_L stopped in the CsI(Tl) hodoscope are analyzed.

We first check the thermodynamic correlation of F_L and F_H . Figures 2(a) and 2(b) present the velocity spectra of triton (a) and ³He (b) in coincidence with various IMFs F_H . It is shown that the shape and the range of the velocity distributions of triton and ³He exhibit insignificant dependence on the type of F_H , although the coincident yield differs. Normalizing the yields to the histograms in the case where F_H is ⁷Li, one



FIG. 2. The spectra of the velocities (in unit of *c*) of triton (a) and ³He (b) in coincidence with various F_H of $A_H = 6$ to 11. The lower panels present the mean velocity $\langle v \rangle$ of *t* (c) and ³He (d), with the solid (open) circles denoting the coincident F_H of larger (smaller) neutron richness, respectively. The shadowing band denotes the standard deviation of each corresponding spectrum.



FIG. 3. (a) Yield ratio of $R(t/{}^{3}\text{He})$ in coincidence with different IMFs, the solid (open) circles represent the IMF of larger (smaller) neutron richness at a given mass number. (b) $R(t/{}^{3}\text{He})$ as a function of the neutron richness $\delta I_{\rm H}$ of $F_{\rm H}$. *k* and y_0 are the slope and the intercept parameters of the linear fit, respectively. The error bars are of only statistical origin.

sees that all the spectra exhibit scaling behavior, as shown in the insets in (a) and (b). Furthermore, the mean velocity $\langle v \rangle$ of triton and ³He are presented in (c) and (d), varying with the type of F_H. The shadowing band represents the standard deviation σ_v of each individual v spectrum. The solid (open) symbols denote the F_H with larger (smaller) neutron richness, respectively. Clearly, both $\langle v \rangle$ and σ_v are nearly constant, showing insignificant dependence on the N/Z of F_H. It suggests that the two A = 3 isobars experience the same dynamic process, regardless of whether the correlated IMFs are rich or deficient in neutrons.

Surprisingly, however, the IDOF features differently. Figure 3(a) presents the yield ratio of $t/{}^{3}$ He, written as $R(t/{}^{3}\text{He})$, as a function of A_{H} . Again, the solid (open) symbols correspond to the more (less) neutron-rich F_H at a given mass (here, neutron-rich only has its relative meaning in the F_H pair with the same mass). The IMF ⁸Be is reconstructed by two correlated α particles. It is seen that the ratio $R(t/{}^{3}\text{He})$ splits in two groups. A ping-pong motion modality of the N/Z in cluster emission is suggested: after a neutronrich IMF (solid) is emitted, the $R(t/{}^{3}\text{He})$ is smaller, indicating that a less neutron-rich F_L follows. Oppositely, if the F_H is less neutron rich (open), $R(t/{}^{3}\text{He})$ is larger, indicating that F_L is averagely more neutron-rich. The ping-pong behavior of the N/Z of the two clusters demonstrates that the isospin content of F_L depends on the N/Z of F_H emitted at earlier chance. This tendency is further presented in panel (b), where the $R(t/{}^{3}\text{He})$ is plotted as a function of the relative neutron richness of F_H defined by $\delta I_H = (N_H - Z_H)/A_H$. The solid line is the least square linear fit to the data points. A nonzero minus slope k is convinced at 5.8 σ confidential level, evidencing that the isospin compositions of F_H and F_L are anticorrelated.

Model independently, the anticorrelation between the N/Z of F_L and F_H infers how the IDOF evolves during the decay of the highly excited system formed in HIRs. If more (less) neutrons are carried away by the IMF, the following LCP carries less (more) neutrons, as a consequence of neutron and proton (isospin) conservation in the reaction system of finite size. It suggests that for the decay of such a finite size system, the neutron richness of the emitted particles is balanced near a



FIG. 4. The correlation between the N/Z of the initial system and the remaining nuclei. The asterisk denotes the initial 86 Kr + 208 Pb system, and the colored contour represents the probability distribution of the remaining nuclei after the emission of F_H and F_L. The circles denote the cluster decay of various superheavy nuclei, with the black (blue) ones being the parents (daughters). The inset displays the yield distribution of the *n*-induced fission of 235 U on the N - Z plot.

certain value, so that the remaining system, possibly destined to emission residue or fission, possesses a certain N/Z close to that of the initial system, see below.

In order to show the isospin balancing effect clearly, Fig. 4 presents the N - Z correlation of the remaining nuclei, obtained by subtracting F_L and F_H from the initial system. The N/Z = 176/118 relationship is indicated by the red dashed line, and the asterisk denotes the location of the ⁸⁶Kr + ²⁰⁸Pb system. It is shown that the remaining system situates in the vicinity of the dashed line, indicating that the average N/Z of the remaining system is the same as the initial value of ⁸⁶Kr + ²⁰⁸Pb. Here, we can only count the two clusters measured, including A = 3 isobars and the IMFs. Because of the anticorrelation demonstrated in Fig. 3, it is reasonable to assume the undetected decay particles have a similar N/Zto the detected products and hence do not drag the distribution center away from the dashed line.

Interestingly, the cluster radioactivity observed in various superheavy nuclei [55–57] is similar, as shown by the circles in the plot. The cluster decay channels are taken from [56]. It is seen that the daughters (blue) are distributed near the same N/Z line of the parent nuclei (black). Moreover, if one inspects the fission of heavy nuclei, for instance, the neutron induced fission of 235 U plotted in the inset, the N/Z of the fission fragments follows the same N/Z value of the parent nucleus ²³⁵U [58]. Because of the presence of $E_{sym}(\rho)$, which finally goes into the Q value, the N/Z of the daughters inherits that of the parents. This is commonly observed in fission and in the cluster decay of heavy nuclei. Our experiment extends the commonality to the cluster emission of the finite and highly excited system formed in HIRs. The commonality also supports that the $E_{sym}(\rho)$ is at work in the anticorrelation of the N/Z of F_L and F_H emission, in addition to the neutron and proton number conservation.

Comparison to transport model simulations. To understand the isospin transport behavior, the reaction process is simu-



FIG. 5. Yield ratio of $R(t/{}^{3}\text{He})$ by ImQMD simulations as a function of δI_{H} with $\gamma = 0.4, 0.5, 0.7, \text{ and } 2.0$, respectively. The results with b = 1 to 7 fm are represented by the open symbols, on top of which the *b*-averaged results are presented by the solid circles with the linear fit (dashed line).

lated using the improved quantum molecular dynamics model (ImQMD05) [59] followed by a statistic decay afterburner GEMINI [60,61]. The ImQMD05 model [59] was developed from the original version of the quantum molecular dynamics code proposed by Aichelin *et al.* [62,63], and has been a useful tool to explore the reaction dynamics of HICs in low and intermediate energies for understanding the nuclear phenomena. For brevity, we limit our discussion here to the mean field part of the ImQMD05 model, where the symmetry potential energy is involved. The local nuclear potential energy density functional in the ImQMD05 model is written as

$$\begin{aligned} V_{\rm loc} &= \frac{\alpha}{2} \frac{\rho^2}{\rho_0} + \frac{\beta}{\eta + 1} \frac{\rho^{\eta + 1}}{\rho_0^{\eta}} + \frac{g_{\rm sur}}{2\rho_0} (\nabla \rho)^2 \\ &+ \frac{g_{\rm sur, iso}}{\rho_0} [\nabla (\rho_{\rm n} - \rho_{\rm p})]^2 + g_{\rho\tau} \frac{\rho^{8/3}}{\rho_0^{5/3}} + \frac{C_{\rm s}}{2} \frac{\rho^{\gamma + 1}}{\rho_0^{\gamma}} \delta^2, \end{aligned}$$
(1)

where ρ , ρ_n , ρ_p are the nucleon, neutron, and proton density, respectively. $\delta = (\rho_n - \rho_p)/(\rho_n + \rho_p)$ is the isospin asymmetry degree. In this work, the parameters in Eq. (1) are determined by the MSL0 parameter set [64], in which the parameter γ is adjusted to mimicked the various density behavior of symmetry potential energy. The simulations cover the impact parameter range of $1.0 \le b \le 11.5$ fm with steps of $\Delta b = 1.5$ fm. For each b, 5×10^5 events were simulated. At the end of the ImQMD calculations, clusters are recognized by a minimum spanning tree (MST) algorithm [63,65] widely used in the quantum molecular dynamics calculations. The information of the excited cluster is input into the GEMINI model to perform statistical decay calculations. The same velocity and polar angle cuts are adopted to t and ³He as in experiment. To avoid the ambiguity of the correlation, we exclude the events containing more than one F_H with the same $N_{\rm H}$ and $A_{\rm H}$, but set no cut on the multiplicity of $F_{\rm L}$.

Figure 5 presents the $R(t/{}^{3}\text{He})$ ratio as a function of δI_{H} for different $E_{\text{sym}}(\rho)$ with $\gamma = 0.4, 0.5, 0.7, \text{ and } 2.0, \text{ respec-}$

tively. The value of $R(t/{}^{3}\text{He})$ depends sensitively on $E_{\text{sym}}(\rho)$, consistent with [34], but less sensitive on *b*, as shown by the open symbols in panels (a)–(d). The *b*-weighted average ratios are depicted by the solid circles with linear fit (dashed line). The experimental trend is replotted in solid lines for comparison. Interestingly, the anticorrelation trend between $R(t/{}^{3}\text{He})$ and δI_{H} is qualitatively reproduced, but the slope is generally underestimated in the simulation. The superstiff ($\gamma = 2.0$) and the supersoft ($\gamma = 0.4$) parametrizations of $E_{\text{sym}}(\rho)$ are disfavored by the large deviation from the experimental trend. By varying γ from 0.5 to 0.7, the simulations cross over the experimental area.

The comparison has an important implication. If one only looks at the integrated $R(t/{}^{3}\text{He})$, i.e., the projection of the data points to the ordinate, apparently $\gamma = 0.7$ is favored, as shown in Fig. 5(c). Nevertheless, the distribution of integrated $R(t/{}^{3}\text{He})$ is broadened since its average value varies with the N/Z of the coincident IMF, even with a well-determined impact parameter. In other words, the measurement of the anticorrelation between $R(t/{}^{3}\text{He})$ and δI_{H} provides an extra dimension to reveal finely the isospin dynamics in presence of clustering in the finite system, which is hardly realizable through the integrated $R(t/{}^{3}\text{He})$ as a one-body quantity. Since the discrepancy on the slope is still evident between the data and the simulations, we do not quantify $E_{\text{sym}}(\rho)$ more precisely here. Instead, with this new line, we suggest that the modeling of clustering in HIRs with transport theory requires further efforts, even for the A = 3 clusters, of which the origin is very complicated.

It is important to ensure that the experimental coverage is mainly contributed by the intermediate velocity source. From the velocity distribution shown in Fig. 1(c) and 1(d), one sees no separated contribution from the projectile-like fragment (PLF) source, which is characterized by small transverse velocity and longitudinal one close to that of projectile. To be more convincing, the velocity distribution of ImQMD calculations is studied. Figure 6 presents the scatter plots of



FIG. 6. The scatter plots of longitudinal vs. transverse velocity distributions in ImQMD simulation for triton (a), ³He (b), IMFs with $6 \le A \le 11$ (c), and heavy residues with A > 30 (d). The experimental angular and velocity cuts are presented in (a) to (c).

longitudinal vs. transverse velocity distributions for the products with different masses in ImQMD simulations. Unlike the heavy residues of A > 30 shown in (d), where both components of the PLF and the target-like fragment (TLF) are clearly seen, the LCPs with A = 3 do not exhibit obvious contribution from PLF in the angular range of our study, as shown in panels (a) and (b). With increasing the mass, the distribution becomes more forward peaked but is still far from the location of PLF with A > 30, as shown in panel (c) for F_H under investigation. In our analysis, in order to suppress maximally the contribution from the PLF source, we adjusted the lower angular cut from 10° to 20° . When the cut is loosened to 10° and F_H with $A \leq 14$ can be included, the ping-pong modality remains robustly clear and the conclusion is not changed.

Experimentally, it is also important to select the certain centrality of the reaction. As presented in Fig. 5, a selection of the impact parameter increases the precision of constraining $E_{\text{sym}}(\rho)$. But this is not done due to the lower statistics of four-body events. Nevertheless, because our SSDTs covers only part of a solid angle, it is less likely that two or more LCPs and IMFs can be recorded by the SSDT array in a very peripheral reactions. It is presumable that two-body correlation in the SSDTs has some selection ability to the impact parameter, particularly one fragment is in an intermediate mass range. Again, one can check this presumption using transport model simulations by counting the correlations of F_L and F_H out of the total events. Defining the total weight of the pair correlation by

$$W_{\rm LH} = b \cdot n_{\rm LH}(b) / N_{\rm tot}(b), \qquad (2)$$

where n_{LH} are the number of the correlated pairs of F_L and F_H counted in N_{tot} events simulated at a given impact pa-



0.4 3 0.2 5 b/fm

FIG. 7. The weight of the events containing F_L and F_H correlation as a function of impact parameter *b* in ImQMD simulations with $\gamma = 0.5$.

rameter *b*. The velocity and polar angle cuts are taken as the same as in experiment. As plotted in Fig. 7, the weight starts at a very low level at central collisions and reaches the top at semicentral ones before decreasing rapidly at large *b*, suggesting that the correlation observable selects mainly the semicentral collisions. It supports the effectiveness of the range of 1.0 < b < 11.5 fm taken in the simulations for the comparison presented in Fig. 5.

Conclusion. Transport of isospin degree of freedom and cluster formation are two extremely important aspects of HIRs. To understand the two aspects, the A = 3 isobars are analyzed in coincidence with fragments of $6 \le A \le 11$ in 25 MeV/u ⁸⁶Kr + ²⁰⁸Pb reactions. While the velocity spectra of triton and ³He show scaling behavior over the type of the intermediate mass fragments, the yield ratio $R(t/{^{3}\text{He}})$ exhibits evident anticorrelation with the N/Z of the latter, suggesting the ping-pong modality of the N/Z of the emitted particles. The commonality that the remaining system inherits the N/Zof the initial system, which has been reported in cluster decay and fission of superheavy nuclei, is extended to the cluster emission from HIRs in Fermi energy region. In addition to the yield ratio of $t/{}^{3}$ He, the anticorrelation between the N/Zof the two clusters, being sensitive to the isospin dynamics in a finite-size system, provides a benchmark to test the transport theory in terms of cluster formation and isospin dynamics, and helps eventually achieving stringent constraint of $E_{\rm sym}(\rho)$ near the saturation density using heavy ion reactions.

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