



## Letter

# Revealing nucleon-nucleon correlation effects through sub-Coulomb transfer reactions in $^{92}\text{Mo} + ^{54}\text{Fe}$

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## ABSTRACT

Cross sections for neutron and proton transfer channels were measured in the proton-rich system  $^{92}\text{Mo} + ^{54}\text{Fe}$  using the PRISMA magnetic spectrometer. The energy dependence of the absolute transfer cross sections was investigated over a beam energy range spanning from slightly above to below the Coulomb barrier. A significant enhancement was observed in the  $2p$  and  $2p + 2n$  pickup and stripping channels compared to expectations based on independent single-nucleon transfer, suggesting strong correlations in the multinucleon transfer mechanism.

## 1. Introduction

Multinucleon transfer reactions have been the focus of extensive experimental [1–9] and theoretical [10–15] efforts aimed at identifying optimal conditions to maximize the production cross sections of the more exotic isotopes. Despite significant progress, the predictive power of current reaction models remains limited, partly due to unresolved questions about the relevant degrees of freedom that must be incorporated into the reaction mechanism, including the not yet fully understood role of nucleon-nucleon correlations. These correlations, induced by the pairing interaction, are known to play a fundamental role in defining properties of nuclei as finite quantum many-body systems, both in their ground and low-lying excited states. Understanding their role in the evolution of the collision, and the relative importance of single-nucleon transfer compared to more complex processes involving the transfer of nucleon pairs, and how these affect the observed cross sections, remains a challenge for theory and experiment alike [16–18].

Significant advances have been made in studying neutron-neutron correlations, for example in closed-shell  $^{96}\text{Zr} + ^{40}\text{Ca}$  [19], superfluid

$^{116}\text{Sn} + ^{60}\text{Ni}$  [20,21] and relatively heavy  $^{206}\text{Pb} + ^{118}\text{Sn}$  [22] systems, where excitation functions were measured at bombarding energies from the Coulomb barrier to far below it. Notably, the  $^{116}\text{Sn} + ^{60}\text{Ni}$  study was the first in heavy-ion collisions to show agreement between experimental transfer probabilities and microscopic models incorporating neutron-neutron pairing correlations. Very recently, the features evidenced in the behavior of the transfer probabilities for this system have been characterized as the nuclear analog to the Josephson effect [23–25].

The role of proton-proton and neutron-proton correlations remains significantly less understood [18,26–28]. Systematic studies of proton transfer have been much less extensive than those for neutrons, both for light-ion and heavy-ion-induced reactions [16,18,29–32]. In reactions involving heavy ions, there is a unique opportunity to simultaneously study the transfer of single nucleons, nucleon pairs, and multiple nucleons within the same reaction. While enhanced two-proton transfer yields have been frequently observed, strong two-proton-two-neutron transfer has been reported only in selected systems [17,18,33]. However, most of these studies were performed at or above the Coulomb barrier, where the interpretation of the data is complicated by the

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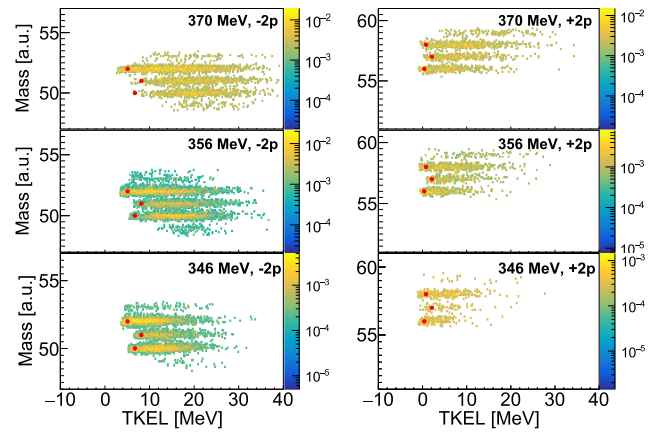
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combined effects of absorption and interference generated by rainbow phenomena. A systematic investigation at sub-barrier energies, where nuclei interact at large distances under quasi-elastic conditions, ideal for probing nucleon-nucleon correlations, has been lacking due to the inherently low transfer cross sections and the demanding experimental conditions. Insights into correlated transfer have emerged from experiments at and below the Coulomb barrier with lighter ion beams, such as  $^{16}\text{O}$ , on  $^{208}\text{Pb}$  targets, that have revealed enhanced  $-2p$  and  $-2p-2n$  transfer, with the latter dominating for  $^{18}\text{O}$  [34,35]. Recently, in the heavier  $^{116}\text{Sn} + ^{60}\text{Ni}$  system, proton-proton correlations were found to significantly influence the yields of  $-2p$  and  $-2p-2n$  transfer channels (i.e. two-proton and two-proton-two-neutron stripping from the light reaction partner) at sub-barrier energies [28]. Nevertheless, data on two-proton transfer channels, particularly on the pickup side, remain scarce at sub-barrier energies, limiting a complete understanding of the underlying mechanisms.

In this letter, we present the first results on multinucleon transfer reactions involving proton-rich nuclei, using the  $^{92}\text{Mo} + ^{54}\text{Fe}$  system studied with high mass, charge and  $Q$ -value resolution at several beam energies spanning above, near, and below the Coulomb barrier. This system is of particular interest as it involves a nucleus below the  $Z = 28$  shell closure, close to the  $N = Z = 27$  region, where neutrons and protons occupy orbitals of the same shell. Given that transfer occurs at the nuclear surface, proton-rich systems offer an ideal environment to study proton correlations. Although this region has been investigated in light-ion reactions, including those with radioactive beams [36–39], it remains largely unexplored in heavy-ion collisions, especially at sub-barrier energies. Our high-resolution measurements provide absolute total cross sections for both proton stripping and pickup channels, offering a systematic view of the transfer dynamics. In particular, the proton pickup channels, which are much less affected by evaporation, provide a more direct probe of nucleon-nucleon correlation effects. The observation of enhanced yields for channels involving the transfer of multiple nucleons, especially pairs or even clusters, provides compelling evidence for such correlations between the transferred nucleons affecting the dynamics.

## 2. The experiment

The measurement was performed in inverse kinematics using a  $^{92}\text{Mo}$  beam with average currents of  $\sim 2$  pA, employing the superconducting PIAVE-ALPI accelerator complex of Legnaro National Laboratories (LNL). The  $100 \mu\text{g}/\text{cm}^2$   $^{54}\text{Fe}$  targets consisted of a strip of 2 mm sandwiched between  $20 \mu\text{g}/\text{cm}^2$  C layers, and had an isotopic enrichment of 99.9%. The  $^{56}\text{Fe}$  impurities were at the level of  $6 \times 10^{-4}$  compared to  $^{54}\text{Fe}$ . The bombarding energy of ALPI has been varied between 370 and 340 MeV, with a precision of  $\sim 2\%$ . Three lowest energies (305, 293, and 280 MeV) were delivered by the XTU Tandem accelerator, providing energy accuracy better than 0.1%, to measure Rutherford scattering, which was used to determine the PRISMA solid angle and to check the beam energy. One energy (346 MeV) was measured using Tandem + ALPI. To minimize beam time losses from tuning of the ALPI cavities, a thick  $85 \mu\text{g}/\text{cm}^2$  C degrader foil was used to lower the beam energy by  $\sim 5$  MeV, allowing measurement at one additional energy for each ALPI setting. This approach enabled the measurement of cross sections over a range of bombarding energies, spanning from above the Coulomb barrier to well below it. Normalization between different runs was ensured by two collimated silicon surface barrier monitor detectors placed at  $\theta_{\text{lab}} = 48^\circ$  and  $53^\circ$  with respect to the beam direction and at a distance of  $\sim 40$  cm from the target. The monitors detected the Rutherford-scattered Fe-like (as well as  $^{12}\text{C}$ ) recoils. To more accurately determine the ALPI beam energy, we required that the energies of Rutherford-scattered target-like Fe and C ions agreed, via a minimization procedure, with those measured in the runs where only the Tandem was used. This results in a beam energy determination with an accuracy better than 1%.



**Fig. 1.** Two-dimensional mass vs. TKEL matrices for two-proton transfer: stripping channels with Cr isotopes (left) and pickup channels with Ni isotopes (right) detected in PRISMA, shown at the indicated beam energies. The colour scale shows counts normalized to the monitor detector counts. One sees the clear separation between different masses. The ground-state-to-ground state  $Q$  values are indicated by red points.

The experiment was performed using the high-efficiency, large-acceptance magnetic spectrometer PRISMA [40,41], where Fe-like recoils were detected at  $\theta_{\text{lab}} = 22^\circ$ , corresponding to  $\theta_{\text{cm}} = 136^\circ$ . Here we briefly recall the main characteristics of the spectrometer and its detector system. A position-sensitive micro-channel plate detector [42] is placed at the entrance of the spectrometer, providing a start signal for time-of-flight measurements and two-dimensional position signals. Ions pass through the optical elements of the spectrometer (a quadrupole and a dipole) and enter a focal plane [43] which consists of a multi-wire parallel plate avalanche gas detector, providing timing and two-dimensional position signals with resolutions similar to the entrance detector. An array of transverse-field multi-parametric ionization chambers (IC) follows, providing nuclear charge ( $\Delta E$ ) and total energy ( $E$ ). In order to obtain the optimum nuclear charge ( $Z$ ) resolution, the direction followed by the different ions reaching the IC in a broad range of kinetic energies and directions was taken into account. Mass identification has been based on an event-by-event reconstruction of the ion trajectories, using two-dimensional entrance and exit positions and the time of flight through the spectrometer [40,44,45]. To illustrate the channel separation and the almost background-free conditions, we display in Fig. 1 two-dimensional spectra of mass vs total kinetic energy loss (TKEL) for the two-proton stripping and pickup transfer channels (the terms stripping and pickup refer to the light reaction partner) at three selected beam energies, above, near and below the Coulomb barrier, i.e., at  $E_{\text{lab}} = 370, 356$  and  $346$  MeV. Notably, larger yields are, on average, associated with more positive ground-state-to-ground-state  $Q$  values, that are indicated by a red point, which is consistent with a broader range of accessible excitation energy in the final nuclei. This behavior reflects the impact of the  $Q$ -value window in the transfer flux. Nuclear structure properties of both the initial and final nuclei may, as well, influence the observed distributions.

## 3. Results and discussion

### 3.1. Total kinetic energy loss distributions

Total kinetic energy loss distributions for the  $1p$ ,  $2p$  and  $2p+2n$  stripping and pickup transfer channels at selected bombarding energies above, near and below the barrier are shown in Fig. 2. The distributions, which incorporate excitation energies of both light and heavy partner, were constructed assuming binary kinematics and momentum conservation, using the measured energies of detected ions, integrated over all

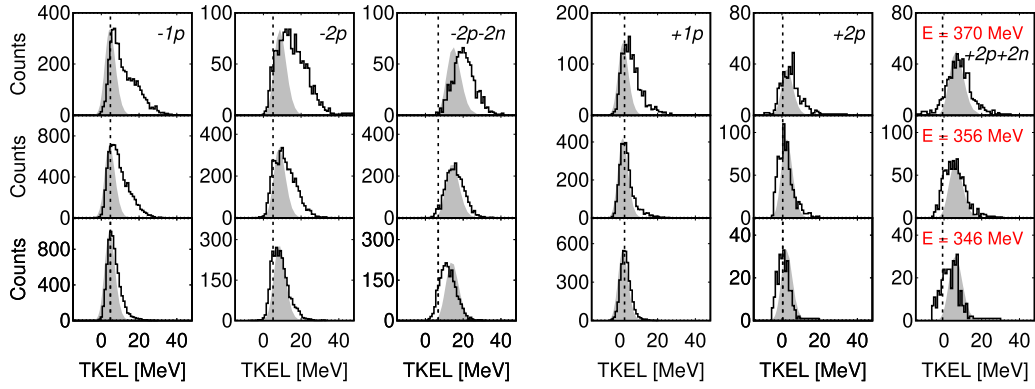


Fig. 2. TKEL distributions for  $1p$ ,  $2p$  and  $2p + 2n$  stripping and pickup transfer channels at the indicated energies. Vertical dotted line corresponds to the ground-state-to-ground-state  $Q$ -value. GRAZING calculations are shown with gray filled histograms.

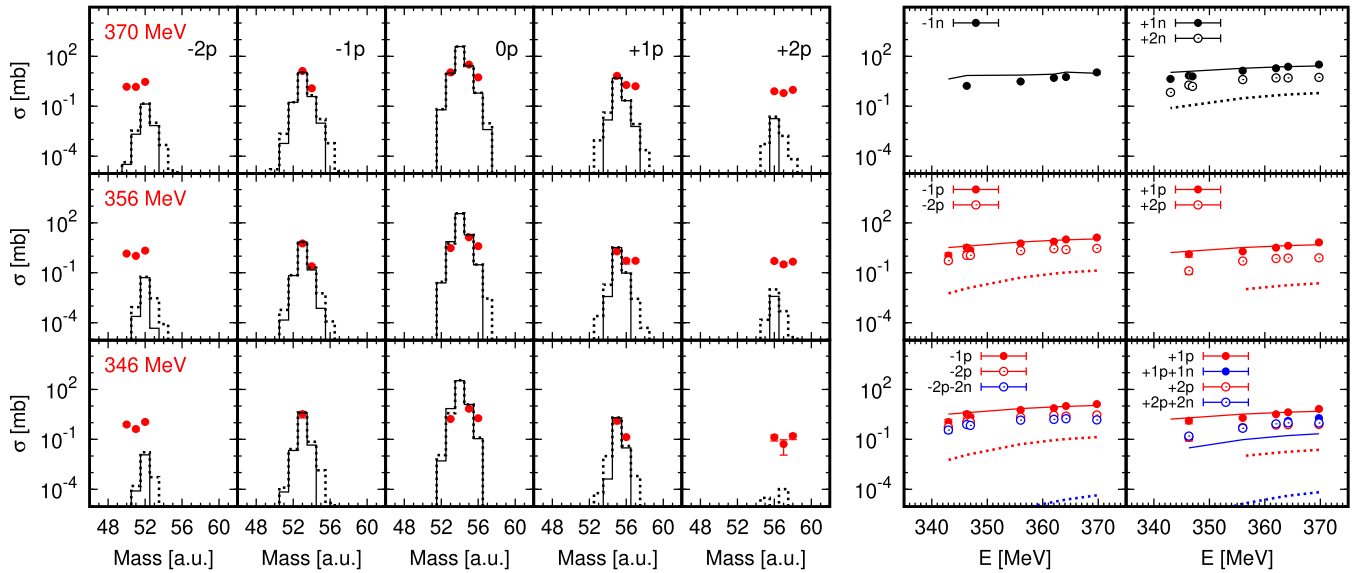


Fig. 3. Left panels: experimental data (points) and GRAZING calculations without (dotted histograms) and with the evaporation of neutrons (full black lines) of total angle- and  $Q$ -value-integrated cross sections for various transfer channels at three selected beam energies. These represent cases above, near, and below the barrier, respectively. Experimental uncertainties are purely statistical and are generally smaller than the symbol size. In the GRAZING calculations, the ion-ion potential radius was reduced by 0.2 fm, the level density for neutrons was increased by 20%, and the form factor strengths for neutrons and protons were modified via correction factors of 0.5. Two right panels: cross sections for the main transfer channels as a function of beam energy (full and empty points) compared with GRAZING calculations with the evaporation of neutrons (lines), with different transfer channels distinguished by color.

PRISMA angles. The width of the elastic + inelastic peak (not shown) is consistent with the energy resolution of PRISMA. Ground-state-to-ground-state  $Q$  values,  $Q_{gs}$ , are indicated by vertical black dotted lines.

Above the barrier, one-proton transfer channels have TKEL distributions with a narrow component centered around the  $Q_{gs}$  value, indicative of direct, quasi-elastic, transfer. A broader component at higher energy losses, more pronounced for the  $-1p$  channel, reflects contributions from deep-inelastic processes. The distributions for  $2p$  and  $2p + 2n$  stripping and pickup transfer channels are progressively broader with increasing number of transferred nucleons and are peaked above  $Q_{gs}$ , indicating increased energy losses and population of more highly excited states.

As the bombarding energy decreases, and particularly below the barrier, all TKEL distributions become narrower, reflecting a transition to predominantly direct processes. The distributions for pure one- and two-proton transfer channels peak closer to  $Q_{gs}$ , with significantly reduced high-TKEL tails. The  $2p + 2n$  stripping and pickup channels exhibit a smooth, broad distributions peaked about 5 MeV above  $Q_{gs}$  and extending up to 20 MeV, suggesting population of excited states in the reac-

tion partners, even at sub-barrier energies. Since higher mass channels have lower primary cross section, evaporation should not significantly influence the measured cross sections, especially for the proton pickup channels.

### 3.2. Total cross sections

The angle- and energy-integrated cross sections for the relevant transfer channels are summarized in Fig. 3 at three selected beam energies. The absolute normalization of the differential cross sections was obtained using measurements at deep sub-barrier energies, where only elastically scattered Fe isotopes are detected in the PRISMA magnetic spectrometer. In this regime, dominated by Rutherford scattering, the spectrometer's effective solid angle could be precisely determined using known Rutherford cross sections and the monitor detectors. These normalization factors were then consistently applied across all beam energies to extract absolute cross sections, independent of any model assumptions.

The pure neutron pickup channels ( $0p$ ), involving one- and two-neutron transfer, as well as one-neutron stripping channel, exhibit iz-

able cross sections. The cross sections for pure one- and two-proton stripping and pickup channels are also relatively large and comparable in magnitude, as are those for the  $2p + 2n$  stripping and pickup channels. A gradual decrease in cross sections with decreasing beam energy is observed for all channels, as expected from reduced reaction phase space.

The experimental cross sections are compared in Fig. 3 with calculations from the GRAZING code [46–48]. In this semiclassical approach, two colliding ions interact through a Coulomb plus nuclear interaction and may exchange nucleons. The two nuclei are described as ensembles of independent nucleons, with degrees of freedom that include surface vibrations and single-particle degrees of freedom. Surface mode excitations are treated using a macroscopic approximation, with form factors proportional to the radial derivative of the ion-ion potential and strengths given by experimental  $B(E\lambda)$  values. The model, for each transfer mode, stripping and pickup of neutrons and protons, uses a representative form factor that is parameterized in accordance with Ref. [49], taking into account the single-particle properties of the two colliding ions. The different single-particle states that participate in the transfer process are described by introducing average single particle level densities. The exchange of nucleons is treated independently and in the successive approximation.

In comparing GRAZING results with experimental data one has to take into account that the closed  $N = 28$  shell nucleus  $^{54}\text{Fe}$  has the smallest charge radius among all measured Fe isotopes, with a deep minimum, as shown by high-precision laser spectroscopy measurements [50–52]. Therefore, the radius of the ion-ion potential was reduced for the present system by 0.2 fm compared to standard values, which accounts for the correct fall-off (onset of absorption) of the experimental elastic + inelastic over Rutherford cross sections. Accordingly, the strength of the transfer form factors had to be reduced (by a factor close to two) at the measured internuclear distances.

The one-neutron pickup channel shows a higher cross section than the corresponding stripping channel, in agreement with theoretical predictions, as illustrated in Fig. 3. The two-neutron pickup is found to be enhanced, both with respect to the one-neutron channel and to theoretical calculations. After the transfer of two neutrons, a sharp decline in the cross section is predicted, of over an order of magnitude, for three- and four-neutron transfer channels, whose yields fall below the present experimental sensitivity. These trends are observed at all measured bombarding energies. The dependence of the absolute cross sections on beam energy is illustrated more clearly in the two rightmost panels of Fig. 3, which summarize the results for all measured energies. The one-neutron stripping cross sections decrease more steeply with decreasing beam energy than predicted by GRAZING, possibly due to an overestimation of evaporation effects in the calculations arising from the much stronger nearby channel at sub-barrier energies. The yield for the two-neutron stripping channel could not be reliably extracted owing to contamination from neighboring channels. While the one-neutron pickup cross sections are reasonably well reproduced by the calculations across the measured energy range, two-neutron pickup cross sections are larger than predicted. These larger cross sections, particularly at sub-barrier energies where evaporation effects are minimal and transfer is dominated by quasi-elastic processes, suggest that neutron-neutron correlations may play an important role in this system.

For the pure one-proton pickup and stripping channels, the agreement between the experimental data and the theoretical calculations remains generally very good. On the proton stripping side, the  $-1p - 1n$  yield could not be reliably extracted due to contamination from nearby channels. The  $-1p + 1n$  channel is a very complex case, as it involves a charge-exchange process in which a proton and a neutron are transferred in opposite directions. Its cross section turns out to be well reproduced by GRAZING, possibly indicating that, in this case, nucleon-nucleon correlations do not play a significant role. In contrast, the cross section for the  $+1p + 1n$  channel appears enhanced compared to the calculations which are based on an independent proton and neutron transfer, sug-

gesting that neutron-proton correlations may play a role in the transfer process.

For multiple proton transfers, the integrated cross sections show a large enhancement for the two-proton pickup and stripping channels compared to both one-proton transfer and theoretical calculations. A particularly striking feature is the presence of strong  $2p + 2n$  channels on both the pickup and stripping sides. In fact, the  $\pm(2p + 2n)$  cross sections are comparable to those of the  $\pm 2p$  channels, with the  $+2p + 2n$  transfer channel even exceeding the  $+2p$  channel in the two-proton pickup distribution. It is interesting to note that the  $-2p$  and  $-2p - 2n$  cross sections become nearly equal at sub-barrier energies. Furthermore, as shown in Fig. 3, the  $\pm(2p + 2n)$  cross sections exceed those of the neighboring  $\pm(2p + 1n)$  channels, deviating significantly from the GRAZING calculations based on uncorrelated, sequential nucleon transfer. This behavior cannot be reproduced assuming an independent particle transfer, where one has a much steeper decrease in the cross section with increasing number of transferred nucleons. Instead, it suggests that nucleon-nucleon correlations may play an important role in the transfer process.

It is important to note that GRAZING has not been extensively tested for proton transfer channels, particularly for proton pickup reactions. In previous studies, discrepancies between experimental data and GRAZING predictions for proton pickup channels were attributed, besides the limited knowledge of the single-particle level density for protons, to the large energy losses observed, for example, in the  $^{40}\text{Ar} + ^{208}\text{Pb}$  system [53]. Therefore, TKEL distributions were also calculated using the GRAZING code and are shown as filled gray histograms in Fig. 2. The theoretical distributions were normalized separately channel by channel for each of the measured energies to allow a direct comparison with the experimental shapes. At the lowest energy, the shapes of the experimental TKEL distributions are well reproduced, indicating that the transfer mechanism is correctly described as a predominantly direct process and that the range of partial waves was correctly included in the code. However (see Fig. 3), the calculated cross sections for the two-proton and two-proton-two-neutron transfer channels, based solely on independent nucleon exchange, significantly underestimate the experimental cross sections.

Experimentally, the  $+2p$  and  $+2n$  channels are enhanced compared to the square of the one-nucleon transfer probabilities. The observed enhancement of two-proton and two-neutron transfer channels should indicate that nucleons of the same species behave like correlated pairs. At the same time, the transfer probabilities of the “ $\alpha$ -like” transfer channel are much larger than the product of the individual  $2p$  and  $2n$  transfer probabilities, or the square of the  $+1p + 1n$  transfer probabilities, which in turn indicates the effect of higher-order correlations. While the inclusion of these higher-order correlations appears to be needed to account for the observed cross sections, this does not directly imply an effect of a spatially localized cluster. Instead, this can be viewed, in a very simplified picture, in terms of  $\alpha$ -type correlations, where four nucleons form dynamically correlated configurations while remaining at relatively large distances moving in the low-density region near the surface of the heavy nuclei [54–56]. In heavy nuclei, the interaction is dominated by the mean field, and consequently  $\alpha$ -type correlations are not expected to play a major role. At variance, the lower nucleon density at the surface may favour the onset of such correlations [54,57–59]. In our experiment, the prominence of  $\alpha$ -type transfer channels increases as the beam energy decreases toward and below the Coulomb barrier, probing more peripheral collisions and thus the nuclear surface. From the current experimental data, it is not possible to determine whether the four nucleons are first coupled as a proton-proton and neutron-neutron pair (pp + nn) [55], and/or if these correlations arise from neutron-proton pairs (np-np) [60]. These observations suggest that multinucleon transfer reactions below the barrier provide evidence of correlations at the nuclear surface, however, their inclusion in reaction models remains a non-trivial task that requires further consideration.

For the two-proton transfer, one must also consider the effect of the  $Q$ -value window. In general, the overall distribution of excitation energy depends on the overlap of  $Q$ -value window with the threshold for allowed states, and the density of states in the reaction products that determines how likely each is to reach a certain level of excitation. It is possible that excited  $0^+$  states, or states with larger angular momentum generated by higher order correlations, may play a significant role. To investigate the population of such excited states, our inclusive data set is compared with light-ion transfer reactions reported in the literature. In particular, light-ion  $\alpha$ -transfer reactions such as  $^{54}\text{Fe}(^6\text{Li}, d)^{58}\text{Ni}$ , which are known for their selective population of states of specific structure, have shown a strong population of the excited  $0^+$  state at 3.53 MeV and  $1^-$  state at 6.02 MeV [61]. This selective excitation has been attributed to the shell structure of the  $^{54}\text{Fe}$  target nucleus which has two proton holes in the  $f_{7/2}$  orbital. In forming the ground state of  $^{58}\text{Ni}$ , these holes would be filled and two neutrons would be added to the  $p_{3/2}$  orbital. The observed strong  $0^+$  excited state may correspond to configurations where all four transferred nucleons occupy orbitals outside the  $f_{7/2}$  shell, possibly the  $p_{3/2}$ , leaving the two  $f_{7/2}$  proton holes. As a result,  $\alpha$ -transfer reactions seem to preferentially populate such configurations where neutrons and protons are in the same orbitals, while weakly exciting the ground state. Similar selectivity has been observed in  $^{54}\text{Fe}(^{16}\text{O}, ^{12}\text{C})^{58}\text{Ni}$  reactions, where a strong population of limited number of states between 4 and 10 MeV was observed [62–64]. Heavy partner was also studied in the  $^{92}\text{Mo}(d, ^6\text{Li})^{88}\text{Zr}$  reaction, where population of  $2^+$ ,  $3^-$  and  $5^-$  states around 2.5 MeV was observed with similar strength [65]. This might be reflected in our TKEL spectra, where the excitation energies of both reaction partners are embedded. It is important to remind that the TKEL distribution for the two-proton-two-neutron transfer channel in the present heavy-ion experiment peaks approximately 5 MeV above the corresponding  $Q_{gs}$ , even below the Coulomb barrier. Moreover, the ground state is only weakly populated, as also observed in light-ion reactions, further confirming the predominantly direct character of the reaction under study. However, one should not assume that all populated states and their population strengths will be the same in light-ion and heavy-ion reaction. Therefore, the present discussion on how the population of specific states in light ion reactions could be associated with (at least part of) the TKEL distributions in our inclusive heavy-ion reaction measurement is kept at a qualitative level only. On the proton stripping side, there is significantly less data from the two-proton-two-neutron transfer reactions available for  $^{50}\text{Cr}$ , and its heavy partner,  $^{96}\text{Ru}$ . However, for nuclei situated near the middle of the proton  $f_{7/2}$  shell, structural differences between the initial and final states are relatively small. In this region, proton configurations tend to be quite similar, which may help explain the relatively similar observations for the  $-2p - 2n$  transfer channel.

#### 4. Summary

In conclusion, the unexpectedly large cross sections observed for the  $2p$  and  $2p + 2n$  stripping and pickup transfer channels at near- and sub-barrier energies in proton-rich heavy-ion reactions could not have been identified without the availability of large solid-angle magnetic spectrometers and detectors with high  $Z$ ,  $A$ , and  $Q$ -value resolution. These results indicate that pairing correlations, and more in general higher order nucleon-nucleon correlations, play a significant role in multinucleon transfer reactions. Such correlations are reflected in the strengths of the two-proton, two-neutron and two-proton-two-neutron channels, which become more evident at the lowest (sub-barrier) energies. These energies correspond to the largest measured internuclear distances, where absorption effects are minimized, offering the most suitable conditions for the manifestation of nucleon-nucleon correlations. Theoretical developments incorporating such correlations are urgently needed, especially in view of forthcoming experiments with radioactive beams in both proton-rich and neutron-rich regions, where a wealth of interesting new effects is predicted.

#### Data availability

Data will be made available on request.

#### Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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