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## Multistep Coulomb excitation of math xmlns="http://www.w3.org/1998/Math/MathML">mmultiscri pts>mi>Ni/mi>mprescripts>/mprescripts>none>/none>m n>64/mn>/mmultiscripts>/math>: Shape coexistence and nature of low-spin excitations

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Phys. Rev. C 106, 044313 — Published 14 October 2022
DOI: 10.1103/PhysRevC.106.044313

### Multi-step Coulomb excitation of <sup>64</sup>Ni: shape coexistence and nature of low-spin excitations

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(Dated: September 14, 2022)

The structure of <sup>64</sup>Ni, the heaviest stable Ni isotope, has been investigated via high-statistics multi-step safe Coulomb excitation to search for shape coexistence, a phenomenon recently observed in neutron-rich <sup>66</sup>Ni and <sup>70</sup>Ni as well as in doubly-magic, N = 40, <sup>68</sup>Ni. The study was motivated by recent, state-of-the-art Monte Carlo shell-model calculations (MCSM), where a Hamiltonian with effective interactions incorporating the monopole tensor force predicts the existence of the shape coexistence, also in the lower-mass  $^{62,64}$ Ni isotopes. A set of transition and static E2 matrix elements for both yrast and near-yrast structures were extracted from the differential Coulombexcitation cross sections. From comparisons between the new results and MCSM as well as other shell-model calculations, a clearer picture of the structure of <sup>64</sup>Ni emerges. Specifically, the low-spin states are shown to be dominated by proton and neutron excitations mainly within the fp shell, with minimal contribution from the  $g_{9/2}$  shape-driving neutron orbital. The agreement between experimental data and MCSM results indicates a small oblate deformation for the  $0^+_2$  level, and a spherical shape for the  $0^+_3$  state. In addition, the small upper limit determined for the B(E2)probability of a transition associated with the decay of the recently observed 3463-keV,  $0_4^+$  state agrees with its proposed assignment to a prolate shape, herewith providing first evidence for triple shape coexistence in a stable Ni isotope.

#### I. INTRODUCTION

At present, knowledge about the nickel isotopic chain extends from the proton dripline at  ${}^{48}$ Ni [1] to the neutron-rich <sup>78</sup>Ni nucleus [2]. This semi-magic (Z = 28)chain provides opportunities to document the evolution of nuclear structure as a function of neutron excess as it spans the neutron shell closures at N = 20, 28 and 50 as well as a subshell closure at N = 40. By now, the nucleus <sup>68</sup>Ni has been shown to exhibit structural aspects consistent with a double shell closure [3, 4]. More recent interest in its structure has arisen, however, from the observation of triple shape coexistence [5, 6]. Indeed, based on comparisons with the results of Monte Carlo shell-model (MCSM) calculations [5], the ground state has been associated with a spherical shape, the 1605-keV,  $0_2^+$  state with oblate deformation, and the 2511-keV,  $0_3^+$  level with a prolate one. The role of excitations associated with different shapes was subsequently emphasized further by the discovery of a 1567-keV,  $0_2^+$  state in <sup>70</sup>Ni [6, 7]. This level is proposed to be associated with a prolate minimum, based on comparisons with the MCSM calculations, and is located roughly 1 MeV lower in excitation energy than the corresponding prolate  $0_3^+$  state in  ${}^{68}$ Ni. Most recently, a similar situation has been discovered in  ${}^{66}$ Ni, where the first four  $0^+$  states are now known [8, 9]. The ground state and the  $0^+_3$ , 2671-keV level are understood as being associated with sphericity, while the  $0^+_2$ , 2443-

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keV and  $0_4^+$ , 2974-keV states are proposed to be located in oblate and prolate minima, respectively. Coexistence and transitions between spherical and deformed structures in this mass region can be understood in the context of so-called Type-II shell evolution, as a consequence of the action of the monopole component of the protonneutron tensor force between the  $\pi f_{5/2,7/2}$  and the  $\nu g_{9/2}$ orbitals [10–12]. Specifically, the occupation of the  $\nu g_{9/2}$ orbital affects the  $\pi(f_{7/2} - f_{5/2})$  gap, thereby facilitating pair excitations across the Z = 28 gap. An enhanced  $\pi f_{5/2}$  occupation also reduces the energy of the  $\nu g_{9/2}$ subshell, resulting in enhanced neutron particle-hole excitations across the N = 40 gap; i.e., multi particle-hole excitations induce changes in the effective single-particle energies and result in level sequences associated with different shapes. Generally speaking, it is worth noting that, as pointed out in Refs. [13, 14], shape coexistence phenomena in the Ni, Sn and Pb regions of the nuclear chart can all be viewed as resulting from multi particlehole excitations.

Extensive experimental work has shown that the N = 40 neutron subshell gap is rather fragile and that deformed structures become yrast as protons are removed. This is the case, for example, with some of the neutronrich Cr, Mn, Fe and Co isotopes, where the level structures are dominated by deformed configurations and collective excitations involving the shape-driving  $\nu g_{9/2}$  orbital [15–31]. In the Ni isotopic chain itself, signatures of collectivity have been reported in <sup>62,63</sup>Ni [32, 33] through the presence of rotational bands at moderate and high spins ( $I \geq 8$ ). Such bands have thus far not been observed in any heavier isotope: only high-spin sequences characteristic of particle-hole excitations have been observed in <sup>64,66,67,68</sup>Ni [34, 35].

The present work focuses on <sup>64</sup>Ni with the objective to determine, with good precision via multi-step Coulomb excitation, the transition probabilities to as many levels as possible near the ground state in order to provide information on their intrinsic structure in a modelindependent manner and to compare the observations with the results of shell-model calculations carried out with the effective interactions used recently to describe the neutron-rich isotopes. In particular, it should be noted that MCSM calculations [5] predict that the presence of potential minima associated with spherical, oblate and prolate shapes seen in the neutron-rich even Ni isotopes persists down to  $^{64}$ Ni and  $^{62}$ Ni. Two excited 0<sup>+</sup> levels have been observed earlier in  $^{64}$ Ni at respective excitation energies of 2867 and 3026 keV [34]. The deexcitation of both states to the  $2^+_1$  level has been observed and B(E2) transitions probabilities were obtained. Their values indicate that the two states are not associated with prolate deformation. Further, it will be shown that the calculations are able to reproduce the general feeding pattern exhibited by the Coulomb excitation data, and provide an understanding for the absence of collective excitations.

Most recently, while the present work was nearing com-

pletion, evidence for the predicted shape coexistence in  $^{64}$ Ni has been reported in Ref. [36], based on the combination of four experiments carried out at four different laboratories. Proton- and neutron-transfer reactions, neutron capture on a highly radioactive  $^{63}$ Ni (2 GBq) target, nuclear resonance fluorescence and Coulomb excitation had to be measured to reach the sensitivity required to probe shape coexistence in  $^{64}$ Ni. As such, some of the results reported here were first introduced in Ref. [36] as part of the experimental evidence supporting the interpretation in terms of triple shape coexistence.

#### II. EXPERIMENT AND ANALYSIS

Multi-step Coulomb excitation of the nucleus <sup>64</sup>Ni was performed at the ATLAS facility at Argonne National Laboratory where a  $0.5 \text{ mg/cm}^2$  isotopically-enriched  $^{208}$ Pb target, with a 6  $\mu$ g/cm<sup>2</sup> Al front layer and a 40  $\mu g/cm^2$  C backing, was bombarded by a <sup>64</sup>Ni beam at an energy of 272 MeV, a value 14% below the Coulomb barrier. Gamma rays of interest were detected with the GRETINA tracking array [37] in coincidence with the two reaction partners measured by the CHICO2 compact heavy ion counter [38]. At the time of this experiment, 12 GRETINA modules were available, providing a total of 48 highly-segmented coaxial HPGe crystals. The 20 position-sensitive parallel-plate avalanche counters of CHICO2, arranged symmetrically around the beam axis, covered 68% of the solid angle around the target with resolutions (FWHM) of  $1.6^{\circ}$  and  $2.5^{\circ}$  or better in the polar ( $\theta$ ) and azimuthal ( $\phi$ ) angles, relative to the beam axis. Figure 1 illustrates the ability of CHICO2 to discriminate between the two reaction partners in a plot of the difference in their time-of-flight,  $\Delta T_{tof}$ , as a function of the scattering angle. A time resolution (FWHM) of  $\sim 1.2$  ns was achieved in this measurement, and the



FIG. 1: (Color online) Time-of-flight difference between the projectile and target recoils as a function of scattering angle measured with the CHICO2 detector. A clear separation between the  $^{64}$ Ni (bottom) and  $^{208}$ Pb (top) ions is observed.



FIG. 2: (Color online) Total  $\gamma$ -ray spectrum obtained from Coulomb excitation, encompassing the full azimuthal range for CHICO2. The spectrum has been corrected for the Doppler shift assuming <sup>64</sup>Ni kinematics. All labeled  $\gamma$  rays are associated with transitions in <sup>64</sup>Ni. The three most significant transitions for the present discussion are placed in boxes, and correspond to the  $2_1^+ \rightarrow 0_1^+$ ,  $0_2^+ \rightarrow 2_1^+$ , and  $0_3^+ \rightarrow 2_1^+$  transitions, respectively.

mass resolution,  $\Delta M/M$ , was determined to be about 5%.

With CHICO2, the trajectories of the reaction partners could be determined on an event-by-event basis. These were subsequently used in the precise correction of the  $\gamma$  rays for the Doppler shift. Figure 2 presents the spectrum measured in coincidence with <sup>64</sup>Ni projectiles: the  $\gamma$  rays can be associated with known transitions in <sup>64</sup>Ni [34, 39] with the exception of the broad bump around 2.6 MeV. The latter corresponds to the <sup>208</sup>Pb  $3_1^- \rightarrow 0_1^+$  ground-state transition for which the Doppler shift correction is inappropriate. The level scheme of Fig. 3 identifies all the <sup>64</sup>Ni transitions observed in the present work.

For the analysis,  $\gamma$ -ray spectra were obtained for seven ranges of the <sup>64</sup>Ni scattering angle:  $30^{\circ} - 40^{\circ}$ ,  $40^{\circ} - 50^{\circ}$ ,  $50^{\circ} - 60^{\circ}$ ,  $60^{\circ} - 70^{\circ}$ ,  $70^{\circ} - 80^{\circ}$ ,  $96^{\circ} - 130^{\circ}$ , and  $130^{\circ} - 160^{\circ}$ , in order to study the dependence of the Coulomb excitation probabilities on scattering angle. As is customary for this type of measurement, the intensities of all the transitions in Fig. 2 were extracted for each angular range, and corrected for both the detector efficiency and the geometry of the experimental set up. The resulting yields were subsequently analyzed with the semi-classical Coulomb excitation code, GOSIA [40], a coupled-channel, leastsquares fit code which derives matrix elements from a standard  $\chi^2$  analysis. The electromagnetic matrix elements of interest are parameters in a procedure comparing the measured  $\gamma$ -ray yields with calculated ones. At the onset, known B(E2) probabilities, lifetimes, branching and mixing ratios [34, 39], summarized in Table I, were included as constraints of the relevant parameters. Note that the Coulomb-excitation analysis only considers relative yields; e.g., all yields are normalized to that of the 1346-keV  $2_1^+ \rightarrow 0_1^+$  transition. The final transition and diagonal matrix elements, together with associated errors can be found in Tables II and III, respectively.

TABLE I: Literature branching and mixing ratios as well as lifetimes for selected transitions used in constraining the  $\chi^2$  minimization in GOSIA. The lifetime data was taken from Ref. [41]. All others are from ENSDF [39].

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Dranching Ratio	Literature	GOSIA
$(2_3^+) \to 0_1^+/(2_3^+) \to 2_1^+$	0.6(2)	0.61
$(2_3^+) \to 2_2^+/(2_3^+) \to 2_1^+$	0.8(3)	0.63
$2_6^+ \to 2_1^+/2_6^+ \to 0_1^+$	0.33(10)	0.38
Mixing Ratio		
$2_2^+ \to 2_1^+$	0.75(20)	0.69
Lifetime (ps)		
$2^+_1$	1.29(5)	1.28

#### III. RESULTS

The level scheme displayed in Fig. 3 contains the thirteen transitions observed in the experiment. Ten of these were reported in Ref. [34], the most recent work on  $^{64}$ Ni, where excited states were populated in quasi-elastic and deep-inelastic reactions of a 430-MeV  $^{64}$ Ni beam on a thick  $^{238}$ U target. The three additional  $\gamma$  rays placed in Fig. 3 are the 1606-keV transition originating from a 4216-keV,  $4^+$  state and two transitions of 1930 and 3276 keV linking an excited  $2^+$ , 3276-keV level with the  $2_1^+$  and ground states, respectively. These three transitions had also been observed previously [39]. In the context of this paper, the observation of the 1522- and 1680-keV  $\gamma$  rays linking the 2867- and 3026-keV 0<sup>+</sup> excitations to the  $2^+_1$  yrast state are particularly noteworthy as these two levels could possibly be associated with different <sup>64</sup>Ni minima within the Type-II shell evolution picture described above. Furthermore, when comparing the present data with those from the in-beam work of Ref. [34], it is worth noting the difference in the general feeding pattern of observed levels in both measurements with Coulomb excitation populating low-spin, non-vrast states considerably more strongly than is the case following complex reactions above the barrier.

The transition matrix elements, reduced transition



FIG. 3: Level scheme depicting all thirteen known <sup>64</sup>Ni transitions observed in this Coulomb excitation experiment. Parentheses indicate tentative spin-parity assignments. The subscripts on the spin quantum numbers refer to the sequence in which levels of the same spin and parity are observed in this work.

TABLE II: Summary of E1, E2, and M1 matrix elements and reduced transition probabilities for <sup>64</sup>Ni deduced in the present work. Units for reduced transition strengths are  $\mu_N^2$ ,  $e^2 \text{fm}^2$ , and  $e^2 \text{fm}^4$  for M1, E1, and E2 transitions, respectively. Accordingly, E1, E2, and M1 matrix elements are listed in units of  $eb^{1/2}$ , eb and  $\mu_N$ . Here,  $\lambda$  is either E or M and L takes values of 1 or 2. The last column presents the reduced transition probabilities in Weisskopf units (W.u.). Note that the uncertainties are quoted in a format based on whether the errors are symmetric or not.

$E \; (keV)$	$I_i^{\pi} \to I_f^{\pi}$	Mult $(\lambda L)$	$E_{\gamma} \; (\text{keV})$	$\langle i   M(\lambda L)   f \rangle$	$B(\lambda L\downarrow; i\to f)$	$B(\lambda L\downarrow; i \to f) $ (W.u.)
1346	$2^+_1 \rightarrow 0^+_1$	E2	1346	0.268(16)	144(18)	9.5(12)
2277	$2^+_2 \rightarrow 0^+_1$	E2		$0.005\substack{+0.002\\-0.001}$	$0.05^{+0.05}_{-0.02}$	$0.003^{+0.003}_{-0.001}$
2277	$2^+_2 \rightarrow 2^+_1$	E2	931	$0.149_{-0.004}^{+0.007}$	$44^{+4a}_{-2}$	$2.9^{+0.3}_{-0.2}$
2277	$2^+_2 \rightarrow 2^+_1$	M1	931	$1^{+1}_{-3}$	$0.2^{+0.6}_{-0.2}$	$0.11_{-0.11}^{+0.34}$
2610	$4^+_1 \rightarrow 2^+_1$	E2	1264	$0.244_{-0.008}^{+0.013}$	$66^{+7}_{-4}$	$4.4^{+5}_{-3}$
2610	$4^+_1 \rightarrow 2^+_2$	E2		$0.42^{+0.05}_{-0.03}$	$196^{+47}_{-28}$	$13^{+3}_{-2}$
2867	$0^+_2 \rightarrow 2^+_1$	E2	1522	0.070(2)	49(3)	3.2(2)
2867	$0^+_2 \rightarrow 2^+_2$	E2		0.27(4)	729(216)	48(14)
2972	$(2_3^+) \to 0_1^+$	E2	2972	0.009(1)	0.16(4)	0.011(2)
2972	$(2^+_3) \rightarrow 2^+_1$	E2	1626	0.05(2)	$5^{+5}_{-3}$	$0.3^{+0.3}_{-0.2}$
2972	$(2^+_3) \to 2^+_1$	M1	1626	$-0.003^{+0.041}_{-0.033}$	$0.000002^{+0.0003854}_{-0.0000002}$	$0.000001\substack{+0.000215\\-0.000001}$
2972	$(2^+_3) \rightarrow 2^+_2$	E2		$0.03^{+0.08}_{-0.07}$	$1.8^{+22.4}_{-1.8}$	$0.12^{+1.58}_{-0.12}$
2972	$(2^+_3) \rightarrow 2^+_2$	M1		$-0.19^{+0.04}_{-0.08}$	$0.007_{-0.006}^{+0.003}$	$0.004^{+0.002}_{-0.003}$
2972	$(2_3^+) \to 4_1^+$	E2		0.26(5)	135(52)	9(4)
3026	$0^+_3 \rightarrow 2^+_1$	E2	1680	0.030(2)	9.0(12)	0.59(8)
3026	$0^+_3 \rightarrow 2^+_2$	E2		$0.25^{+0.05}_{-0.07}$	$625^{+250}_{-350}$	$41^{+16}_{-23}$
3166	$4_2^+ \rightarrow 2_1^+$	E2	1820	0.249(4)	68.9(22)	4.5(2)
3276	$2^+_4 \rightarrow 0^+_1$	E2	3276	0.030(2)	1.80(24)	0.12(2)
3276	$2^+_4 \rightarrow 2^+_1$	E2	1930	-0.069(5)	9.5(1.7)	0.63(9)
3276	$2^+_4 \rightarrow 2^+_1$	M1	1930	$0.02^{+0.03}_{-0.05}$	$0.00008^{+0.00042}_{-0.00008}$	$0.00005\substack{+0.00024\\-0.00005}$
3396	$4^+_3 \rightarrow 2^+_1$	E2	2050	0.047(5)	2.5(5)	0.16(3)
3560	$3^1 \rightarrow 2^+_1$	E1	2214	0.035(6)	0.018(6)	0.017(6)
4216	$4^+_4 \rightarrow 2^+_1$	E2		0.101(7)	11.3(16)	0.8(1)
4216	$4^+_4 \rightarrow 4^+_1$	E2	1606	$-0.9^{+0.7}_{-0.1}$	$900^{+1944}_{-200}$	$59^{+128}_{-13}$
4216	$4^+_4 \to 4^+_1$	M1	1606	$1.3_{-3.0}^{+0.5}$	$0.19\substack{+0.17 \\ -0.19}$	$0.11\substack{+0.10\\-0.11}$

 $^a{\rm The}$  value of 73(8)  $e{\rm b}$  previously quoted for this transition in Ref. [36] is in error. The present value of  $44^{+4}_{-2}$  eb is correct.

TABLE III: Summary of diagonal matrix elements along with the associated spectroscopic (Q) quadrupole moments in units of *eb* for <sup>64</sup>Ni low-lying states deduced in the present work.

$E_{level}$ (keV)	$I^{\pi}$	$\langle I_i   M(E2)   I_f \rangle$	$Q_s$
1346	$2_{1}^{+}$	0.11(3)	+0.08(2)
2276	$2^{+}_{2}$	$0.30^{+0.03}_{-0.05}$	$+0.23^{+0.02}_{-0.04}$
2610	$4_{1}^{+}$	$-0.4_{-0.3}^{+0.1}$	$-0.30^{+0.08}_{-0.23}$
3166	$4^{+}_{2}$	$0.1^{+0.1}_{-0.3}$	$+0.08^{+0.08}_{-0.23}$

TABLE IV: Comparison of level lifetimes determined in the present work with the adopted values taken from the ENSDF database [39].

E (keV)	$I^{\pi}$	Lifetime (ps)					
L (KeV) 1		ENSDF	Present Work				
1346	$2_{1}^{+}$	1.57(5)	1.28(16)				
2610	$4_{1}^{+}$	2.5(4)	2.13(23)				
2867	$0_{2}^{+}$	2.09(14)	2.04(13)				
2972	$2^{+}_{3}$	$0.19^{+0.19}_{-0.07}$	2.58(12)				
3026	$0_{3}^{+}$	5.2(17)	6.5(9)				
3276	$2_{4}^{+}$	0.35(4)	1.1(8)				

probabilities, and their associated uncertainties are displayed in Table II, while Table III presents the static matrix elements for the four lowest-lying states along with the corresponding spectroscopic quadrupole moments. At the onset of the minimization process, the matrix element for the excitation of the  $\mathbf{2}_1^+$  level from the ground state was kept fixed at the value reported by Allmond et al. [41] following single-step Coulomb excitation of the  $^{64}$ Ni +  $^{12}$ C system. Once the  $\chi^2$  minimum for the full data set was determined, this constraint was removed for subsequent minimizations. The latter were used to determine the correlated uncertainties. The value of 0.268(16)eb reported in Table II for the  $\langle 0_1^+ | M(E2) | 2_1^+ \rangle$  matrix element agrees with the 0.268(5) eb one of Ref. [41], herewith providing confidence in the results of the present analysis. This confidence is further validated by the agreement between the literature data included as constraints in the minimization and the experimentally deduced values (Table I). No Coulomb excitation data for other higher-lying <sup>64</sup>Ni states was available prior to this work. However, a  $B(E2; 4_1^+ \rightarrow 2_1^+)$  transition probability of 6.6 (1.0) W.u. was extracted from a lifetime measurement carried out in conjunction with a g-factor determination [42]. This value is of the same order as the  $4.4^{+5}_{-3}$ W.u. value obtained in the present study. Comparisons are extended further in Table IV, where adopted level lifetimes from the evaluated nuclear structure data file (ENSDF) database [39] are compared with corresponding values derived from the present measurement. It can be seen that the agreement between the two data sets is satisfactory with values of the same order of magnitude in each case as well as similar trends as a function of level energy. Overall, all the measured B(E2) transition probabilities displayed in Table II correspond to values

of the order of a few single-particle units ( $\sim 1-10$  W.u.), consistent with non-collective excitations.

#### IV. DISCUSSION

The present <sup>64</sup>Ni data have been compared with three different sets of shell-model calculations. The first of these, labelled as ii44, is aimed specifically at exploring the role of neutron multi particle-hole excitations from the fp shell into the  $g_{9/2}$  orbital. Thus, the model space is comprised of the  $f_{5/2},\,p_{3/2},\,p_{1/2}$  and  $g_{9/2}$  neutron states and the Hamiltonian incorporates the jj44pna effective interaction proposed by Lisetskiy et al. [43]. The latter is based on an analysis and fit of experimental data on the  $^{57-78}$ Ni isotopes as well as on those for the  $^{77}$ Cu  $^{100}$ Sn isotones available at the time (2004). In addition, the role of excitations involving both protons and neutrons was investigated in calculations within the  $f_{7/2}$ ,  $f_{5/2}$ ,  $p_{3/2}$ , and  $p_{1/2}$  model space with a Hamiltonian including the GXPF1A effective interaction [44]. This interaction has been fine-tuned on large data sets in fp-shell nuclei, and has proved successful in reproducing many structural properties in A = 40 - 70 isotopes ranging from Ca to Ni, including the onset of new neutron subshell closures at N = 32 and 34 [44–46]. Calculations with this interaction are labelled as fp hereafter. Finally, a more global view of excitations in <sup>64</sup>Ni was attempted with calculations within the Monte Carlo Shell-Model framework (MCSM) of Refs. [12, 47–49]. These calculations are of the same type as those presented earlier for  ${}^{6678}$ Ni [8, 12]. In this case, however, the model space encompasses protons and neutrons in the full fp shell with, in addition, the  $g_{9/2}$  and  $d_{5/2}$  orbitals. The Hamiltonian is based on the modified A3DA effective interaction used for comparisons with data, for example, on neutron-rich Ni isotopes in Refs. [5, 6, 8].

A comparison between the level energies observed in the present work and the results of the various calculations is presented in Figs. 4 (a) - (d). Note that negativeparity states dominate the structure of <sup>64</sup>Ni above the  $4^+$  levels [34]. These excitations were not observed in the present work, with the exception of the 3560-keV  $3_1^$ state which is included in Fig. 3. Hence, they will not be discussed here beyond stating that they are understood as involving mostly one-neutron particle-hole excitations from the fp shell to the  $g_{9/2}$  orbital, based on the shell-model calculations presented in Ref. [34], and those carried out for the present work within the jj44pna Hamiltonian (not shown).

The two more conventional shell-model calculations of the positive-parity states (jj44 and fp) reproduced the data rather well: see Figs. 4 (c) and (d). Quantitatively, results of the former with the jj44pna interaction appear to reproduce the data somewhat better with a root-meansquare (rms) deviation of 242 keV for the states observed in the present study. The corresponding value for the fp results, obtained with the GXPF1A interaction, is 314



FIG. 4: (Color online) Comparison of the results of jj44, fp, and MCSM excitation energy calculations with the <sup>64</sup>Ni states observed in the present Coulomb excitation experiment. Calculated positive-parity levels with spin 0, 2, and 4 up to 4.5 MeV are included. The length of the horizontal lines indicates the spin value. The red lines in the MCSM panel indicate states with relatively large occupancies of  $g_{9/2}$  neutron particles and  $f_{7/2}$  proton holes. The blue line in the fp panel indicates the first level with a relatively large number of  $f_{7/2}$ proton holes, while the red lines in the jj44 panel signal states with a large  $g_{9/2}$  neutron occupancy.

keV. Furthermore, the sequence of levels as a function of spin and excitation energy is reproduced more closely in the jj44 computations. Thus, starting from the  $0^+$ ground state, two  $2^+$  levels are computed to lie lower than the first  $4^+$  excitation, which is in turn followed by the  $0_2^+$  state. It is worth pointing out that the agreement between data and calculations for the  $0^+_3$  level is worse than that for the  $0^+_2$  state as the former is computed nearly 359 keV too high in excitation energy while the latter is predicted to lie lower by only 182 keV. The jj44 results also predict the  $0^+_3$  state to be higher in excitation energy than the  $4^+_2$  level, a result at variance with the data. Within the fp approach, the  $0^+_2$  level is calculated 630 keV too low, but agreement between data and theory is more satisfactory for the  $0^+_3$  state with calculations locating the excitation 215 keV higher. However, the fp Hamiltonian places a number of  $2^+$  and  $4^+$ states below the  $0^+_3$  level, a feature not exhibited by the data. More generally, from Fig. 4 (c) it is clear that the fp level sequence is compressed with respect to the data. This is likely due to the increasing role of neutron pairing brought about by the presence of the  $g_{9/2}$  orbital close to the Fermi surface as one approaches <sup>68</sup>Ni.

Figure 4 (b) provides similar comparisons between the measured and calculated level structures, but within the MCSM framework. In this instance, the 298-keV rms deviation is comparable to those reported above, and well within the level of accuracy to be expected for any of the three calculations. Here as well, the computed states give rise to a spin sequence with less similarity to the data. For example, the  $0^+_2$  state is predicted almost 780

keV too low, while the calculated  $0_3^+$  excitation is low as well, although only by 30 keV. Furthermore, the  $0_2^+$ level is calculated to lie lower than the  $2_2^+$  state, and the ordering of the  $2_3^+$  and  $4_1^+$  levels is reversed as well.

Since <sup>64</sup>Ni possesses a closed major proton shell, it was thought a priori [34] that most of the low-energy states would be associated predominantly with neutron excitations. This is supported by the relative success of the jj44 calculations. However, a closer look at the  $0^+_3$  state, specifically, indicates that this level is, in fact, reproduced best by the MCSM calculations, followed by the fp ones, and that the deviation is largest in the jj44 space. This suggests that the configuration of the  $0^+_3$  level involves a combination of both proton and neutron excitations rather than the expected dominant neutron ones, which reproduce the other low-spin states. The configurations dominated by two neutrons in the  $g_{9/2}$  orbital (red lines in panels (b) and (d) of Fig. 4) start at 2.1 MeV for the MCSM calculations and 3.7 MeV in the case of jj44 computations. Comparison with the experimental energies suggest that the  $(\nu g_{9/2})^2$  configurations start near 3 MeV.

Tables V and VI list the measured strengths and spectroscopic quadrupole moments alongside those obtained from the two shell-model calculations within the ji44 and fp model spaces as well as from the MCSM approach. Transitions for which an experimental value is not listed in the table were not determined via the analysis with GOSIA, but the calculated values are included due to their relevance for the ensuing discussion. As noted above, decays involving the  $3^{-}_{1}$  level at 3560 keV are not listed as the three calculations have not been extended to the negative-parity states. However, the decay from this state to the  $2^+_1$  level is characterized by an E1 transition strength of only 0.0170(2) W.u., strongly implying single-particle character. This strength is also comparable to those of similar  $3^- \rightarrow 2^+_1$  transitions in the other even Ni isotopes: they are all of the order of a few mW.u. [50]. It is noted that, while all three calculations correctly predict a positive quadrupole moment for the  $2^+_1$  state, only the fp computation, and to a lesser extent, the ji44 interaction are able to provide a better agreement with the experimental data in terms of the absolute magnitude. All three values are, however, consistent with a near-spherical ground state. In contrast, the large magnitude and positive sign determined experimentally for the  $Q_s(2_2^+)$  moment are not reproduced in any of the three calculations.

## A. Transition probabilities and occupancies for the $2^+$ states

The computations of reduced transition probabilities within the shell model require the use of effective charges. All three calculations adopt the conventional value of  $e_p = 1.5e$  for the proton effective charge. However, while the MCSM and fp computations also adopt the canon-

TABLE V: Comparison of experimental reduced transition strengths with those obtained via fp, jj44, and MCSM calculations. Units for reduced transition strengths are  $\mu_N^2$  and  $e^2 \text{fm}^4$  for M1 and E2 transitions, respectively. Here,  $\lambda$  is either E or M and L takes values of 1 or 2.

$I^{\pi} \rightarrow I^{\pi}$	Mult $(\lambda L)$	$F_{\rm c}$ (koV)	$B(\lambda L\downarrow; i \to f)$						
$I_i \rightarrow I_f$	$\operatorname{Mult}(\lambda L)$	$L_{\gamma}$ (kev)	Exp	fp	jj44	MCSM			
$2^+_1 \to 0^+_1$	E2	1346	144(18)	139.0	142.5	158.9			
$2_2^+ \to 0_1^+$	E2		$0.05\substack{+0.05\\-0.02}$	2.0	2.2	0.49			
$2_2^+ \to 2_1^+$	E2	931	$44^{+4}_{-2}$	139.6	185.9	127.3			
$2_2^+ \to 2_1^+$	M1	931	$0.2^{+0.6}_{-0.2}$	0.014	0.011	0.011			
$4_1^+ \to 2_1^+$	E2	1264	$66^{+7}_{-4}$	28.9	0.22	138.5			
$4^+_1 \rightarrow 2^+_2$	E2		$196^{+47}_{-28}$	61.5	43.2	88.5			
$0^+_2 \rightarrow 2^+_1$	E2	1522	49(3)	70.9	1.8	182.4			
$0^+_2 \rightarrow 2^+_2$	E2		729(216)	69.2	21.9	695.7			
$(2^+_3) \to 0^+_1$	E2	2972	0.16(4)	7.4	0.25	0.28			
$(2_3^+) \to 2_1^+$	E2	1626	$5^{+5}_{-3}$	20.2	6.2	0.10			
$(2_3^+) \to 2_1^+$	M1	1626	$0.000002\substack{+0.000385\\-0.000002}$	0.026	0.0054	0.00001			
$(2_3^+) \to 2_2^+$	E2		$1.8^{+22.4}_{-1.8}$	7.7	1.4	161.8			
$(2_3^+) \to 2_2^+$	M1		$0.007\substack{+0.003\\-0.006}$	0.028	0.067	0.001			
$(2_3^+) \to 4_1^+$	E2		135(52)	0.48	11.5	20.2			
$0_3^+ \to 2_1^+$	E2	1680	9.0(12)	0.21	1.5	36.2			
$0_3^+ \to 2_2^+$	E2		$625^{+250}_{-350}$	31.9	2.1	6.1			
$4_2^+ \to 2_1^+$	E2	1820	68.9(22)	133.7	140.1	114.7			
$4_2^+ \to 2_2^+$	E2			2.8	2.1	14.7			
$2^+_4 \rightarrow 0^+_1$	E2	3276	1.80(24)	2.4	0.006	5.7			
$2^+_4 \rightarrow 2^+_1$	E2	1930	9.5(1.7)	11.2	0.2	11.3			
$2^+_4 \to 2^+_1$	M1	1930	$0.00008\substack{+0.00042\\-0.00008}$	0.0022	0.000009	0.011			
$4_3^+ \to 2_1^+$	E2	2050	2.5(5)	8.2	10.1	6.2			
$4_3^+ \to 2_2^+$	E2			3.9	0.06	120.1			
$4_3^+ \to 4_2^+$	E2			24.6	9.6	108.1			
$4_4^+ \to 2_1^+$	E2		11.3(16)	3.8	1.9	1.8			
$4_4^+ \to 4_1^+$	E2	1606	$900^{+1944}_{-200}$	1.7	5.4	0.08			
$4_4^+ \to 4_1^+$	M1	1606	$0.19\substack{+0.17\\-0.19}$	0.34	0.018	0.0079			

TABLE VI: Experimental spectroscopic quadrupole moments,  $Q_s$  for low-lying states in <sup>64</sup>Ni in comparison with results of the shell-model calculations discussed in the text.

$E_{level}$ (keV)	$I^{\pi}$	$Q_s$ (eb)	fp	jj44	MCSM
1346	$2^{+}_{1}$	+0.08(2)	+0.12	+0.04	+0.20
2276	$2^{+}_{2}$	$+0.23^{+0.02}_{-0.04}$	-0.10	-0.02	-0.03
2610	$4_{1}^{+}$	$-0.30^{+0.08}_{-0.23}$	+0.29	+0.26	+0.35
3166	$4_{2}^{+}$	$+0.08^{+0.08}_{-0.23}$	+0.08	+0.14	+0.27

ical 0.5e value for the neutron effective charge, the jj44 ones use  $e_n = 1.2e$ . As stated above, the jj44 calculations only consider neutron excitations, including those involving neutrons in the  $g_{9/2}$  orbital. As protons are ignored in this case, their contribution to the B(E2) value is mimicked by boosting  $e_n$  to the value required to reproduce the  $B(E2; 2_1^+ \rightarrow 0_1^+)$  transition probability.

The strength of the 1346-keV ground state transition is computed satisfactorily as the measured value of 144(18)  $e^2 \text{fm}^4$  is reproduced within errors by all three calculations (see Table V). This result is significant only for the fp and MCSM calculations as they are carried out with the standard effective charges. In fact, this agreement can be viewed as reinforcing confidence in the present data. In the case of the jj44 computations, the calculated  $142.5 \ e^{2} \text{fm}^{4}$  value solely reflects the fact that the adjustment of the neutron effective charge seems appropriate. An inspection of the occupancies in the three calculations (see Table VII) reveals that the proton  $f_{7/2,5/2}$ ,  $p_{3/2,1/2}$ configurations in the fp and MCSM spaces are similar for the  $2_1^+$  level; e.g. the  $\pi g_{9/2}$  and the  $\nu g_{9/2}$  and  $\nu d_{5/2}$ occupations are found to be negligible in the MCSM approach. It is worth noting that this observation of a small occupation of proton orbitals across the shell gap is valid for all the levels discussed in the present work. Furthermore, the respective neutron occupancies in the MCSM and fp results are similar in the  $0^+_1$  and in the  $2^+_1$  levels. Hence, no significant difference between these two calculated strengths should be expected. In addition, the neutron occupancies in the MCSM and jj44 calculations are similar as well, with the  $\nu g_{9/2}$  orbital occupancy in the wavefunction at only  $\sim 0.6$ .

The  $2_2^+ \rightarrow 0_1^+$  transition strength is experimentally determined to be quite small, with a value of  $0.05^{+0.04}_{-0.02}$   $e^2 \text{fm}^4$ . Indeed, all three calculations reproduce this value rather well, with the MCSM one matching it best with a strength of 0.49  $e^2 \text{fm}^4$ . For the  $2_2^+ \rightarrow 2_1^+$  transition, the

τπ				π		ν					
1"	Calculation	$f_{7/2}$	$p_{3/2}$	$f_{5/2}$	$p_{1/2}$	$g_{9/2}$	$f_{7/2}$	$p_{3/2}$	$f_{5/2}$	$p_{1/2}$	$g_{9/2}$
$0^+_1$	fp	7.51	0.37	0.1	0.03		7.92	3.22	4.03	0.83	
-	jj44							3.04	3.57	0.74	0.65
	MCSM	7.35	0.40	0.1	0.03	0.10	7.84	3.14	3.58	0.82	0.58
$2^{+}_{1}$	fp	7.38	0.48	0.1	0.03		7.92	3.22	4.03	0.83	
1	jj44							3.04	3.57	0.74	0.65
	MCSM	7.1	0.59	0.14	0.05	0.10	7.82	3.10	3.31	1.07	0.66
$2^{+}_{2}$	fp	7.41	0.46	0.1	0.03		7.94	3.4	3.83	0.84	
2	jj44							3.12	3.57	0.92	0.39
	MCSM	7.0	0.65	0.19	0.07	0.09	7.80	3.05	3.47	0.87	0.77
$2^{+}_{3}$	fp	7.41	0.44	1	0.03		7.94	3.36	3.62	1.08	
5	ji44							2.24	4.88	0.26	0.62
	MCSM	6.38	1.04	0.34	0.14	0.09	7.69	2.68	3.05	1.16	1.39
$2^{+}_{4}$	fp	7.58	0.32	0.09	0.02		7.93	2.12	4.19	0.77	
4	ji44							3.24	3.24	1.11	0.41
	MCSM	7.33	0.44	0.09	0.03	0.09	7.86	3.19	3.47	1.09	0.36

TABLE VII: Orbital occupancy values associated with  $2^+$  states in  ${}^{64}$ Ni, as predicted by the fp, jj44, and MCSM calculations. The occupancy values for the  $0_1^+$  state are included for reference. All proton occupancy values correspond to zero for the jj44 computation due to the fact that proton excitations were excluded from the model space (see text for details).

strengths determined by all three calculations are larger than that to the ground state. They are also larger than the  $44_{-2}^{+4} e^2 \text{fm}^4$  measured value. While the fp and MCSM strengths are comparable in this case, the jj44 calculation overpredicts the value even more, computing a strength of 185.9  $e^2 \text{fm}^4$ . This overprediction is likely due to the artificially large effective charge of the neutrons applied in the jj44 calculations. Hence, this overprediction in comparison to the other two values also suggests that the majority of the wavefunctions involved in the  $2^+_1$  and  $2^+_2$  levels occur within the fp space. Moreover, all three calculations also reproduce small M1 values similar to the small  $0.1_{-0.1}^{+0.6} \mu_n^2$  value observed experimentally.

A closer examination of the neutron occupancies for the  $2^+_2$  level reveal a general agreement between all three computations. The most significant differences between the MCSM calculations and the conventional shell model approaches appear to be slight reductions in the  $f_{7/2,5/2}$ and  $p_{3/2}$  MCSM occupancies, compensated by a larger occupancy of the  $g_{9/2}$  neutron orbital. The MCSM value of 0.77 for this  $g_{9/2}$  occupation number is larger than the analogous value of 0.39 in the jj44 space, indicating that this increase in occupancy is likely one of the main disparities between the wavefuction associated with the  $2^+_2$  level and the  $0^+_1$  state. Additionally, while the occupation numbers for the proton orbitals remain mostly unchanged between the  $2^+_2$  level and the ground state in the fp space, and the MCSM values agree for the most part with those of the fp calculations, the MCSM occupancies still indicate some movement of protons from the  $f_{7/2}$  to the  $p_{3/2}$  orbital. Hence, the calculations of the  $2_2^+ \rightarrow 0_1^+$  transition strength indicate that proton excitations may play a minor role in the  $2_2^+$  wavefunction.

A rather small transition strength of 0.16(4)  $e^2 \text{fm}^4$  was measured for the  $2_3^+ \rightarrow 0_1^+$  transition, which both the MCSM and jj44 calculations reflect well (see Table V). The fp calculation, on the other hand, produces a considerably larger value of 7.4  $e^2$  fm<sup>4</sup>, suggesting that this transition is likely dominated by neutron excitations outside the fp model space. This interpretation is supported by the measured strength of the  $2^+_3 \rightarrow 2^+_1$  transition, which is calculated well by both the MCSM and jj44 computations but, once again, overpredicted by the fp calculations. The small M1 transition strength further emphasizes the prevalence of neutron excitations. The small experimental  $2^+_3 \rightarrow 2^+_2$  transition strength of  $1.8^{+9.6}_{-8.4} e^2 \text{fm}^4$ is predicted reasonably well by the fp calculations and even more so by the jj44 ones, whereas the MCSM result drastically overshoots, predicting a value of  $161.8 \ e^2 \text{fm}^4$ . Here, not much more is revealed by the weak B(M1)transition. The final decay mode involving the  $2^+_3$  level worth noting is the  $2_3^+ \rightarrow 4_1^+$  transition. The large experimentally determined strength of 135(52)  $e^2 \text{fm}^4$  is best reproduced by the ji44 and MCSM calculations but with significantly smaller values of 11.5 and 20.2  $e^2$  fm<sup>4</sup>, respectively, whereas the fp value of  $0.42 \ e^2 \text{fm}^4$  is even lower. From the de-excitation pattern of the  $2^+_3$  level, it appears that this state is dominated by neutron excitations and probably involves the  $g_{9/2}$  intruder orbital.

The proton occupancies for the  $2_3^+$  state differ somewhat between the fp and MCSM calculations, with the MCSM results indicating a drop in the  $f_{7/2}$  occupation number and a redistribution of protons, predominantly to the  $p_{3/2}$  orbital. Similar to the  $2_2^+$  level, the MCSM results predict a relatively high contribution from the neutron  $g_{9/2}$  orbital (1.39 neutrons). Notably, the jj44 occupancies display a significant drop in both p orbitals and an increase in the  $f_{5/2}$  one, in addition to the  $g_{9/2}$  occupancy. This suggests that it is likely the larger neutron occupancy that brings the MCSM calculations in agreement with the observed  $2_3^+ \rightarrow 0_1^+$  transition strength. The  $2_3^+ \rightarrow 2_2^+$  decay branch, on the other hand, is more challenging to comprehend by following the same reasoning. It is understandable in the jj44 calculations that the disparity in the occupancy of the  $g_{9/2}$  orbital between the two states would lead to a smaller transition strength. However, assuming that this applies for neutrons in the MCSM result as well, it must then be concluded that the rearrangement of the protons in the MCSM case (with respect to the fp one) leads to such a large effect in the transition strength.

The measured strength of 1.80(24)  $e^2 \text{fm}^4$  for the  $2^+_4 \rightarrow$  $0_1^+$  transition is well reproduced by both fp and MCSM calculations, while the jj44 value, corresponding to 0.006  $e^{2}$ fm<sup>4</sup>, appears to be too small and, thus, argues for the strength being associated with excitations predominantly within the fp space. The same conclusion is reached for the  $2_4^+ \rightarrow 2_1^+$  transition, with the fp and MCSM calculations, once again, closely predicting the observed value of 9.5(1.7)  $e^{2}$  fm<sup>4</sup>, while the jj44 computation provides too small a value  $(0.2 \ e^2 \text{fm}^4)$ . Unfortunately, the M1 transition strength does not reveal much in this case, as each set of calculations predicts it to be quite small. The proton occupancies of the  $2_4^+$  level with respect to that of the ground state are nearly the same for both the fp and MCSM calculations, suggesting that the difference in wavefunctions between the two levels largely arises from neutron contributions. In terms of neutron occupancies, the fp calculations compute a drop in the  $p_{3/2}$  neutron number, while the MCSM calculations do not. Instead they indicate a drop in the  $g_{9/2}$  orbital occupancy with respect to the ground state. However, the fact that the fp value reproduces the data well in this case may indicate that  $g_{9/2}$  orbital excitations do not play as significant of a role in this state, and that, instead, the redistribution of neutrons to the  $p_{3/2}$  orbital may be more relevant. With respect to the  $2_1^+$  level, both fp and MCSM calculations predict a small reorganization of protons from the  $p_{3/2}$ state back to the  $f_{7/2}$  orbital, but, for the most part, the proton occupancies remain quite similar between the two states. From the considerations above, it can be concluded that the  $2_4^+$  level corresponds to an excitation within the fp shell.

A trend similar to that discussed for the  $2^+$  states is observed for the four  $4^+$  levels populated in the present study, but these are not all discussed here. Rather, a detailed analysis of their configurations and occupancies can be found in Ref. [51]. The calculations all indicate that the low-lying levels in <sup>64</sup>Ni are dominated by excitations involving the  $\nu(f_{5/2}, p_{3/2}, p_{1/2})^8$  neutron configuration so that, to first order, the B(E2) transition probabilities computed within the fp and jj44 model spaces would be expected to be similar, except for an overall scaling factor coming from the excitation of a  $f_{7/2}$  proton into the  $\pi(f_{5/2}, p_{3/2}, p_{1/2})$  configuration. This expectation is not borne out for the  $B(E2; 4^+_1 \rightarrow 2^+_1)$  probability (Table V) where the fp calculations are in better agreement with experiment, herewith indicating that the effective Hamiltonians within the  $\nu(f_{5/2}, p_{3/2}, p_{1/2})^8$  model space are still different for the two interactions. In the case

of the  $4_1^+ \rightarrow 2_1^+ E^2$  matrix element, the difference can be attributed to a large cancelling contribution involving  $f_{5/2}$  neutrons [51] which is absent in the fp model.

## B. Transition probabilities and occupancies for the $0^+$ states

The main focus of this work is on the nature of the excited  $0^+$  states and their relationships with the  $2^+$  and  $4^+$  levels. In this regard, both the MCSM and fp interactions provide strong indications about the nature of  $0_2^+$  state. The measured strength of  $49(3) e^2 \text{fm}^4$  for the  $0^+_2 \rightarrow 2^+_1$  transition is relatively large. It is overestimated by both MCSM and fp calculations with respective values of 182.4 and 70.9  $e^2 \text{fm}^4$ . This, however, is a satisfactory result considering the degree of agreement between data and calculations observed for the  $2^+$  and  $4^+$  states. The most striking feature in this respect is the small jj44 strength of  $1.8 \ e^2 \text{fm}^4$ . Furthermore, the data for the  $0^+_2 \rightarrow 2^+_2$  decay mode lead to a similar observation: the 729(216)  $e^{2}$  fm<sup>4</sup> strength determined by the present measurement is large, and both the MCSM  $(695.7 \ e^2 \text{fm}^4)$  and the fp  $(69.2 \ e^2 \text{fm}^4)$  computations predict large strengths, while those within the jj44 space produce the smallest value of 21.9  $e^{2}$  fm<sup>4</sup>. This suggests that the wavefunction for the  $0^+_2$  level is dominated by configurations within the fp space, and that neutron excitations alone cannot fully account for the description of this state. This conclusion is reinforced by exploring the proton and neutron occupancies of Table VIII. It can be seen, from comparing the  $0^+_2$  and  $2^+_1$  levels, that the proton occupancies remain the same within the fp calculations. On the other hand, the MCSM result indicates a reduction of the  $f_{7/2}$  orbital population for the  $0^+_2$  level, which appears to be redistributed into the  $p_{3/2}$  and  $f_{5/2}$ orbitals. In terms of the neutrons, the occupancies of these two states are, once again, similar for the fp calculations, with the main difference being a reduction in the  $f_{5/2}$  occupation and a slight increase in the  $p_{1/2}$  one. With regard to the MCSM computations, a significant increase in the  $g_{9/2}$  occupation value up to 1.96 neutrons is computed. This is correlated with a depletion in that of both the  $p_{3/2}$  and  $f_{5/2}$  orbitals. Correspondingly, the jj44 calculations confirm a small  $g_{9/2}$  neutron occupation. It can, thus, be concluded that the occupancies reinforce the observations drawn from the B(E2) strengths and, as neutrons contribute with the opposite sign to the reduced transition probability, the MCSM strengths would be expected to be smaller than those of the fp calculations, as the comparison between experiment and theory indicate.

The MCSM calculations predict the largest strength with a value of 36.2  $e^2 \text{fm}^4$  for the  $0^+_3 \rightarrow 2^+_1$  transition, which is relatively close to the experimental value of 9.0(12)  $e^2 \text{fm}^4$ . While the jj44 calculations are also within an order of magnitude, the fp ones are significantly underestimated. This difference with the observa-

τπ		$\pi$						ν				
$I^{*}$	Calculation	$f_{7/2}$	$p_{3/2}$	$f_{5/2}$	$p_{1/2}$	$g_{9/2}$	$f_{7/2}$	$p_{3/2}$	$f_{5/2}$	$p_{1/2}$	$g_{9/2}$	
$0^{+}_{1}$	fp	7.51	0.37	0.1	0.03	· · · ·	7.92	3.22	4.03	0.83		
-	jj44							3.04	3.57	0.74	0.65	
	MCSM	7.35	0.40	0.1	0.03	0.10	7.84	3.14	3.58	0.82	0.58	
$2^{+}_{1}$	fp	7.38	0.48	0.1	0.03		7.92	3.22	4.03	0.83		
1	jj44							3.04	3.57	0.74	0.65	
	MCSM	7.1	0.59	0.14	0.05	0.10	7.82	3.10	3.31	1.07	0.66	
$0^{+}_{2}$	fp	7.38	0.49	0.1	0.04		7.92	3.47	3.33	1.28		
2	jj44							3.56	2.58	1.5	0.35	
	MCSM	5.97	1.30	0.46	0.18	0.08	7.62	2.40	2.79	1.18	1.96	
$0^{+}_{3}$	fp	7.61	0.29	0.09	0.02		7.9	2.67	4.97	0.46		
0	jj44							4.88	2.24	0.26	0.62	
	MCSM	7.29	0.47	0.10	0.03	0.10	7.82	3.44	3.12	1.20	0.38	

TABLE VIII: Orbital occupancy values associated with  $0^+$  states in  ${}^{64}$ Ni, as predicted by the fp, jj44, and MCSM calculations. The occupancy values for the  $2^+_1$  state are included for reference. All proton occupancy values correspond to zero for the jj44 computation due to the fact that proton excitations were excluded from the model space (see text for details).

tions for the  $0_2^+$  level indicates that, in this case, the  $g_{9/2}$ orbital plays a larger role, which cannot be accounted for within the fp space. Moreover, it appears that both proton and neutron excitations must be included, based on the examination of the  $0^+_3 \rightarrow 2^+_2$  transition strength. Here, the measured strength is large  $[625^{+250}_{-350} \ e^2 \text{fm}^4]$ , and mimicked to a degree by the fp calculations (31.9)  $e^{2}$  fm<sup>4</sup>), whereas the ji44 and MCSM computations derive strengths smaller by an order of magnitude. From these, it can be concluded that proton configurations within the fp space, as well as neutron ones, including those from the  $g_{9/2}$  orbital, are involved in the  $0^+_3$  wavefunction. A further examination of the predicted orbital occupancies supports this interpretation. For the protons, the occupancy values are quite similar for both the fp and MCSM model spaces, as concluded above from the transition strengths. For the neutrons, the occupation of the  $g_{9/2}$  orbital in the jj44 calculations is nearly the same as that of the  $2^+_1$  state, which is consistent with the observation that, in this case, the jj44 calculations perform better than the fp ones. However, the  $g_{9/2}$  occupancy in the ji44 calculations is larger than that given by the MCSM ones, which could perhaps account for the fact that the MCSM computations come closest to reproducing the experimental value; i.e., a smaller  $g_{9/2}$  occupation leads to a larger transition strength. Overall, the patterns observed from the transition strengths and the occupancy values for the  $0_2^+$  and  $0_3^+$  states can, thus, be understood in a consistent picture.

While the detailed discussion of transition strengths and orbital occupancies presented above provide an understanding of the multi-step Coulomb excitation data described in this work, it is, however, worthwhile to examine more globally the general trends exhibited by the data and to compare these with the calculations. Figs. 5 and 6 provide the so-called transition networks generated from the shell-model calculations within the fp and jj44 spaces, respectively. Here, the excitation energies of the



FIG. 5: (Color online) Plot of the reduced transition strengths connecting states in  $^{64}$ Ni, as predicted by the jj44 calculations (see text for details).

calculated states are represented as dots while the connecting lines represent the E2 decay paths. The widths of the various lines are directly proportional to the calculated reduced transition probabilities. Note that, for each spin value, many more states than discussed thus far are presented, i.e., the plots extend up to 6-7 MeV above the yrast line.

Both interactions clearly indicate that the ground state and the  $2_1^+$  level are linked by a strong transition strength. This, in turn, implies that the feeding of higherspin states proceeds predominantly through multi-step Coulomb excitation via the  $2_1^+$  level, as seen in the data. Furthermore, both calculations within the two model spaces predict that, beyond the  $2_1^+$  state, the excitation path splits in two directions with nearly comparable



FIG. 6: (Color online) Plot of the reduced transition strengths connecting states in  $^{64}$ Ni, as predicted by the fp calculations (see text for details).

strengths and feeds the  $2^+_2$  and  $4^+_2$  states. This trend is indeed seen in the data: the measured strengths of the  $2_2^+ \rightarrow 2_1^+$  and  $4_2^+ \rightarrow 2_1^+$  transitions are of the same order  $(2.9_{-0.2}^{+0.3}$  W.u. vs 4.5(2) W.u.) and only slightly lower than the 9(1) W.u. probability measured for the ground-state transition as the widths in the two calculations suggest. Based on the jj44 results (see Fig. 5), the  $4_1^+$  state should not be observed. In this instance, the data favor the fp model space as the  $4_1^+ \rightarrow 2_1^+$  strength has been determined experimentally, and found to be smaller than that linking the  $4_1^+$  and  $2_2^+$  states, in qualitative agreement with the calculations in this space. In fact, the similarity between the data and the fp calculations extends clearly to the  $0^+_2$  level, where the computed de-excitation pattern also mirrors that seen in the measurements. Specifically, the noted absence of a  $0^+_2 \rightarrow 2^+_1$ transition in the jj44 prediction is not supported by the data. Indeed, the fact that all the transitions out of the  $0_2^+$  state in Fig. 6 are of comparable width, i.e., of the same order of magnitude, can be viewed as a signature for a small degree of collectivity. This observation agrees with the results of the MCSM calculations that associate the  $0^+_2$  state with a small, oblate deformation.

In Figs. 5 and 6, signatures of strong collectivity are manifest through the appearance of sequences with the same strength linking successive states separated by two units of angular momentum. While no such band is present in the shell-model calculations within the fp space, one is clearly visible within the jj44 computation (Fig. 5). Indeed, a rather clear  $8^+ - 6^+ - 4^+ - 2^+ - 0^+$  sequence appears, terminating in the calculated  $0_4^+$  level at an excitation energy of 3.646 MeV. Assuming that the jj44 calculations are a close representation of the data, the question then arises as to why this  $0_4^+$  state, and by extension, the cascade of collective E2 transitions

built on it was not observed in the present investigation. As indicated in Fig. 5, the strengths of transitions linking the states in the cascade with the lower-lying, near yrast states are small. In terms of multi-step Coulomb excitation, this implies that the cross section for populating the  $0_4^+$  state must be small as well and that, hence, the associated transitions such as the  $0_4^+ \rightarrow 2_2^+$  link would be below the detection limit of the experiment. It is also worth noting that the entire band in Fig. 5 is not calculated to become yrast, even at spin 10 or above. This would also account for the fact that such a collective sequence was not observed in measurements following deep inelastic reactions with the  $^{64}$ Ni +  $^{238}$ U [36] and  $^{26}$ Mg +  $^{48}$ Ca [51] systems.

#### C. Evidence for a $0^+_4$ state

From the discussion above and the calculations illustrated in Figs. 5 and 6, it is clear that observing a  $0_4^+$ state through multi-step Coulomb excitation would be very challenging, if not impossible. Nevertheless, the decay patterns delineated in Fig. 5 as well as the results of similar calculations within the MCSM framework (not shown) stimulated further attempts to identify possible candidates for a  $0^+$  level associated with a prolate minimum. Following a neutron capture measurement [36], a  $0_4^+$  state at 3463 keV which decays in part via a 1187keV transition to the  $2^+_2$ , 2277-keV level was observed. As expected, the  $0^+_4 \rightarrow 2^+_2$  transition was not observed in the present Coulomb excitation data. However, an upper limit of  $1.3 \ e^2 \text{fm}^4$  or 0.08 W.u. was established for the  $B(E2; 0^+_4 \rightarrow 2^+_2)$  transition strength from the spectra at backward angles. With this value, the similarity between the low-spin structures of  ${}^{66}$ Ni and  ${}^{64}$ Ni is striking. Indeed, both nuclei reveal the presence of three excited  $0^+$ states with similar de-excitation patterns. Specifically, the  $0^+_2$  levels in both <sup>66</sup>Ni and <sup>64</sup>Ni decay toward the respective  $2_1^+$  states with strengths of 4.3 W.u. and 3.2(2)W.u., while the  $0^+_3$  states have corresponding B(E2) values of 0.1 and 0.59(8) W.u., respectively. Likewise, the 0.2 W.u. value reported for the  $0^+_4$  state in <sup>66</sup>Ni compares with the 0.08 W.u. upper limit reported here for  $^{64}$ Ni. In view of these observations and based on the results of the MCSM calculations, it is concluded that the  $0^+_1$  and  $0_3^+$  levels are associated with a spherical shape, the  $0_2^+$ state with an oblate one, and the  $0_4^+$  state with a prolate shape. As predicted by these calculations, the excitation energy of the three modes increases with decreasing neutron number: the measured  $0^+_2$  energy increases from 2443 to 2867 keV between <sup>66</sup>Ni and <sup>64</sup>Ni, that of the  $0_3^+$ levels increases from 2671 to 3026 keV, and the  $0^+_4$  states are located at 2974 and 3463 keV, respectively. Hence, the structure of <sup>64</sup>Ni is understood in terms of shape coexistence based on the available experimental evidence. The reader is referred to the work of Ref. [36] for an in-depth discussion.

#### V. CONCLUSIONS

The electromagnetic properties of low-lying states and the possible presence of shape coexisting states in  $^{64}$ Ni were investigated following a multi-step Coulomb excitation measurement. The experiment was performed at the ATLAS accelerator facility using the  $\gamma$ -ray tracking array, GRETINA, in conjunction with the CHICO2 particle detector. Transition and diagonal matrix elements were extracted from the measured  $\gamma$ -ray yields using GOSIA, a semiclassical coupled-channel analysis code. Reduced transition probabilities and spectroscopic quadrupole moments deduced from the matrix elements were compared with those predicted by conventional shell-model calculations with the jj44 and fp effective interactions as well as those within the Monte Carlo shellmodel (MCSM) framework. Transition strengths for the de-excitation of the known excited  $0^+$  states were found to be reproduced well by theory and, in general, supported the view that the nucleus, <sup>64</sup>Ni, closely resembles its neighbor <sup>66</sup>Ni, herewith favoring an interpretation in terms of shape coexistence. In particular, transition strengths and quadrupole moments indicate that the ground and  $0^+_3$  states are spherical, and the  $0^+_2$  level is characterized by a small oblate deformation. In addition, the convergence between the experimentally deduced B(E2) strengths and the calculated ones indicates that most excitations observed in the present data are confined to the fp shell. Furthermore, while more study is necessary in order to further assess the nature of these states, the agreement between experiment and theory, alongside the trends seen in the Ni chain, make a strong

case for shape coexistence in this nucleus.

#### VI. ACKNOWLEDGEMENTS

The authors gratefully acknowledge help with the experiment from K. Auranen, C. M. Campbell, H. L. Crawford, A. B. Hayes, D. Cline, and Krishichayan. This material is based upon work supported by the U.S. Department of Energy, Office of Science, Office of Nuclear Physics, under Grants No. DE-FG02-97ER41041 (UNC), No. DE-FG02-97ER41033 (TUNL), No. DE-FG02-08ER41556 and No. DE-SC0020451 (MSU), No. DE-FG02-94ER40848 (UML), No. DE-FG02-94ER4084 (Maryland), and Contracts No. DE-AC02-06CH11357 (ANL), No. DE-AC52-07NA27344 (LLNL), No. DE-AC02-05CH11231 (LBNL), No. DE-SC0020451 (MSU), by the National Science Foundation under Grants No. PHY-1565546 (MSU), and by the UNC Startup Funds of ADA. The MCSM calculations were performed on the K computer at RIKEN AICS and Oakforest-PACS operated by JCAHPC (hp160211, hp170230, hp180179, hp190160, hp200130). This work was supported in part by MEXT as Priority Issue on Post-K computer (Elucidation of the Fundamental Laws and Evolution of the Universe) and Program for Promoting Research on the Super- computer Fugaku (Simulation for basic science: from fundamental laws of particles to creation of nuclei) and by JICFuS. This research used resources of ANLs ATLAS facility, which is a DOE Office of Science User Facility.

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