

Shape Coexistence at the sub-shell closure 40 Emmanuel Clément

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UNIVERSITE CAEN NORMANDIE Habilitation à diriger des recherches

 Par

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Shape Coexistence at the sub-shell closure 40

Soutenue le 11 Décembre 2019 devant la commission d'examen :

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à Nassima, Zaccharie et Balthazar

 $\gamma\text{-rays}$ always tell the truth

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Introduction

The nuclear shape coexistence is a spectacular and universal phenomena reflecting the competition between different microscopical configurations at low excitation energy corresponding to different nuclear shapes in a single nucleus [1]. In the nuclear chart, in most of the case, the nuclear shape coexistence involves a spherical configuration that coexistences with a deformed configuration. The corresponding states reflect the competition between spherical gap from major or sub-shell closure and the residual energy of the nuclear interaction which minimize the energy of the deformed configuration. The ground state and the low lying excited states corresponding to different shapes coexist in the same nucleus with a difference in excitation energy typically lower than 2 MeV. It illustrates the subtile equilibrium of the nuclear interaction near the ground state. This phenomena is therefore a strong constraint for modern nuclear models and its experimental description remains a challenge involving advanced heavy-ions facilities and experimental setup. In a unified description of shape coexistence [1] for nuclei close to proton shell closures, e.g. Z=20 (Ca [2]), 28 (Ni [3]), and 82 (Pb-Hg [4]), spherical configurations corresponding to closed shells compete with deformed configurations resulting from multi-particle multi-hole excitations above the proton shell closures that interact with neutrons which fill high-j intruder orbits. The significant gain in binding energy for this configuration arising from the pairing and the proton-neutron correlations energy decreases the excitation energy of the corresponding 0^+_2 state in even-even nuclei so that it appears at low excitation energy or even becomes the ground state [1]. Shape coexistence has been the subject of numerous theoretical works and almost all theoretical approaches have been compared with available experimental data. Recently, a major achievement was reached by the use of Monte Carlo Shell Model calculations. These calculations provide for the first time a microscopic description in the neutron rich Ni and Zr isotopes and in neutron deficient Hg isotopes with an accurate reproduction of the experimental data [5, 6, 7, 8]. A second achievement is the first measurements of spectroscopic quadruopole moment of short lived excited states using post-accelerated radioactive beams at ISOL facilities and safe Coulomb excitation technique [9, 10, 11, 12]. Very recently, single nucleon transfer shed a new light on the field [13]. The last decade has also shown that this exotic phenomena is not rare. From isolated case, islands of occurrence appeared all along the nuclear chart, at proton shell closures closed by high j-orbits, involving therefore a possible strong proton-neutron interaction via the tensor force. Finally, some cases appeared in the recent years close to doubly magic nuclei (42 Ca [14], 80 Ge [15], 56 Ni [16], 78 Ni [17]).

The experimental study of shape coexistence is long standing effort. Many experimental probes can be used to discover or characterized shape coexistence in an isotopic chain or in a given nuclei. Shape changes such as the transition from spherical to deformed nuclei can be identified by the

measurement of the excitation energy of the first excited 2^+ state in even-even nuclei. In odd-mass nuclei, having a non zero spin in the ground state, the charge radii or the spectroscopic quadrupole moment extracted from laser spectroscopy, are a clear signature of a shape change. The most emblematic case is the discovery of the charge radii staggering in the Hg isotopes [18, 19, 7] between the ground state and the isomeric state, suggesting a shape coexistence between spherical and deformed configuration. In heavy elements, α -decay have been used to identify excited 0⁺ shape isomer in the light Pb isotopes [20]. E0 $0^{+m} \rightarrow 0^+$ transition by e^- decay is a unique signature of shape coexistence [21, 22, 23, 24, 25]. A large value of the monopole transition strength $\rho^2(E_0)$ reflects a large difference in deformation or/and a large mixing of the 0^+ states wave function [26]. This large mixing can lead to the repulsion of the 0^+ states and increase the excitation energy of the 0^+_2 state. If both the monopole and quadrupole strengths are known [27], the mixing amplitude of the wave functions can be deduced. This strength distribution related to a significant mixing perturbates the relative excitation of the first 2^+ to the ground state (that increases) and the reduced E2 transition probability (which is reduced) from an extrapolation of the collective band at higher angular momentum. This low spin perturbation was interpreted as a signature of mixing related to shape coexistence. A complete illustration is the case of the neutron deficient Kr isotopes [28, 29, 30, 31, 32]. Ideally, one would like to measure directly the shape of both configurations. The quantities directly related to the charge distribution are the charge radii and the electric spectroscopic quadrupole moment. In odd mass nuclei, with states that differ from zero spin, such measurement can be achieved by laser spectroscopy under the condition that both the ground state and the shape isomer live long enough for the measurement. Deformation parameter for a 0^+ state cannot be directly measured since the zero spin prevents to define an orientation needed to define a shape. First 2^+ excited states built on top of the corresponding 0^+ state live from few pico-second to few nano-second. Laser spectroscopy remains impossible. The last decade has revolutionized the study of shape coexistence with the possibility of having post-accelerated radioactive beam at the Coulomb barrier in so-called ISOL facilities like GANIL-SPIRAL1, REX-ISOLDE and ISAC-TRIUMF. With beams intensity > 10^3 pps and efficient γ arrays like EXOGAM [33], MINIBALL [34] and TIGRESS [35], multistep safe Coulomb excitation [36, 37, 38, 39, 40, 41] becomes feasable. From these measurements, using second order effects in the Coulomb excitation cross section, the diagonal E2 matrix element $\langle J^{\pi}|E2|J^{\pi}\rangle$ can be measured in short lived excited states. This quantity is directly related to the spectroscopic quadrupole moment (\mathcal{Q}_s) and its sign. A geometrical conversion allows to determine an axial deformation parameter β_2 . Thus such measurements allow to determine the corresponding shape of excited states and therefore the *unique* way to prove shape coexistence in exotic nuclei involving short lived non-zero spin states.

Shape coexistence with protons and neutrons far from shell closures have been reasonnably well describes within the mean-field approaches using interactions like the Gogny or Skryme functionnals. In these calculations, two clear minima appear in the potential energy surface calculated as a function of the deformed axial (β) and triaxial (γ) degree of freedom. Each minima corresponds to a $J^{\pi} = 0^+$ state in even-even nuclei, from which collective structures are built. In the vicinity of proton major or sub-shell closure, a more microspical description of the low lying 0^+ state was developped by K. Heyde and co-workers [42]. Starting from the nuclear shell structure in medium-heavy and heavy nuclei, the excitation energy for low-lying 0^+ intruder states was investigated. Taking a simplified model with two particle-two hole (2p-2h) excitations across closed shells, the effects of pairing and proton-neutron (monopole and quadrupole components) with high-*j* intruder orbits residual interaction was highlighted. The resulting contribution of the pairing and quadrupole proton-neutron interaction allow the deformed state to gain enough binding energy to become the ground state. The spherical configuration doesn't vanish but appears as an excited configuration. It is a caracteristic of the shape coexistence phenomenum that it is not only an onset of deformation (as for N=90) or the local appearance of a spherical shape (as magic nuclei). But both configurations, spherical and deformed, coexist at low excitation energy and are in competition for the ground state configuration. Only subtle effects in the nuclear interaction make one or the other configuration a candidate for the ground state as minimum of the potential energy of the system. The microscopical description calls for more elaborated calculations. This was recently achieved using Monte Carlo Shell Model calculation in several region benchmarking the phenomenom of shape coexistence [5, 6, 7, 8]. However, it is worth mentioning that, in spite of all these great progresses, the comparaisons with spectroscopic data are always partial. Indeed, most of the advanced theoretical models are compared with experimental data of a given publication. Often, partial level schemes are compared on few isotopes and time to time reduced transition probabilities. Rarely, spectroscopic quadrupole moment from both excited states and ground states are compared at the same level. Even more rarely, monopole transition strength $\rho^2(E_0)$, often evidencing for the first time shape coexistence by the observation of a $0^+ \rightarrow 0^+$ transition, are systematically compared. It can also be underlight that so far, the so-called ab - initio calculations did not yet investigated such shape competition. It will be interesting to evaluate if such spectacular phenomena emerge from first-principle approaches giving more insight in the microscopical origin of the deformation in atomic nuclei and by extension to shape coexistence. Finally, symmetries can play a major role. The islands of inversion have also been described in the Quasi-SU(3) and Pseudo-SU(3) dynamical symmetries framework [43, 44] and can be applied to many area of shape coexistence.

The sub-shell closure at nucleons number 40 plays a major role in the framework of the study of the shape coexistence. This sub-shell closure is not a strong shell closure defining magic or semimagic nuclei as can be clearly seen from the mass measurements. Its appears as the 4^{th} major shell gap in the oscillator potential plus angular momentum but without spin-orbit. It is closing the spdfsequence, last orbit being the l=2 2p, by the l=4 1q orbit. This natural $\Delta=2$ difference calls for large quadrupole collectivity effect when nucleons are excited from the spdf shells to the g orbit. Thanks to the spin-orbit term, the 1g orbit is further splitted between the $1g_{9/2}$ (down in energy) and the $1g_{7/2}$ (increased energy) which define finally the strong N, Z = 50 shell closure responsible for the doubly magicity character of ^{100,132}Sn and ⁷⁸Ni isotopes. This proximity in energy between the $1g_{9/2}$ orbit and the pf shells is the seed for the competition between spherical states with nucleons occupying the pf shells and deformed configurations involving nucleons excitation into the $1g_{9/2}$ orbit. The occupancy of the g-orbit only doesn't provide sufficient gain in binding energy to allow the deformed configuration to become the ground state. Similarly to the N = 20 and N = 28 island of inversions, thus shape coexistence, are triggered by a proton-neutron interaction reducing the spin-orbit splitting induced by the tensor term of the interaction in its $\Delta l=0$ or $\Delta l=1$ channels. Proton-neutron interactions are the final trigger of shape coexistence provinding enough correlation energy to reduce the excitation energy of the deformed configuration. At N = 20, the $p - n \Delta l = 0$ interactions are $\pi d_{5/2} - \nu d_{3/2}$ and $\pi d_{3/2} - \nu d_{3/2}$ as at N = 28, the $\Delta = 1$ interactions are $\pi d_{5/2} - \nu f_{7/2}$ and $\pi d_{3/2} - \nu f_{7/2}$. This scheme can also be applied to isotopes involving *either* a proton or a neutron number 40 or *both* a proton and a neutron number 40 making this nucleon number unique in the nuclear chart.

At Z~ 40 and N~60, the $\pi(g_{7/2} - g_{9/2})$ spin-orbit splitting is reduced when neutrons are promoted into the $\nu g_{7/2}$ and less significantly into the $\nu h_{11/2}$ orbits from the $\nu d_{5/2}$ orbit. Neutron holes interact strongly in the $\Delta l=1$ channel with proton holes in the $\pi f_{5/2}$ orbit created by pair excitation across the Z=40 gap into the $\pi g_{9/2}$ orbit. This proton pair further interacts with a pair of promoted neutron in the $\nu g_{7/2}$ orbit in the $\Delta l=0$ channel. These strong p-n interactions induce the sudden and unique onset of deformation leading to shape coexistence at N=60 in neutron rich Sr/Zr elements which desappears for elements Z > 40 and Z < 37. My contribution in this mass region is described in chapter II.

At Z~28 and N~ 40, the $\pi(f_{5/2} - f_{7/2})$ spin-orbit splitting is reduced when neutron are excited across the N=40 gap and populate the $\nu g_{9/2}$ orbit from the $\nu f_{5/2}$ orbit. Neutron holes interact strongly in the $\Delta l=0$ channel with proton holes in the $\pi f_{7/2}$ orbit created by the pair excitation across the Z=28 gap into the $\pi p_{3/2}$ orbit. The resulting deformed configuration competes at ~ 2MeV excitation energy with the spherical ground state in the ⁶⁸Ni isotope, and is almost degenerated in the ⁶⁷Co isotopes (Z=27) to become the ground state in the Z=26 ⁶⁶Fe nuclei. My contribution in this shape coexistence scenario is described in chapter III.

At $Z \sim N \sim 40$, the description in term of particle-hole excitation of proton interacting with neutron populating high-j intruder orbits is not directly applicable. Being at and near the N=Z line, neutron-deficient isotopes of Ge, Se, Kr, Sr and Zr do not follow the large isospin asymmetry leading to shape coexistence with respect to the two previous cases. Both proton and neutron occupy same orbits and have similar occupancy of the $g_{9/2}$ orbit. This isospin symmetry seems to inihibit the competition between spherical and deformed configuration. A shape coexistence scenario between spherical and deformed states was proposed in ^{70,72,74}Ge and will not be discussed here. In neutron deficient, Se, Kr, Sr and Zr, prolate and oblate configurations compete for the ground state giving rise to a unique prolate-oblate shape coexistence scenario with very large mixing of the wave functions by opposition to the previous cases. The nearly identical microscopic configuration of both proton and neutron arise from the fact that naturally proton and neutron occupy same orbits. By opposition to the $A \sim 100$ and 68 Ni regions where a proton-neutron interaction drives the shape competition, the $N \sim Z$ region could be explained by mean field effects. The weak occupancy of the $g_{9/2}$ orbit gradually increases along the N=Z line and can be correlated to the level of triaxility of the ground state. My contribution in this shape coexistence scenario is described in chapter IV.

Partie I

Avant-Propos

Chapter 1

Avant-Propos

One of the key parameter in the description of the nucleus is its mass distribution. Since the first observations of rotational bands similar to molecules in nuclei, it is well established that they are breaking the spherical symmetry and get deformed. Almost all nuclei are far from the spherical shape. This deviation is called nuclear deformation. The most simplest nucleus, ²H, has a non-zero quadrupole moment which indicates that on average a preference for a non-spherical shape for the nuclear interaction. The question of the description of the nuclear deformation, both theoretically and experimentally, remains a challenge in nuclear physics. On the theoretical side, one could summarize the open question to which degrees of freedom are relevant for the description of the nuclear shape. This lead to the development of non-spherical mean-field model at different orders or group-symmetry based approaches. The dominant symmetry is the axial symmetry with the so-called prolate and oblate deformation. Smaller corrections on the potential energy can be obtained by including the triaxial or the octupole degree of freedom.

Proton and neutrons create electric and magnetic fields in their orbits inside the nucleus. The V potential created by the charge distribution can be written in multi-pole moments such as :

$$V(R) = \underbrace{\frac{1}{R} \int \rho(r) dr}_{Electric\ monopole} + \underbrace{\frac{1}{R^2} \int z\rho(r) dr}_{Dipolar\ term} + \underbrace{\frac{1}{R^3} \int (3z^2 - r^2)\rho(r) dr}_{Quadrupolar\ term} + \dots$$
(1.1)

The nuclear surface R can be written using the spherical shape corrected by the normalized spherical harmonic such as :

$$R(\theta,\phi) = R_0(1 + \sum_{\lambda=0}^{\lambda} \sum_{\mu=-\lambda}^{\lambda} \alpha_{\lambda\mu} Y_{\lambda\mu}(\theta,\phi))$$
(1.2)

The λ, μ indexes defined the degree of symmetry of the nuclear surface. When limited to the triaxial degree of freedom, the three non null terms are $\alpha_{2,0}$, $\alpha_{2,2}$ and $\alpha_{2,-2}$. The later can be written as a function of two deformations parameters β and γ defined as :

$$\alpha_{20} = \beta cos\gamma$$

$$\alpha_{22} = \alpha_{2-2} = \frac{1}{\sqrt{2}}\beta sin\gamma$$
(1.3)

 β represents the axial elongation and γ is the asymmetry also called the triaxial degree of freedom. Using the ellipsoidal description of the nuclear volume, β is proportional to the difference between the long and short axis such as $\beta = \frac{4}{3}\sqrt{\frac{\pi}{5}} \left(\frac{c-a}{R}\right)$.

The comparison of measured and calculated nuclear deformation and its evolution with mass, charge and angular momentum is a cornerstone of the nuclear researches. On the experimental side, there are many methods to estimate the nuclear deformation. However measuring directly β or γ is not possible. From the observation of excited states, one can deduce from geometrical models an axial deformation parameter β using for instance the excitation energy of the first 2^+ state. In collective models, the reduced transition probabilities measures the wave functions overlap between different excited states under a given electromagnetic operator. For instance the B(E2, $2^+_1 \rightarrow 0^+_1$) gives the wave function overlap between the 2^+_1 and 0^+_1 states using the electric quadrupole operator. For all multi-polarities, the reduced transition probability is proportional to the matrix element between states and can be written such as :

$$B(E\lambda, I_i \to I_f) = \frac{|\langle I_f || M(E\lambda) || I_i \rangle|^2}{2I_i + 1}$$
(1.4)

In deformed nuclei, the B(E2)'s are large, underlining that excited states have the same structure but differ by their angular momentum. Similarly, under the assumption of geometrical models, a β parameter can be deduced from the B(E2, $2_1^+ \rightarrow 0_1^+)$.

The most accurate quantity to determine the nuclear deformation is the electric spectroscopic quadrupole moment. Its amplitude determines the overall deformation and its sign the prolate or oblate character. Similarly to the reduced transition probability, the spectroscopic quadrupole moment is proportional to the mean value of the E2 operator for a given state such as, in the laboratory frame :

$$eQ_s = \sqrt{\frac{16\pi}{5}} (2I+1)^{-1/2} \langle II20 \mid II \rangle \langle I \mid \mid M(E2) \mid \mid I \rangle$$
(1.5)

This quantity can be translated in the nucleus reference frame (intrinsic quadrupole moment) by :

$$eQ_0 = \sqrt{\frac{16\pi}{5}} \frac{1}{\sqrt{2I_i + 1}} \frac{\langle I_f \| M(E2) \| I_i \rangle}{\langle I_i K20 | I_f 0 \rangle}$$
(1.6)

With $I_f \neq I_i$, one deduces a transitional quadrupole moment which is related to the reduced transition probability by :

$$B(E2, I_i \to I_f) = \frac{5}{16\pi} (eQ_0)^2 \langle I_i K20 | I_f 0 \rangle^2$$
(1.7)

With $I_f = I_i$, the static quadrupole moment (Q_0) can be deduced and can be translated in β such as :

$$Q_0 = \frac{3}{\sqrt{5\pi}} Z R^2 \beta \left(1 + \frac{1}{8} \sqrt{\frac{5}{\pi}} \beta \right) \tag{1.8}$$

A direct translation from the intrinsic to the laboratory frame is given by :

$$Q_0 = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}Q_s$$

In excited states, the reduced transitional probability can be deduced from the measurement of their lifetime such as :

$$\frac{1}{\tau} = 12.26B(E2,\downarrow)E_{\gamma}^5(1+\alpha)$$

 τ is in [ps], the B(E2) in $[e^2 f m^4]$ and E_{γ} in [MeV]. The α coefficient is the total conversion coefficient. In this work, lifetimes ranging from 1 to 100 ps are measured using the plunger technique [45].

Experimentally, the ground state spectroscopic quadrupole moment can be measured at zero angular momentum in odd-mass nuclei using laser spectroscopy techniques [46]. For excited states, the spectroscopic quadrupole moment can be measured using the Coulomb excitation technique below the Coulomb barrier. At first order, the cross section is proportional to the Rutherford scattering cross section and the probability to populate the final state from the ground state under the electromagnetic field of the nucleus. The cross section can be written such as :

$$\underbrace{\frac{d\sigma}{d\Omega_{i\to f}}}_{ulomb\ cross\ section} = \underbrace{\frac{d\sigma}{d\Omega}}_{Rutherford\ cross\ section} \underbrace{\frac{\sum_{M_iM_f} |b_{if}^{Tot}|^2}{2I_i + 1}}_{P_{i\to f}\ probability}$$
(1.9)

The b_{if}^{Tot} amplitude has a first order (1) and a second order (2) terms :

Co

$$b_{if}^{(1)} = \frac{4\pi Z_1 e}{i\hbar} \sum_{\lambda\mu} \frac{1}{2\lambda + 1} \langle i \mid M(E\lambda, \mu) \mid f \rangle S_{E\lambda,\mu}$$

$$b_{I_f M_f I_i M_i}^{(2)} = \left(\frac{1}{i\hbar}\right)^2 \sum_n \int_{-\infty}^{+\infty} \langle I_f M_f \mid V(t) \mid I_n M_n \rangle e^{i\omega_{fn}t} dt$$
$$\times \int_{-\infty}^t \langle I_n M_n \mid V(t') \mid I_i M_i \rangle e^{i\omega_{ni}t'} dt'$$

with $S_{E\lambda,\mu}$ the Coulomb integrals. The total probabilities is :

$$P_{i \to f} \propto \mid b_{if}^{Tot} \mid^2 \propto \mid b_{if}^{(2)} \mid^2 + \mid b_{if}^{(1)} \mid^2 + 2 \mid b_{if}^{(1)} b_{if}^{(2)} \mid$$

The first order, (1), is proportional to the transitional E2 matrix element only while the second order, (2), is proportional to the diagonal E2 matrix element, i.e. the spectroscopic quadrupole moment via the interference term $|b_{if}^{(1)}b_{if}^{(2)}|$ [38]. The measurement of the Coulomb excitation cross section allows therefore to deduce both reduced transition probabilities and spectroscopic quadrupole moment.

Shape coexistence is a particular phenomenon in the study of the nuclear deformation. In that case, a significantly different microscopical configuration than the ground state can give rise to a different shape, corresponding to a second minimum in the potential energy, possibly degenerated with the ground state. The study of this phenomenon remains a challenge for both experimental and theoretical works. The modern challenges are the determination of the shape parameters of the coexisting structures, the coupling between them and how well they can be described by the theories. In the calculations, the main motivation is to understand the origin of the shape coexistence. At the mean-field level, the question is to identify the relevant symmetries. In a more microscopical approach, it is essential to identify the relevant nucleon excitations and coupling between them. Theoretical calculations reached in the last decade a very high level of description of the experimental quantities, in particular based on the Monte Carlo Shell Model approach. The counter part is that the description in proton pair excitation above major shell gap coupled to neutron occupying intruder orbits is not experimentally verified. A long term prospective is therefore the possible direct constrain on this description.

In this manuscript, I have focussed my studies on the shape coexistence phenomena involving the nucleon sub-shell closure 40. The experimental work uses the three experimental techniques described previously: high resolution prompt γ -ray spectroscopy of exotic nuclei to extract the low spin levels scheme, excited states lifetime measurements using the plunger technique and safe Coulomb excitation of radioactive beams. This manuscript is not an introduction to the field. Introductions to the subject, to the experimental techniques and their analysis are available in my PhD manuscript [47].

On the contrary, the aim of this work is to summarize very briefly my experimental achievements and put them in the perspectives of more recent experimental works and theoretical calculations. The goal is to identify if the collection of experimental data, on each mass region in which I produced a result, have consistencies and if not, trying to identify them. In chapter 3, controversial experimental results are discussed in great details in the light of the most recent experimental and theoretical works. In all chapters, my personal experimental work are compared and placed in larger systematics including in-beam, β -decay and ground state properties experimental results. The counter part of this choice is that general introductions on each mass region are very brief. Part of the work in chapter 3 are the results of the PhD thesis of A. Dijon [48] and I. Celikovic [49] at GANIL.

In spite my significant involvement in the experimental activities at GANIL through the EX-OGAM and AGATA projects [50, 51], this manuscript will not discussed this aspect of my careerer.

Partie II

Shape coexistence at A=100, Z=40

Chapter 2

Shape coexistence at A = 100, Z=40

2.1 Introduction

Neutron-rich nuclei with A~ 100 are amongst the best examples of interplays between microscopic and macroscopic effects in the nuclear matter. A rapid onset of quadrupole deformation is known to occur at N=60 in the neutron-rich Zr and Sr isotopes, making this region an active area for both experimental and theoretical studies. Already in the 60's, S.A.E. Johansson investigated the properties of light fission fragments of 252 Cf and observed an island of large and constant deformation around A=110 [52]. Since , a large set of experimental data becomes available in this mass region from ground state to high spin excited states.

A compilation of the mass and the charge radii $(\delta \langle r^2 \rangle)$ measurements was recently published in [46]. In fission fragments with A~100, the two neutron separation energy shows that the binding energy of Rb, Sr, Y and Zr isotopes rapidly increases at N=60 [53]. At the same neutron number, the systematics of the $\delta \langle r^2 \rangle$ shows an abrupt increase of the nucleus radii. Therefore, the onset of stability has been interpreted as a consequence of a dramatic increase of the ground-state deformation. This onset of deformation is unique in the nuclear chart. Indeed, it occurs at exactly N=60 for several elements (Rb, Sr, Y and Zr) ; it is sudden (from spherical N=58 isotones to highly deformed N=60 isotones) ; it is not observed for Mo and Kr isotopes. This low-Z border in the Kr isotopes of this phenomenon has been recently established by means of mass measurements, where no deviation from the standard trend toward the drip line was observed at N=60 [54]. This localized effect points to the interaction between specific proton and neutron orbitals.

The systematics of the excitation energy for the first 2⁺ states in the Sr and Zr isotopic chains show a sudden drop at N=60 (Fig.2.1) and by applying a simple geometrical model, one can relate it to a change of deformation from $\beta_2 = 0.1$ to $\beta_2 = 0.4$. On the other hand, in Kr isotopes, the energy of the first 2⁺ state decreases smoothly between ⁹⁴Kr₅₈ and ¹⁰⁰Kr₆₈ indicating instead a smooth increase of the deformation [55, 56]. At N=60, the evolution of the E(4⁺)/E(2⁺) ratio



Figure 2.1: Systematics of experimental excitation energy of the 2_1^+ state as a function of the neutron number.

between Zr/Sr and Kr shows an abrupt change of trend consistent with a change in microscopical evolution [57].

The uniqueness of this shape change is illustrated in figure 2.2. The systematics of the excitation energy of the 2_1^+ states in neutron rich Sr and Sm isotopes is presented as a function of the neutron number added to the last neutron shell closure, N=50 and N=82 respectively. A maximum of excitation energy is observed at the shell closure and a minimum when 10 neutrons are added. In between, the transition from the spherical shape associated to the shell closure to the highly deformed 2_1^+ is gradual in Sm isotopes where it is sudden in Sr isotopes. Due to the sudden onset of deformation, this shape transition at N=60 and Z~40 has recently been identified as a *Quantum Phase Transition* with N as the order parameter [54, 58].

Finally, low-lying 0^+ states, indicating possible shape coexistence, have been identified in the Zr and Sr chains and, similar to the 2^+_1 state, a drop of the 0^+_2 energy is observed at N=60 (Fig.2.3). A shape coexistence scenario was therefore proposed where the 0^+_2 states for N<60 correspond to a deformed configuration, which then become the ground state at N=60, while the spherical configuration of the ground state for N<60 becomes non-yrast.

The structure of neutron-rich Sr isotopes beyond the first 2^+ state has been studied extensively in the past. In ⁹⁶Sr, the ground-state band was shown to have a vibrational-like character, and the small $B(E2; 2_1^+ \rightarrow 0_1^+)$ value extracted from the lifetime of 7(4) ps [59] is consistent with a nearly spherical ground state. Two low-lying 0^+ states at 1229 and 1465 keV were established by G. Jung *et al.* [60] and interpreted as candidates for a deformed band head, supporting the shape



Figure 2.2: Systematics of experimental excitation energy of the 2_1^+ states in neutron rich Sr and Sm isotopes as a function of the neutrons added to the shell closure.

coexistence scenario. An extremely strong electric monopole transition of $\rho^2(E0) = 0.185(50)$ was observed between these two states [23, 24], indicating the presence of a sizeable deformation and strong mixing between the configurations. In ⁹⁸Sr, the ground-state band has a rotational character, and the large B(E2) values between the excited states, deduced from lifetime measurements [61, 62, 63, 64, 59, 65, 66], are consistent with a deformed character of the ground state. A low-lying 0_2^+ state at 215.3 keV was established by F. Schussler *et al.* [25] and interpreted as the band head of a presumably spherical structure. A strong electric monopole transition of $\rho^2(E0) = 0.053(5)$ was measured between the 0_2^+ and the 0_1^+ states, again supporting the shape coexistence scenario [25, 67].

While these observables yield informations on the configuration of the ground state band and strongly support the shape coexistence scenario, the exact properties of co-existing non-yrast structures and the degree of their mixing with the ground-state configuration can only be inferred from both transition rates and spectroscopic quadrupole moments.

2.2 Coulomb excitation experiment results

The spectroscopic quadrupole moments and reduced transition probabilities in ^{96,98}Sr have been measured by low-energy Coulomb excitation of post-accelerated radioactive ion beams at REX-ISOLDE, which provided firm evidence for shape coexistence and configuration inversion in the



Figure 2.3: Systematics of experimental excitation energy of the $0^+_{2,3}$ states as a function of the neutron number.

neutron-rich Sr isotopes [10, 11, 68]. The corresponding level schemes are presented in figure 2.4. To summarize the output of this rich set of experimental data :

- A Molecular extraction of a 96 Sr¹⁹F⁺ beam was developed further broken in the REX-EBIS to deliver a pure secondary 96 Sr beam
- In REX-Trap/REX-EBIS $\beta\text{-decay}$ of $^{98}\mathrm{Rb^+}$ was developed to produce a secondary post-accelerated $^{98}\mathrm{Sr}$ beam
- 2(9) E2 transitional matrix elements have been measured in 96 Sr (98 Sr), respectively.
- 1(5) E2 diagonal matrix elements have been measured in ⁹⁶Sr (⁹⁸Sr), respectively for the first time.
- The B(E2) transition strengths are in good agreement with the results from the most recent lifetime measurements.
- Using the precise lifetimes measurements as additional input in the analysis of the Coulomb excitation data enhanced the sensitivity to the reorientation effect and allowed extracting spectroscopic quadrupole moments for several excited states for the first time.
- In ⁹⁶Sr, the diagonal matrix element of the 2⁺₁ state is found small and compatible with 0 in the error bars, and compatible with a nearly spherical state.



Figure 2.4: Experimental level schemes for ⁹⁶Sr and ⁹⁸Sr in [10, 11, 68].

- In ⁹⁸Sr, the diagonal matrix elements of the 2⁺₁, 4⁺₁, 6⁺₁ and 8⁺₁ states are large and negative underlying the large prolate deformation of the ground state band. On the contrary, the 2⁺₂ diagonal matrix is found compatible with 0 indicating a spherical state similar to the 2⁺₁ in ⁹⁶Sr. This is the first ever proof of shape coexistence in Sr isotopes and supports the shape inversion between ⁹⁶Sr and ⁹⁸Sr.
- The B(E2; $2_1^+ \rightarrow 0_1^+$) in ⁹⁶Sr is close to the B(E2; $2_2^+ \rightarrow 0_2^+$) in ⁹⁸Sr supporting their similar nature. In addition, the B(E2; $2_2^+ \rightarrow 0_1^+$) in ⁹⁸Sr are found an order of magnitude smaller than the B(E2; $2_2^+ \rightarrow 0_2^+$), suggesting a very weak mixing between the wave function of the coexisting states.
- The systematics of B(E2)'s along the ground state band is found similar in Sr and Zr as good rotational bands.
- They differ from the Mo isotopes which present a change from axial to triaxial rotational band.
- The transitional quadrupole moments calculated from the $B(E2,2_1^+ \rightarrow 0_1^+)$ of 96,94 Kr, 96 Sr, 98 Zr and 100 Mo are found very similar supporting the fact that they lie out of the island of deformation.
- The spectroscopic quadrupole moments of the 2⁺₁ state deduced from the diagonal E2 matrix elements in ^{96,94}Kr, ^{96,98}Sr and ¹⁰⁰Mo are found very similar and low, suggesting a certain level of triaxility in the deformed ⁹⁸Sr, similarly to the deformed ¹⁰⁰Mo.

- The matrix elements were interpreted in a phenomenological two-band mixing model for 98 Sr. The mixing angle for the 0⁺ (2⁺) states yield 0.87(1) (0.99(1)) respectively underlying the very weak mixing of the wave function in spite of their proximity in excitation energy.
- The purity of the wave functions increases rapidly with spin, explaining the increase in the spectroscopic quadrupole moments between the different 2⁺ and 0⁺ states.
- The quadrupole sum rule formalism was applied to derive shape parameters for the 0⁺ states.
- The first rotational invariant, Q^2 (similar to $|\beta_2|$), is found small and very similar for the 0_1^+ (0_2^+) states in ⁹⁶Sr (⁹⁸Sr) respectively, confirming their weakly deformed character. On the contrary, the value is found large for the 0_1^+ in ⁹⁸Sr supporting the large deformation of the ground state band.
- The second rotational invariant, $cos3\delta$ (similar to γ) can only be calculated in ⁹⁸Sr with the available E2 matrix elements. It shows that both 0⁺ states are triaxial with a value of $\gamma \sim 20^{\circ}$. The similarity of the electromagnetic moments between the 0⁺₁ in ⁹⁶Sr and 0⁺₂ states in ⁹⁸Sr suggests that the 0⁺₁ in ⁹⁶Sr should also present a certain level of triaxility.
- The sharp transition is associated to a very weak mixing between competing configuration, involving triaxility at very low spin, in contrast to the N~Z cases in Kr and neutron deficient Hg for instance.
- The Davydov-Filippov model was applied on the $\langle 2_1^+ | E2 | 2_1^+ \rangle / \langle 2_1^+ | E2 | 0_1^+ \rangle$ matrix elements ratio as a function of the γ triaxial parameter. The experimental ratio is compatible with $\gamma \sim 27^\circ$ close to the 20° extracted from the quadrupole sum rule formalism.
- The transition strengths and quadrupole moments were compared to theoretical calculations going beyond the static mean-field approach using the Gogny D1S interaction which well reproduce the shape coexistence in N=Z nuclei (see Chapter 4).

As mentioned, we applied a simple two-states mixing model to the measured reduced matrix elements in ⁹⁸Sr. In this model, the observed physical states $|I_1^+\rangle$ and $|I_2^+\rangle$ may be expressed as linear combinations of pure prolate and spherical configurations, $|I_n^+\rangle$ and $|I_s^+\rangle$, respectively:

$$|I_1^+\rangle = +\cos\theta_I \times |I_p^+\rangle + \sin\theta_I \times |I_s^+\rangle$$
$$|I_2^+\rangle = -\sin\theta_I \times |I_s^+\rangle + \cos\theta_I \times |I_s^+\rangle$$

Experimental data, in particular E2 matrix elements, can be used to calculate the mixing amplitudes, $\cos^2\theta_I$, between the two pure (unperturbed) configurations. Following the method described [9], and using the complete set of E2 matrix elements in ⁹⁸Sr we have extracted, small mixing angles for the 0⁺ and 2⁺ wave functions [10, 68, 11]. An evaluation of the mixing angles for the 0⁺ and 2⁺ states requires a large set of E2 matrix elements which are not available for many nuclei in this mass region. Assuming a negligible mixing between 2⁺ states, one can extract an approximate mixing angle for the 0⁺ states via

$$\tan \theta_0 = \frac{\langle 0_2^+ | E2 | 2_1^+ \rangle}{\langle 0_1^+ | E2 | 2_1^+ \rangle}.$$

Elements	N = 58	N = 60
Pd	0.93	0.86
Ru	0.86	0.92
Mo	0.63	0.84
Zr	not measured	0.84
Sr	0.84	0.88

Table 2.1: Values of $\cos^2 \theta_0$ for the mixing angles between the 0⁺ states in N=58,60 nuclei extracted using the approximation described in the text.

The $\cos^2 \theta_0$ values obtained using the above expression for N=58,60 Sr, Zr, Mo, Ru and Pd nuclei are presented in Tab 2.1. For the $0_2^+ \rightarrow 2_1^+$ transition in ⁹⁸Zr, only an upper limit of the B(E2) is known. The result for ⁹⁸Sr is similar to the values determined for neighbouring nuclei. A rather weak mixing between coexisting structures is observed for nuclei in this mass region, both inside and outside the region of deformation. ¹⁰⁰Mo presents a certain level of mixing which could be related to a certain level of triaxiality as proposed from the systematics in B(E2) along the rotational band [69].

2.3 Q_s systematics

The diagonal E2 matrix elements extracted from the Coulomb excitation analysis can be translated into spectroscopic quadrupole moments, defined in the laboratory frame, using :

$$Q_s = \sqrt{\frac{16\pi}{5}} \frac{1}{\sqrt{2I+1}} (I, I, 2, 0|I, I) \langle I \| E2 \| I \rangle$$

It can be compared to the spectroscopic quadrupole moments of the ground states obtained from laser spectroscopy for odd-mass neighbouring nuclei [70]. In order to eliminate spin, mass and charge dependences, we further convert the Q_s values to intrinsic quadrupole moment and to quadrupole deformation parameters β_2 , assuming an axial symmetry:

$$\mathcal{Q}_s = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}\mathcal{Q}_0 = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)}\frac{3}{\sqrt{5\pi}}ZeR^2\beta_2$$

where R is defined as R= $1.25 \times A^{1/3}$ [fm].

The comparison for all known Q_s is shown in Fig.2.5 as a function of the neutron number for the odd-neutron Kr, Sr, Zr, Mo and Ru nuclei, and for the odd-proton Rb, Y, Nb and Tc isotopes [70]. The spherical neutron Nilsson orbitals are also indicated. The nuclei with N<60 have a low deformation with $\beta_2 \leq 0.2$. Our measurement of the spectroscopic quadrupole moment for the 2_1^+ state in ⁹⁶Sr fits well with the systematics. For N \geq 60, the deformations of the ground states of odd-mass isotopes, and in the ground state band of ⁹⁸Sr, are consistently large, whereas the β_2 value for the 2_2^+ state in ⁹⁸Sr is as low as those observed for N<60. Unfortunately, quadrupole moments for ^{90,92}Rb, ^{91–93,95–98}Y, ^{94–98,100}Nb and ^{99–101}Mo have not been measured yet. The spectroscopic quadrupole moments in ^{88,90,92}Kr, ^{90,92,94}Sr and ^{92–94,96–100}Zr are unknown. In the



Figure 2.5: Quadrupole deformation parameter β_2 as a function of the neutron number in the Kr – Ru isotopic chains. The parameter is calculated assuming an axial symmetry. The values calculated from diagonal E2 matrix elements measured in the present experiment for $J\neq 0$ states in 96,98 Sr are compared to those obtained from laser spectroscopy for the ground states of odd-mass nuclei. States belonging to the ground state band (2^+_2) are labelled in red (blue), respectively. Spherical Nilsson orbits are mentioned for neutron.

stable ^{94,96}Mo and ^{98,100,102}Ru nuclei the quadrupole moments of the 2_1^+ states were measured using the reorientation effect with light-ion beams and yielded two alternative solutions in the Coulomb excitation analysis leading to different conclusions on the deformation. The data are therefore not displayed in the figures. This limitation could be overcome using multi-step Coulomb excitation with high-Z collision partners. The systematics shown in Fig. 2.5 is further extended by including the spectroscopic quadrupole moments of the 2_1^+ states in ^{94,96}Kr [55, 71], ^{98,100}Mo [72, 69] and ¹⁰⁴Ru [73], extracted from low-energy Coulomb excitation experiments. They follow consistently the trend observed in laser spectroscopy. Between N=50 and 58, when the $d_{5/2}$ and $s_{1/2}$ orbits are filled, the deformation is uniformly low ($\langle \beta_2 \rangle \sim 0.1$) for all elements from Kr to Tc. From N=60, all deformation parameters are uniformly high ($\langle \beta_2 \rangle \sim 0.5$) for all elements from Rb to Tc. Due to the large error bars, no conclusion can be drawn for ⁹⁶Kr. Unfortunately, ¹⁰²Mo is not yet measured as possible high even-Z border of the island of deformation. The onset of deformation when adding 2 neutrons to N=58 (ie. the $s_{1/2}$ orbit) to N=60 (ie filling the $g_{7/2}$) is consistently observed in both methods.

This systematics highlights a particular behaviour at N=59. As the onset of deformation is abrupt for all elements from Z=37 to 41 when adding a pair of neutrons, at N=59 a gradual effect

as function of the proton number emerges. The ${}^{96}_{37}$ Rb₅₉ deformation is estimated at $\beta_2 \sim 0.17$, slightly higher than the N \leq 58 systematics and well below the N \geq 60 systematics. For ${}^{103}_{44}$ Ru₅₉, the deformation is evaluated at $\beta_2 \sim 0.4$, slightly lower than the N \geq 60 systematics. Therefore, a more gradual shape change is suggested when only one neutron is added to the $g_{7/2}$ and when increasing the proton number in the pf shells. The shape change seems highly correlated to the occupancy of both proton and neutron in the mentioned orbits. Unfortunately, this gradual change between Sr and Tc cannot be quantified as no laser spectroscopy of this N=59 isotone (1/2 ground state spin) was performed so far.



Figure 2.6: Experimental quadrupole deformation of the ground state for odd-neutron Kr, Sr, Zr, Mo and Ru nuclei, and for the odd-proton Rb, Y, Nb and Tc isotopes, and for the 2_1^+ state in even-even nuclei, as a function of the proton and neutron number. The sizes of the squares are proportional to β_2 under the assumption of axial symmetry. The Pseudo (*italic*) and Quasi (**red**) SU(3) orbitals are shown (see text).

In figure 2.6, the deformation is represented in the (N,Z) plan. The sizes of the squares in the plot are proportional to β_2 . These systematics suffers from several missing experimental points. Large quadrupole deformations are observed for the ground states of odd-mass nuclei and for the first 2⁺ states in even-even nuclei for elements between Z=37 and Z=44 at N=60 and beyond. With this two dimensional systematics, the island of deformation appears clearly and shows its boundaries. It appears that the Kr isotopes lie outside the island of deformation since the spec-

troscopic quadrupole moment in 96 Kr₆₀ is low with respect to those in 97 Rb₆₀ and 98 Sr₆₀. This may indicate that for Z=36, the two neutrons beyond N=58 occupying the $g_{7/2}$ are insufficient to induce a large quadrupole deformation. The completion of the systematics, in particular for the Kr isotopic chain and N=59 isotones, are vital informations to further constrain the microscopic interpretation of the onset of deformation at N=60.



Figure 2.7: Algebraic deformation amplitude (β_2) in the (N,Z) plan from measured spectroscopic quadrupole moments. *Top*: for ground state or 2^+_1 states. *Bottom*: for isomers or $2^+_{2,3}$ states

In figure 2.7, the algebraic deformation is deduced (β_2). The blue (green) squares indicate prolate (oblate) deformations respectively. The first matrix shows the deformation for the ground states in odd-nucleon isotopes and the 2_1^+ state for even-even isotopes. The second matrix displays the long lived isomeric state deformation in odd-proton or odd-neutron isotopes extracted from the laser spectroscopy. In the case of the even-even nuclei, the 2_2^+ or 2_3^+ state built on top of the 0_2^+ state with the largest B(E2) is shown. The first systematics is similar to figure 2.6 and highlights the onset of deformation of prolate character. The second systematics reveals the lack of experimental data in spite of the large set of known long lived isomers. Unfortunately, only spectroscopic quadrupole moment in Y isotopes have been measured. In even-even nuclei, 98,100 Mo and 98 Sr have measured spectroscopic quadrupole moments. Consistently, for isotopes with N \leq 58, non-yrast 2⁺ states and isomers have very large deformation of prolate character in Mo and oblate character in Y isotopes coexisting with weakly deformed ground state. Beyond N=58, only one deformation in excited configuration is measured in ⁹⁸Sr having a weak oblate deformation. This systematics shows again the consistency between the laser spectroscopy measurement and the Coulomb excitation data and the shape inversion at N=60. Below Z=40, the non-yrast configuration seems to be systematically of oblate character.

2.4 Overview with other experimental data



2.4.1 Bands head overview

Figure 2.8: Systematics of ground and isomeric states spin/parity and confirmed deformation in Zr, Y and Sr isotopes. Orange stars indicate that the deformation parameter was extracted from laser spectroscopy. The yellow stars highlight the deformation measurement by safe Coulomb excitation. The pink stars underline the conclusion from the neutron transfer experiment in ⁹⁶Sr. Shapes without star are speculative deformation assignment.

Spectroscopy of fission fragments with mass $A \sim 100$ produced in fission sources or induced fission of actinide has triggered a large experimental effort world-wide to probe the scenario of shape coexistence. Large set of γ -ray spectroscopy and electron spectroscopy at high spins are available in the literature. The onset of collectivity and deformation change in the ground state is now clearly established, even if not for all isotopic chains. On the contrary, the shape coexistence description is incomplete and systematics of excited and ground state deformation are very scarce. With the development of post-accelerated radioactive ion beams, experimental data with "direct" methods are available and improve our description of shape coexistence in this mass region. The spin and parity of the ground state and long-lived isomers give strong indication on the involved Nilsson orbits. Figure 2.8 shows the systematics of measured ground states and isomeric states spin/parity and highlight confirmed deformation in Zr, Y and Sr isotopes as a function of the neutron number. Isotones are presented in the same column and a colour code indicates the element. For clarity, figure 2.9 shows the orbitals involved in the present discussion.



Figure 2.9: Shells involved in the mass A=100 region in the spherical shell model. The position of the $2d_{3/2}$, presented with a dashed line is adopted from the calculations in [58]. The orbitals energy spacing is arbitrary. The microscopical configuration of the spherical ground state of 96 Sr is shown.

Even neutron number

For even neutron number, the systematics is shown for ytrium (Z=39). Using spherical Nilsson orbits, the single proton occupies the $2p_{1/2}$ orbit as proven by the confirmed $J^{\pi} = 1/2^{-}$ ground state assignment up to neutron number N=58. The onset of deformation is triggered by proton-particle hole excitation across Z=40. The confirmed $J^{\pi} = 5/2^{+}$ ground state for ^{99,101}Y involves either a single proton into $2d_{5/2}$ lying above the $g_{9/2}$ orbits or a strongly coupled deformed even-even core and a single proton into the $[422]5/2^+$ Nilsson orbital (prolate deformed $1g_{9/2}$). This second configuration is favoured from both the laser spectroscopy [74] and in-beam γ -rays spectroscopy [75, 76, 77] with a deformation $\beta \simeq 0.4$.

In spherical ytrium isotopes, very long lived isomers have been observed. From laser spectroscopy, a spin/assignment $J^{\pi} = 9/2^+$ was deduced together with a large spectroscopic quadrupole moment indicating a large *oblate* deformation [70]. This is compatible with a single proton excited into the [404]9/2⁺ Nilsson orbital (oblate deformed $1g_{9/2}$). In the deformed configuration, one can expect a large fragmentation of the neutron occupancy in the $1\nu g_{7/2}$ and $1\nu h_{11/2}$ orbits.

Beyond N=60, the ground states are highly deformed and no isomeric state has been reported so far.

Even proton number

For even proton number, the systematics is shown for strontium (Z=38) and zirconium (Z=40). In figure 2.8, both at N=57 and N=59, Sr have a confirmed $J^{\pi} = 1/2^+$ ground state. This spin assignment in the supposed spherical 97 Zr and 95 Sr (N=57) ground state and is compatible with a single neutron in the $3s_{1/2}$ spherical Nilsson orbit. This assumption is supported by the local maximum of the 2^+_1 excitation energy in 96,98 Zr in figure 2.1 corresponding to two neutrons filling the $3s_{1/2}$ spherical Nilsson orbit. With one neutron more in 97 Sr (N=59), this spin/parity assignment to $J^{\pi} = 1/2^+$ is surprising. At N=59, the $3s_{1/2}$ is full and the $\nu 1g_{7/2}$ starts to be filled with one neutron in the conventional spherical Nilsson orbit. In a spherical framework, the $3s_{1/2}$ orbit is the only available low energy J=1/2 level for N=59 nucleus. As mentioned earlier from the proposed Q_s systematics, the N=59 appears as a transitional region and could present a certain level of deformation. According to MCSM calculations [58], in Zr isotopes, the neutron $2d_{3/2}$ orbit is placed between the $3s_{1/2}$ and the $1g_{7/2}$ and could provide a $J^{\pi} = 1/2^+$ spin/parity when deformed (1/2[400]). Therefore, the possible weakly deformed ground state at N=59 must be based on a mixture of weakly deformed $s_{1/2}$, $d_{3/2}$ and $g_{7/2}$ neutron orbits which differ from the large deformation of the ground state beyond N=60.

Isomeric states have drawn a particular attention below N=60 as possible candiate for shape isomers [78, 79, 70]. Long-lived $9/2^{+m}$ states have been identified in several odd Sr and Zr isoropes but unfortunately no spectroscopic quadrupole moment were measured so far. However, rotational bands have been clearly established supporting the deformed character, and therefore shape coexistence, in these isotopes. It is proposed in [79] that the $9/2^{+m}$ isomeric state involves a neutron excitation from the deformed 9/2[404] (g_{9/2})orbit from the ⁷⁸Ni core into the deformed 3/2[541](h_{11/2}).

In the deformed Nilsson diagram for excited configurations, at N = 59, the odd neutron can be placed either on the 3/2[541] (h_{11/2}) orbital, making a 3/2⁻ rotational band, or the 3/2[411] (g_{7/2}) orbital, making a 3/2⁺ rotational band. The deformation is larger than at N = 58 but the 9/2[404](g_{9/2}) orbital still limits the deformation. The deformed 3/2⁺ and 3/2⁻ excited band heads have been observed experimentally (not shown in the figure, see [78]) and are based on the single neutron occupancy of these deformed orbits. In the deformed region, at N=61 and 63, laser spectroscopy have established the deformed (prolate) character of ¹⁰¹Zr and ¹⁰¹Sr ground states. Tentative spins and parities are $(3/2^+)$ and $(5/2^-)$ respectively. At N = 61 the situation is analogous to that at N = 59. Two additional deformed bands with the single neutron in the 3/2[411] (g_{7/2}) and 5/2[532] (h_{11/2}) orbitals are observed. It is worth mentioning that the $9/2^{+m}$ K-isomer rotational band (similar to N=59) still exists at N=61 in the deformed ¹⁰¹Zr. It means that it is not the configuration based on the $\nu 9/2[404]$ excitation which becomes the ground state at N=60. In other words, the deformed ground state beyond N=60 does not involve, from the experimental point of view, an excitation from the ⁷⁸Ni core. As described later in the section 2.5.2, Monte Carlo Shell Model Calculations lead to the same conclusion and do not involve any neutrons excitation from the ⁷⁸Ni core to reproduce the large deformation beyond N=60.

2.4.2 Transition probabilities overview

Numerous γ -rays experimental data set exist probing the onset of deformation, from level schemes to B(E2) values by lifetime measurement, which give more indirect proof of the *shape* and microscopical configurations compared to Coulomb excitation experiments. A list of recent relevant works in the last 5 years in which I have been involved is listed below in addition to the Coulomb excitation data presented before.

- $B(E2;2^+_1 \to 0^+_1)$ value in ⁹⁰Kr [80].
- Abrupt shape transition at neutron number N=60: B(E2) values in 94,96,98 Sr from fast γ timing, [81].
- Evolution of nuclear shapes in odd-mass yttrium and niobium isotopes from lifetime measurements following fission reactions, [77].
- Experimental study of the lifetime and phase transition in neutron-rich ^{98,100,102}Zr, [82].
- Evidence for coexisting shapes through lifetime measurements in ⁹⁸Zr, [8]

As a result, the B(E2;2⁺₁ \rightarrow 0⁺₁) systematics is now completed from N=50 to N=60, ie covering from the neutron shell closure to the shape transition, for Zr, Sr and Kr isotopes. The systematics is presented in figure 2.10 where the B(E2) in W.u. are normalized by the mass A for each isotopes. This representation first proposed by R. F. Casten and N. V. Zamfir [83] illustrates the amount of nucleons participating to the collectivity. If the ratio equals unity, it illustrates that all nucleons participate to the collective behaviour. In a nearly spherical nucleus, this ratio should be close to 0 whereas in highly collective nucleus it should approach 1. In the case of the Sr/Zr isotopes, the onset of collectivity follows the two extremes. Below N=60, the ratio is very close to 0. For ^{96,98}Zr, the local maximum of the 2⁺₁ excitation energy (see figure 2.1), reflecting the $\nu s_{1/2}$ occupancy, corresponds to a local minimum in figure 2.10. From N=60, the ratio saturates where all nucleons participate to the collective behaviour suggesting an almost *perfect* rigid rotor.



Figure 2.10: Systematics of the $B(E2;2_1^+ \rightarrow 0_1^+)$ in W.u. normalized by the mass A as a function of the neutron number for Kr, Sr and Zr isotopes.

2.4.3 Rotational band and configuration mixing

From the collective model of A. Bohr and B. R. Mottelson, in a rotational band, the spectroscopic quadrupole moment, ie the intrinsic moment of the 2_1^+ state, equals the transitional quadrupole moment that one can convert from the $B(E2;2_1^+ \rightarrow 0_1^+)$ as follow :

$$\mathcal{Q}_s(I,K) = \frac{3K^2 - I(I+1)}{(I+1)(2I+3)} \cdot \mathcal{Q}_0$$

where Q_0 is the intrinsic quadrupole moment of the *I* state (ie in the nucleus frame) and Q_s is the spectroscopic quadrupole moment (ie in the laboratory frame). *I* is the angular momentum with its projection *K*. In the rotational model

$$B(E2; (I_i, K) \to (I_f, K)) = (5/16\pi) \cdot \mathcal{Q}_0^2 \cdot (I_i K 20 | I_f K)^2$$

For an even-even rotational nucleus and for its first 2^+ , I = 2, K = 0, the rotor model gives :

$$\mathcal{Q}_s(2,0) = -(2/7) \cdot \mathcal{Q}_0$$

Simplifying by \mathcal{Q}_0 , the asymptote limit is

$$\frac{\mathcal{Q}_s(2,0)}{\sqrt{B(E2;(0,0)\to(2,0))}} = -0.906$$

In [84], a global survey of the 2_1^+ states collectivity is proposed using a quantity, R_{QB} , defined as :

$$R_{QB} = -\frac{\mathcal{Q}_s(2_1^+)}{\sqrt{B(E2;0_1^+ \to 2_1^+)}}$$

In rare-earth elements, known to be excellent rotor, the $E_x(4_1^+)/E_x(2_1^+)$ ratio saturates at 10/3 [83] with the R_{QB} ratio nearly equal to unity. The $E_x(4_1^+)/E_x(2_1^+)$ ratio equals 1.2 (3.0) in ⁹⁶Sr (⁹⁸Sr) respectively. The B(E2; $2_1^+ \rightarrow 0_1^+)/A$ ratio yields a value of 0.18 (0.98) in ⁹⁶Sr (⁹⁸Sr) respectively. This indicates that ⁹⁶Sr behaves as a spherical vibrator and ⁹⁸Sr as an almost perfect rotor. From our Coulomb excitation data, spectroscopic quadrupole moments for the 2_1^+ state in ^{96,98}Sr were measured at -22(30) efm^2 and -52(24) efm^2 respectively. The accurate value of the B(E2), leads to $R_{QB}({}^{96}Sr) = 0.46(70)$ and $R_{QB}({}^{98}Sr) = 0.45(28)$. The large error bars on the experimental Q_s do not allow drawing final conclusion for ⁹⁶Sr. However, within the experimental error bars, the value for ⁹⁸Sr is found far from what one expect for a good rotational nucleus.

As mentioned earlier [10, 68, 11], using the full set of E2 matrix elements, the two-level mixing model was applied in ⁹⁸Sr. A mixing angle of 0.87(1) was extracted for the 0_1^+ state wave function while the 2^+ states have almost no mixing. Unperturbated matrix elements can be extracted from which an *unperturbated* intrinsic quadrupole moment for the band can be deduced, later translated in the laboratory frame. The unperturbated matrix elements equals -1.45(2) eb (to be compared to the measured $-0.63 \begin{pmatrix} +0.32\\ -0.28 \end{pmatrix}$ eb for the 2_1^+ state). This leads to an *unperturbated* $\mathcal{Q}_s^{un} = -110(3)$ efm^2 . Using \mathcal{Q}_s^{un} in the R_{QB} equation leads to $R_{QB}^{un}({}^{98}Sr) = 0.96(2)$, is in the rotational limit. As a summary, the weak mixing of the 0^+ states wave functions does not influence significantly the rotational behaviour of the excitation energies and B(E2). However, the measured spectroscopic quadrupole moment for the 2^+_1 state shows a clear perturbation from its rotational behaviour. Using the two-levels mixing model, the unperturbated moment fits well the rotational behaviour. One must underline that \mathcal{Q}_s^{un} doesn't involve the measured $\mathcal{Q}_s(2_1^+)$ (see [9] for the details). An approximate experimental Q_s^{un} can be estimated from the average measured $\langle Q_s^{un} \rangle$ in the 4⁺, 6⁺ and 8⁺ states and equals to -1.6(3) eb [11]. In that case, $R_{QB}^{\langle Q_s \rangle}({}^{98}Sr) = 1.4(5)$, in agreement with the previous observations. As a results, the presented study for 98 Sr shows the influence of the mixing in the shape coexistence and how it can be taken into account in collective nuclei. However, the spectroscopic quadrupole moment of the 2^+_1 state is not discussed here and not used in the present R_{QB} calculations. Its value is found smaller than the Q_s^{un} calculated from mixing model and the average value at higher spins. The difference could be attributed to the mixing of the wave functions. However, from the mixing model, it is found negligible. Therefore, the low value is not attributed to the mixing but to a certain level of triaxility as deduced from the sum rules.

2.4.4 Particle-Core coupling scheme

In the deformed region, one can also underline that lifetime measurements in odd-proton and even-neutron [77] illustrate very well the particle+rotor model (so-called strong coupling between a deformed even-even core and a single particle) [85]. In ⁹⁹Y, the ground state spin/parity is $5/2^+$. In the particle+rotor model, the ground state can be seen as $5/2^+ = \pi [422]5/2^+ \otimes 0^+ (^{98}Sr)$. The proposed $9/2^+$ state at 284 keV in ⁹⁹Y can be part of the multiplet of the $\pi [422]5/2^+ \otimes 2^+ (^{98}Sr)$ configuration. The B(E2; $2^+ \rightarrow 0^+$) established at 0.259(8) e^2b^2 in ⁹⁸Sr [10] is compatible with the measured B(E2; $9/2^+ \rightarrow 5/2^+$) = 0.25(2) e^2b^2 in ⁹⁹Y by [77], supporting very well the particle+rotor model.

At N=58, in spherical ⁹⁶Sr, the 2⁺ state excitation energy is 815 keV with a B(E2; 2⁺ \rightarrow 0⁺) = 0.045(10) e^2b^2 . In the weak coupling scheme, ⁹⁷Y should present a multiplet of states $(1/2^-)\otimes 2^+({}^{96}Sr) = (5/2^-, 3/2^-)$ with a barycentre equal to the excitation energy of the 2⁺ state in ⁹⁶Sr and similar B(E2) to the ground state. A state at 953 keV with proposed spin/parity assignment $(5/2^-, 3/2^-)$ and a lifetime $T_{1/2} \leq 4$ ps is suggested. A more accurate measurement of this lifetime would confirm the spin/parity assignment of the ground state and the weak coupling scheme. In lighter isotopes at N=56, the 2⁺ state excitation energy in ⁹⁴Sr is 837 keV with a corresponding B(E2) = 0.020(8) e^2b^2 . In ⁹⁵Y, two states with confirmed spin/parity at 827 keV (5/2⁻) and 685 keV (3/2⁻) are the obvious candidates for the $(1/2^-)\otimes 2^+({}^{94}Sr)$ multiplet. Unfortunately, the corresponding B(E2) are not measured. If such measurements could confirm the weak coupling scheme, it would bring further indications that the Z=40 is a robust su-shell closure. Indeed such weak-coupling states have been established around large proton shell gap at Z=28, Z=50 and Z=82.

2.4.5 Experimental multi-messenger approach: the ⁹⁶Sr case



Figure 2.11: Multi-messenger studies of the shape coexistence in 96 Sr

Figure 2.11 shows the compilation of several experimental data set for 96 Sr. The low spectroscopic quadrupole moment of the 2^+_1 state and relatively large B(E2) indicate its vibrational nature around a weak deformation [10]. As already mentioned, two 0^+ excited states have been observed at 1229 and 1465 keV. E0 spectroscopy was performed and deduced one of the highest $\rho^2(E0)$ value between the 0^+_2 and 0^+_3 states, indicating a very large difference in deformation and/or large mixing of the wave functions [24]. The E0 branch to the ground state was never established. The 2_2^+ and 2_3^+ states are very close in energy to the 0_2^+ and 0_3^+ states respectively, suggesting a rotational band. The second 4^+ , 6^+ and 8^+ states develop a rotational band but it is very unclear to which band head it belongs. The lifetime of the second 6^+ and 8^+ states was measured and suggest a rather weak deformation [78]. The 95 Sr ground state is a $1/2^+$ state compatible with the spherical Nilsson orbit $\nu[s1/2]$ [86]. Unfortunately, its spectroscopic quadrupole moment was not measured. The neutron transfer reaction ${}^{95}Sr(d,p){}^{96}Sr$ was recently performed [13, 87], which favours spherical component of the populated states are favoured in 96 Sr. It turned out that the ground state is relatively weakly populated as the main spherical strength is found in the low lying 0^+ states, suggesting a rather weak deformation of the ground state (compatible with the spectroscopic quadrupole moment extracted from the Coulomb excitation) and a nearly spherical 0_2^+ state which however presents a large mixing with the deformed 0_3^+ state [13, 87]. From the E0 spectroscopy and the large $\rho^2(E0)(0_3^+ \to 0_2^+)$, the 0_3^+ becomes a natural candidate for the band head of the deformed configuration that becomes the ground state in ⁹⁸Sr. Such measurement was not performed from 97 Zr to 98 Zr having also two excited 0⁺ states. One could assume a similar scenario and draw a direct comparison between the $0^+_{1,2,3}$ states in 96 Sr and 98 Zr.

However, different conclusions were drawn in the N=58 ⁹⁸Zr by [8]. In this work, the B(E2) values in ⁹⁸Zr have been measured and compared to MCSM calculations. The calculations propose the coexistence of three different structures at low spin: a nearly spherical ground state (0_1^+) , a deformed excited 0_2^+ state, and a well-deformed (band-like) structure possibly based on the 0_3^+ state. The calculated proton and neutron occupancies are very similar in both 0_2^+ and 0_3^+ deformed states. Only small differences in the neutron occupancies make the change in deformation. The ⁹⁵Sr(d,p)⁹⁶Sr transfer reaction cross sections propose instead nearly spherical 0_1^+ and 0_2^+ states with a well-deformed 0_3^+ state. The angular distributions treated as a single-step process to the 0_2^+ is best fitted for a l = 0 angular momentum transfer, thus neutron populating the $\nu 2s_{1/2}$ orbital. According to the previous discussions, a deformed configuration would involve a l = 2 ($d_{3/2}$), or l = 4 ($g_{7/2}$) or l = 5 ($h_{11/2}$) angular momentum transfer. The microscopical configuration of the $0_{2,3}^+$ state at N=58 remains therefore puzzling.

2.5 Theoretical calculations for ^{96,98}Sr

2.5.1 Mean field approach

The sharp transition and magnitude of the deformation at $A\sim100$ remain a challenge for theories. Similarly to the experimental effort, numerous theoretical works have been published (more than 100 theoretical papers since the 70's) on the subject. All nuclear models have been used to reproduce this onset of deformation such as the HFB + the generator coordinate method (GCM), the macroscopic-microscopic method, the shell model, the Monte Carlo shell model, the interacting boson model (IBM) approximation, the VAMPIR model and covariant density functional (DF) theory (PC-PK1). The global comparison of all the models is out of the scope of the present discussion.
In the following, the state-of-the-art of mean-field calculations using the Gogny D1S interaction with GCM-GOA and Monte Carlo Shell Model Calculations will be briefly discussed.



Figure 2.12: Comparison between the experimental and calculated level scheme of ${}^{96}Sr$ using the Gogny D1S interaction. The excitation energies are mentioned in brackets. The arrows are proportional to the B(E2) indicated in e^2b^2 . The measured and calculated spectroscopic quadrupole moments are given in efm² next to each state.

We compared the set of E2 matrix elements with calculations using the Gogny D1S interaction with GCM-GOA. The comparison is presented in [10, 68, 11]. Bands assignment in calculated states is not obvious. We developed a graphical method detailed in [11] to construct theoretical bands. In figure 2.12, the calculated level scheme is compared to the experimental one for ⁹⁶Sr. As discussed in [10, 11], the Gogny calculations over estimate the collectivity at N=58 and under estimate the deformation at N=60. The calculated ground state band differs from the experimental ones in B(E2) and predict a very fast increase of the Q_s as a function of spin with a significant contribution of K=2 (30%) for the 2_1^+ state leading to this low Q_s . This is impossible to verify today. The calculations do not predict two 0⁺ excited states in contrast to the experimental situation. The (B) band presents large B(E2) associated to low Q_s with only 50% of K=0 underlying the triaxial character of the state. In the (A) band, large B(E2)'s are associated to large Q_s with a large contribution of K=0.

Figure 2.13 shows the comparison between the Gogny calculations and the VAMPIR approach [88]. The VAMPIR B(E2) are mentioned in red. The agreement is surprisingly good in spite of the very different approaches used in the calculations.



Figure 2.13: Comparison between the experimental and calculated level scheme of ${}^{96}Sr$ using the Gogny D1S interaction and the VAMPIR approach

In figure 2.14, the calculated level scheme is compared to the experimental one for 98 Sr using the Gogny interaction. In both cases, the ground state band has a large prolate deformation. It is worth mentioning that in spite of the differences in absolute values, the Gogny calculations predict a drop of the Q_s between the 4_1^+ and the 2_1^+ states as observed experimentally. A K=2 γ -band is predicted very close to the ground state band but not yet observed experimentally. The nearly spherical configuration is assigned as the (A) band based on the similarities in the B(E2) connecting to the ground state band.

A summary of the experimental $\rho^2(E0)$ transition with the calculated ones using Gogny D1S, Excited VAMPIR and 5DCH (PC-PK1) in table 2.2. It must be underlined that discrepancies are huge and prediction not always calculated for all transitions making the comparison difficult.

2.5.2 Shell model approach

In the Sr and Zr isotopic chains, the spherical-to-deformed transition takes place when going from 58 to 60 neutrons, thus when the $\nu g_{7/2}$ orbital is being filled. Shell model calculations were performed for the Zr isotopic chain in an extended model space [89, 5, 58, 8] and pointed to the $\pi - \nu$ interaction between the spin-orbit partners $\pi 0g_{9/2}$ and $\nu 0g_{7/2}$ as the main mechanism for the shape change: as the $\nu 0g_{7/2}$ orbital is being filled, the Z=40 sub-shell gap between the $\pi 0f_{5/2}$ and $\pi 0g_{9/2}$ effective single-particle energies (ESPE) is reduced, giving rise to multiple particle-hole excitations

Table 2.2: Monopole transition strengths $\rho^2(E0)$ between 0^+ states. Experimental results are compared with those calculated within the beyond-mean-field approach with a five-dimensional collective Hamiltonian and the Gogny D1S force (present study) and using the complex excited VAMPIR approach [88].

		experiment ^{96}Sr	5DCH (Gogny)	Excited VAMPIR
			$\rho^2(E0)~(\cdot~10^3)$	
0_{2}^{+}	0_{1}^{+}		106	66
0^{+}_{3}	0_{1}^{+}		22	
0^{+}_{3}	0_{2}^{+}	185(50)[24]	95	9
		experiment 98 Sr		5DCH (PC-PK1)
0_{2}^{+}	0_{1}^{+}	53(5)[67]	179	117
0^{+}_{3}	0_{1}^{+}		40	
0^+_3	0_{2}^{+}		75	



Figure 2.14: Comparison between the experimental and calculated level scheme of ${}^{98}Sr$ using the Gogny D1S interaction

across the gap. The most recent calculations [5, 58, 8] using MCSMC show a spectacular agreement with the experimental data in even-even nuclei and become the reference in the mass region. In the calculations [90, 8], the 0_2^+ and 0_3^+ below N=60 result from 2p-2h proton excitations (with a possible 4p-4h contribution) from the pf-shell into the $\pi 0g_{9/2}$ orbital. On the neutron side, the $\nu g_{9/2}$ remains fully filled, ie without excitation from the ⁷⁸Ni core. The deformed configurations in even-even excited 0^+ does not involve neutron excitation from the N=50 closed shell. This might explain why (9/2^{+m}) deformed states still remains beyond N=60 in odd-neutron Sr/Zr isotopes as excited configurations. In the deformed configuration, neutrons are well distributed from the d_{5/2} into the s_{1/2},d_{3/2} and more significantly into the g_{7/2} and h_{11/2} as deduced from the previous studies of odd neutron isotopes. A similar mechanism is known to be responsible for the rapid onset of deformation and shape coexistence in neutron-rich isotopes around N= 8, 20, 28 and 40, with different spin-orbit partners [91, 92, 1, 93, 94].

2.6 Conclusions

For the future, several systematics are needed :

• 98 Sr remains a unique case in term of experimental data. More are needed in particular in the N=58 and N=59 to understand the shape evolution along the yrast bands and excited deformed configuration.

- Coulomb excitation of ⁹⁶Sr, Zr and Kr isotopes are mandatory to determine the shape of the different configuration.
- Following the exploratory work on the ⁹⁵Sr(d,p) reaction, single and double nucleon transfer experiment are needed to probe both the proton and neutron microscopic configuration.
- The N=59 isotonic line deserves a particular interest in laser and direct nucleon transfer experiment.
- In the spherical isotopes, the weak coupling scheme was not yet studied in details.
- The differences in structure and deformation of the 0^+_2 and 0^+_3 below N=60 in even-even Sr and Zr isotopes must be clarified.
- Laser spectroscopy of long lived isomeric state from N=50 to N=60 must be pursued as well as for ground state beyond N=60.
- In the global survey of the spectroscopic quadrupole moment, the safe Coulomb excitation of ¹⁰²Mo will make the bridge between the island of large deformation and the triaxial shape region
- Regarding the very large amount of experimental data in the mass region, holistic comparison in theoretical work must be encouraged.

Partie III

Shape coexistence in the neutron rich Ni isotopes

Chapter 3

Shape coexistence at N=40 and Z=28

3.1 Introduction

In isotopes with Z < 28 and N = 40, a new region of deformation was identified and characterized in the recent years. When removing only two protons from 68 Ni to 66 Fe, first excited states and reduced transition probabilities have indicated an onset of collectivity. This mass region presents a difference with the two other cases presented in the manuscript. The Z=28 shell closure is a strong shell closure as revealed by the doubly magic character of 56 Ni [95] and 78 Ni [96, 17] by opposition to the Z=40 sub-shell closure. The corresponding large proton gap implies that, in the scheme of the π particle-hole excitation as standard mechanism leading to shape coexistence, it must involve a substantial gain in $\pi - \nu$ quadrupole correlation energy. Along the Ni isotopic chain, neutrons are added to the $\nu q_{9/2}$ orbital from ⁶⁹Ni to the doubly magic ⁷⁸Ni. The experimental level schemes for the yrast band in even-even nuclei show a 8^+ isomeric state arising from the alignment of a broken neutron pair as an example of seniority $\nu = 2$ scheme. The resulting multiplet should be pure neutron states, ie with very low E2 probabilities. In 70 Ni, the 8^+ and 6^+ states present such character as the enhanced B(E2) for the 2^+ state was interpreted as an indication that the Z=28 proton shell gap, near ⁶⁸Ni, is relatively weak leading to core polarization [97, 98, 99]. From the B(E2) measurements in Ni, it was proposed that the spin-orbit splitting $\pi f_{7/2} - f_{5/2}$ is reduced when filling the $\nu g_{9/2}$ orbital through the tensor term of the nuclear interaction favouring the development of collectivity and possibly shape coexistence. Already in the 80's, M. Girod et al [100] predicted a low lying deformed 0^{+m} state in ⁶⁸Ni as a first prediction of shape coexistence at N=40. Deformed states below N=40 would therefore involved the $\nu g_{9/2}$ as intruder orbit.

⁶⁸Ni is therefore cornerstone of the mass region. The proton shell gap is closed at Z=28 by the $f_{7/2}$ orbits and the next shells are the $p_{3/2}$, $f_{5/2}$ and $p_{1/2}$. On the neutron side, the N=40 gap is defined between the $p_{1/2}$ and the $g_{9/2}$, $d_{3/2}$ orbitals. Experimentally, at N=40, the mass measurements do not show an increase of binding energy [101]. However, ⁶⁸Ni mimics the characteristics of a doubly magic nucleus with a first 2⁺ state at 2 MeV excitation energy, a minimum of the B(E2,

 $0^+ \rightarrow 2^+$) and a first excited state being a 0^+ state. Following the scheme of shape coexistence driven by a π p-h excitation, deformed states would consist in promoting a proton from the $f_{7/2}$ to the $p_{3/2}$, $f_{5/2}$ or $p_{1/2}$ orbitals coupled to a high - j neutron intruder orbits (in the present case the $g_{9/2}$). The later should coexist with spherical configuration and even become the ground state when proton are continuously removed from the $\pi f_{7/2}$ orbit. In ⁶⁷Co, a β -decay isomer was discovered and interpreted as a $\pi(1p-2h)$ excitation across Z=28 and shape isomer [102]. Using the energy of the intruder $\pi(2p-1h)$ state in ⁶⁹Cu and, symmetrically, the $\pi(1p-2h)$ state in ⁶⁷Co, which both lie at N=40, D. Pauwels *et al.* have been able to predict a deformed $\pi(2p-2h)$ state in ⁶⁸Ni. The predicted energy from this analysis leads to a rather low value of 2.2 MeV excitation energy, which can be understood only with an important gain in binding energy from the $\pi - \nu$ residual interactions between the two proton holes and the active valence neutrons across N=40 [3]. The shape coexistence in ⁶⁸Ni has been investigated by the observation of low lying 0⁺ state as a candidate for the deformed proton configuration.

The systematics of the 2^+ excitation energy of Fe isotopes doesn't show any local maximum at N=40 but a continuous decrease from N=36 to N=58, as an indication of collectivity and therefore deformation. In comparison with N=28, this onset of collectivity occurs closer to the proton shell closure (see figure 3.1). As Ar and S isotopes show a local maximum at N=28, already 2 protons from ⁶⁸Ni, no increase of the excitation energy is observed. In Co isotopes, similarly to Cu and Zn in which coexist single particle, collective and core-coupled states [103, 104], are states that would be naturally expected in odd-A masses based on the coupling of the single proton hole in the $f_{7/2}$ with even-even cores states. The corresponding multiplet is composed of the $f_{7/2} \otimes 2^+ = (3/2^-, 9/2^-, 11/2^-)$ states.

The systematics of excitation energies and B(E2) values in the Fe, Co and Ni isotopes show that the $9/2_1^-$ (Co) and 2_1^+ (Ni) excitation energies as well as those of the $3/2_1^-$ (Co) and 2_1^+ (Fe) states behave similarly. It is therefore tempting to interpret the $9/2_1^-$ state as a proton hole coupled to 2_1^+ (Ni) [105] and the $3/2_1^-$ state as a proton particle coupled to the deformed 2_1^+ (Fe). The $11/2^$ states have been recently established and not shown in the figure. Weakly deformed excited states must have all the characteristics of a spherical core-coupled states (weak coupling scheme). The barycentre of the multiplet should equal the 2^+ energy of the even-even Ni core and the B(E2) to the ground state. Deformed states should follow the strong coupling scheme where the intruder neutron orbit $\nu g_{9/2}$ is coupled to the rotational band of the even-even Fe core. The deformed configuration in odd-Co should present a $(2^+ \otimes g_{9/2}) = 11/2^+$, $(4^+ \otimes g_{9/2}) = 13/2^+$ etc ... sequence similar to the 2^+ , 4^+ etc ... rotational band in the even-even Fe core. The low energy excited states $9/2^-$, $11/2^-$ and $3/2^-$ states have been identified in 63,65,67 Co. In the present study, the core-coupled scheme was tested by the systematic lifetime measurements of relevant excited states leading to reduced transition probabilities and compared to large scale shell model calculations.

3.2 Lifetime measurement and transition probabilities around N=40

The onset of collectivity in the mass region was probed by the measurement of the reduced transition probabilities (B(E2) and B(M1)) from lifetime measurements using the plunger technique. A series of experiments were conducted at the GANIL facility using the EXOGAM and AGATA



Figure 3.1: Top panel: Systematics of the experimental energies in the even-N Ca, Ar, S and Si isotopes. Bottom panel: Systematics of the experimental energies in the even-N Fe and Ni isotopes.

HPGe arrays, the VAMOS magnetic spectrometer and a plunger device to measure lifetime of excited states in neutron rich Fe and Co isotopes produced by multi-nucleon transfer between heavy ions. Lifetime in 62,64 Fe 2⁺ and 4⁺ states have shown that by comparison with shell model calculations, the $\nu g_{9/2}$ occupancy triggers the onset of collectivity at N=40 as the full amplitude is reproduced when the $\nu d_{5/2}$ is included [106, 107]. The core coupled assumption in the $(3/2^-, 9/2^-, 11/2^-)$ states in Co isotopes was tested by the measurement of their B(E2) to the

ground state (tentatively assigned to $J^{\pi} = (7/2^{-})$ from the single proton hole in the $f_{7/2}$ orbit) and compared to the E2 transition in the corresponding 2^{+} state in the even-even core. In the following a review of the measured values in Co is presented and enriched from measured performed in other facilities.

3.2.1 The $3/2_1^-$ state

The $B(\text{E2}; 3/2_1^- \to 7/2_1^-)$ value in ⁶³Co and ⁶⁵Co have been deduced from the lifetime measurement in [108, 109]. The measured value in ⁶³Co is found well below that of $B(\text{E2}; 2_1^+ \to 0_1^+)$ in Fe. This indicates that the N = 40 gap has not completely collapsed at Z = 27 or that the $3/2_1^$ state has in fact a single-particle character. As shown in [106, 107], the onset of collectivity in Fe is clearly established at N = 38. If the $3/2_1^-$ state results from the coupling of a proton to the 2_1^+ (Fe) state, then the $B(\text{E2}; 3/2_1^- \to 7/2_1^-)$ value should rise at N = 38, making the measurement of this quantity in ⁶⁵Co very important. The $3/2^-$ lifetime was recently measured in ⁶⁵Co [109] with a value $\tau=5(5)$ ps leading also to a B(E2) = 17(16) W.u to be compared to 22(2) W.u for the B(E2, $2^+ \to 0^+)$ in ⁶⁴Fe and 7(1) W.u in ⁶⁶Ni making the experimental conclusion elusive. It is however presently proposed that it belongs to the $\pi f_{7/2}^{-1} \otimes 2^+$ Ni state multiplet [109].

3.2.2 The $9/2_1^-$ state

The lifetime to the ground state was measured in 63,65 Co in [108]. The $9/2_1^- \rightarrow 7/2_1^-$ transition probability is even more difficult to interpret since the decay occurs by E2 or M1, and the interpretation depends on the assumed multi-polarity. Presently the E2/M1 mixing ratio is not known. Assuming a pure E2 transition, the $B(E2; 9/2_1^- \rightarrow 7/2_1^-)$ value extracted for 63 Co is compatible with the $B(E2; 2_1^+ \rightarrow 0_1^+)$ value in 64 Ni. It is proposed that it belongs to the $\pi f_{7/2}^{-1} \otimes 2^+$ Ni state multiplet in [108, 109].

3.2.3 The $11/2_1^-$ state

In [110, 107, 109], the lifetime of $11/2^-$ states at 1664 (1674) (1480) keV in ⁶¹Co (⁶³Co) (⁶⁵Co) respectively have been measured and were interpreted also as a $\pi f_{7/2}^{-1} \otimes 2^+$ Ni state based on the deduced transition strength similar to the B(E2,2⁺ $\rightarrow 0^+$) in Ni.

3.2.4 The $1/2_1^-$ state

This state plays a major role in the shape coexistence scenario. In [102], D. Pauwels *et al.* reported the observation of a $J^{\pi} = (1/2^{-}) \beta$ isomeric state at 491 keV in ⁶⁷Co and interpreted as a deformed proton intruder state 1p-2h across the Z=28 shell gap. A $(1/2^{-})$ proton state is proposed at 1095 keV in ⁶⁵Co [105]. The E2/M1 mixing ratio of the transition to the $3/2^{-}$ state is unknown [109]. Assuming a pure E2 transition to the $3/2^{-}$ state, the deduced B(E2) from its lifetime is found large, B(E2) = 67(1) W.u. On the contrary, a pure M1 multi-polarity leads to low B(M1). It is presently interpreted as a member of the deformed configuration [109]. This deformed state is not populated in multi-nucleon transfer reaction [108, 111].

	Table 3.1: Measured lifetime (τ [ps]) in odd-mass Co isotopes.				
		A. Dijon	M. Klintefjord	V. Modamio	B. Olaizola
		[108]	[107]	[110]	[109]
$^{63}\mathrm{Co}$	$(3/2^{-})_{995keV}$	15.4(18)			
	$(9/2^{-})_{1383keV}$	0.9(4)			
	$(11/2^{-})_{1674keV}$		0.55(19)	1.0(3)	
$^{65}\mathrm{Co}$	$(3/2^{-})_{882keV}$				5(5)
	$(9/2^{-})_{1480keV}$	$\leq \! 17.3$			
	$(11/2^{-})_{1480keV}$	$\leq \! 17.3$		1.3(5)	
	$(1/2^{-})_{1095keV}$				1803(28)

1 1.0 .. 1. 1 1

Table 3.2: Measured $B(\lambda \mu)$ [W.u.] in odd-mass Co isotopes and even-even Ni isotopes.

		Assuming Pure E2	Assuming Pure M1
N=36			
$^{63}\mathrm{Co}$	$(3/2^{-})_{995keV}$	3.7(4)	-
	$(9/2^{-})_{1383keV}$	12(5)	0.013(4)
	$(11/2^{-})_{1674keV}$	4(1)	-
⁶⁴ Ni	$2^+_{1345.75keV}$	9.04(28)	-
N=38			
$^{65}\mathrm{Co}$	$(3/2^{-})_{882keV}$	17(16)	-
	$(9/2^{-})_{1480keV}$	*	*
	$(11/2^{-})_{1480keV}$	6(2)	-
⁶⁶ Ni	$2^+_{1424.8keV}$	7.6(13)	-
	$(1/2^{-})_{1095keV}$	67(1)	$1.83(3) \times 10^{-3}$

3.2.5Summary of the experimental results

The measured lifetimes are summarized in table 3.1 and deduced transition probabilities in table 3.2. The experiments in [108, 107, 110] correspond to plunger measurements in multi-nucleon transfer reaction using different heavy beams and targets. Even if the range in sensitivity is similar, they differ in populated states making them complementary. The reference [109] is a fast-timing measurement after β -decay populating different states. From these set of measurements, it is concluded that the $(3/2^{-})$, $(9/2^{-})$ and $(11/2^{-})$ states are part of the $\pi f_{7/2}^{-1} \otimes 2^{+}$ Ni multiplet based on the similarities in excitation energy and corresponding B(E2) to the ground state. This conclusion is supported by comparison with shell model calculations. However, one could argue that the experimental conclusion is overstated regarding the uncertainties highlighted in table 3.2. First, the B(E2) for the $(9/2^-)$ state in ⁶³Co is not determined accurately since the $\delta(E2/M1)$ mixing ratio is not measured. The $(3/2^-)$ in ⁶⁵Co has 100% error bar and the relatively precise value for the $(3/2^{-})$ and $(11/2^{-})$ states in ⁶³Co differ significantly from the B(E2) in ⁶⁴Ni. Finally, only the $(11/2^{-})$ state in ⁶⁵Co fits well the weak coupling scheme. More precise measurements are therefore needed to probe experimentally the coupling scheme and track possible deviation from the model to probe the influence of the deformed configurations. Spins and parities must be uniquely determined and $\delta(E2/M1)$ mixing ratio measured. Finally, higher precision lifetime measurements or Coulomb excitation cross-sections are mandatory to extract with smaller error bar the reduced transition probabilities.

The $(1/2^{-})$ is interpreted in comparison with shell model calculation as a deformed proton state involving excitation above the Z=28 gap in [109]. There is, today, no unambiguous experimental data supporting the interpretation. Indeed, the long lifetime measured in [109] arises mainly from the low energy E2 transition to the $(3/2^{-})$ state and cannot be used as an argument on its structure since the $\delta(E2/M1)$ mixing ratio is not measured. A $(1/2^{-})$ is not part of the $\pi f_{7/2}^{-1} \otimes 2^{+}$ Ni multiplet and in the interpretation of a large deformation, the $(1/2^{-})$ spin parity can be related to a single proton excitation to the $p_{3/2}^{1}$ $(1/2^{-}[321])$ orbit. One could speculate that when neutron are added to the $g_{9/2}$ orbit, this state could become the ground state as a result of the onset of deformation established in the Fe isotopes. The ground state spin and parity beyond N=40 in Co isotopes remain unknown. The possible spin/parity inversion of the ground state from $7/2^{-} \rightarrow 1/2^{-}$, as suggested by the excitation energy systematic, is not yet established.

3.2.6 Theoretical calculations

Calculations from [108] are compared with the data in [108]. It is clear that the inclusion of the $1q_{9/2}$ orbital becomes imperative as the neutron number approaches N = 40. This conclusion can be drawn by observing that, towards N = 40, excitation energies are overestimated in a pf model space and B(E2) values are underestimated. Nevertheless, even with the pfg interaction which includes the $1g_{9/2}$ orbital, the excitation energies of the $3/2^-_1$ and $9/2^-_1$ levels are slightly higher and the B(E2) values of the transitions to the ground state are somewhat smaller in ⁶⁷Co compared to lighter Co isotopes, indicative of some effect of an N = 40 sub-shell closure in the calculation. Our measured $B(E2; 3/2_1^- \rightarrow 7/2_1^-)$ value in ⁶³Co falls well below the value calculated with pfg, which indicates that the shell model overestimates the collectivity of the $3/2_1^-$ state. On the other hand, the calculated $B(E2; 9/2^-_1 \rightarrow 7/2^-_1)$ value in ⁶³Co comes out at 11.1 W.u, in excellent agreement with the experimental value of 11.4 (55) W.u, obtained under the assumption of pure E2. However, although the E2 rate is well reproduced by the shell model, it fails to obtain the correct B(M1) value for this transition. This can be inferred from the lifetime of the $9/2^{-}_{1}$ level which is calculated a factor 10 shorter than the measured value of 0.9 (4) ps due to a large calculated $B(M1; 9/2_1^- \rightarrow 7/2_1^-)$ value of 0.36 W.u, obtained with bare nucleon g factors. This result is not significantly altered if the spin g factors are quenched by a factor 0.7.

To what extent are the simple interpretations, mentioned in the introduction, of a $1\pi f_{7/2}$ particle coupled to a 0^+_1 or 2^+_1 in Fe or a $(1\pi f_{7/2})^{-1}$ hole coupled to a 0^+_1 or 2^+_1 in Ni, borne out by the shell-model calculations? This can be illustrated with the example of the $7/2^-_1$ and $9/2^-_1$ states. The most important components of these states are

$$\begin{aligned} |7/2_{1}^{-}\rangle &\approx 0.62 |(1\pi f_{7/2})^{-1} \otimes 0_{1}^{+}(\mathrm{Ni}); 7/2\rangle \\ &\pm 0.38 |(1\pi f_{7/2})^{-1} \otimes 2_{1}^{+}(\mathrm{Ni}); 7/2\rangle + \cdots, \\ |9/2_{1}^{-}\rangle &\approx 0.62 |(1\pi f_{7/2})^{-1} \otimes 2_{1}^{+}(\mathrm{Ni}); 9/2\rangle \\ &\pm 0.31 |(1\pi f_{7/2})^{-1} \otimes 4_{1}^{+}(\mathrm{Ni}); 9/2\rangle + \cdots. \end{aligned}$$
(3.1)

These components represent barely 50% of the total. There is thus considerable fragmentation of the wave function in the shell model and, accordingly, the simple interpretations at best are only qualitatively valid. This fragmentation also explains the large $B(M1; 9/2_1^- \rightarrow 7/2_1^-)$ value obtained in the shell model. No M1 transition is allowed between the main components in the decomposition (3.1) since they involve different core states $(0_1^+ \text{ and } 2_1^+, \text{ respectively})$. On the other hand, the first component of the $9/2_1^-$ state and the second component of the $7/2_1^-$ state are connected by the proton part of the M1 operator, and this leads to the large calculated B(M1) value. Given that the measured lifetime of the $9/2_1^-$ level implies a small $B(M1; 9/2_1^- \rightarrow 7/2_1^-)$ value, a tentative conclusion of our analysis is that the shell model using these interaction predicts too much fragmentation. No new recent measurement of the $3/2^-$ (Fe-like) and $9/2^-$ (Ni-like) lifetime have been reported since.

The measured B(E2) were finally compared to state-of-the-art Shell Model calculations using the LNPS interaction [92]. Calculations reproduce well the experimental level scheme [112] and the measured lifetime of the $11/2^{-}$ states [107]. In [109], the B(E2) are compared to measured values in 65 Co and in particular to the proton deformed $(1/2^{-})$ state. Experimentally, its B(E2) to the $3/2^{-}$ is surprisingly high, assuming a pure E2 transition. This large B(E2) is not compatible with the calculated value. This is unexpected since the interaction has reproduced well the spectroscopic data in the mass region. However, it must be underlined that the experiment was not design to measure the M1/E2 mixing ratio. Assuming instead a pure M1 transition, the calculated B(M1) is 6.10^{-3} Wu to be compared to 2.10^{-2} Wu in LNPS which is reasonably close. This is supporting that the transition is dominated by the M1 component and the corresponding B(E2) probably low. To support the dominant M1 contribution, the Weisskopf estimates can be used. The Weisskopf estimates for the 212.7 keV decay to the $(3/2^{-})$ state in ⁶⁵Co are $\tau = 123$ ns for an E2 and $\tau =$ 3 ps for an M1. The measured value is $\tau = 1.80(2)$ ns. Using the E2 Weisskopf estimates, a mixing ratio $\delta(E2/M1) \sim 0.01$ is calculated and is supporting a large domination of the M1 component. The $(1/2^{-})$ state is interpreted as a deformed proton intruder state. In the even-even Ni core, such intruder appears as a $(2p-2h) 0^+$ state [102]. The shape isomers will be discussed in the next section but as the collectivity of excited states in Co is discussed, a comparison between the $0_{def}^+ \rightarrow 2_1^+$ in the even-even Ni core and the $(1/2^{-}) \rightarrow (3/2^{-})$ will be discussed in the following.

In ⁶⁶Ni, the deformed 0_4^+ proton state is located at 2974 kev with a B(E2) to the 2_1^+ state of 0.2 W.u. [113]. In [114], the LNPS calculations leads to a low B(E2)=8.10⁻³ W.u. The MCSMC from [113] predicts a B(E2)= 6.10⁻³ W.u. Both experimental and calculations are coherent with a low B(E2). The $(1/2^-) \rightarrow (3/2^-)$ B(E2) in ⁶⁵Co is also found small. In ⁶⁸Ni, the deformed proton state is located at 2511 keV (see next section). According to [115], the B(E2) to the 2_1^+ is 2.3 W.u. The published LNPS calculations are 0.9 W.u [115, 116] in reasonable agreement. The LNPS wave function for the $1/2^-$ state at 1290 keV in ⁶⁵Co has been published in [109] and has great similarities with the wave function of the 0⁺ proton deformed states in ⁶⁶Ni. The deformed proton



Figure 3.2: Level scheme and B(E2) in W.u for the ^ACo and ^{A+1}Ni isotopes. The member of the $2^+(^{A+1}Ni) \times \pi f_{7/2}^{-1}$ multiplet and the deformed $1/2^-$ states are shown for the Co isotopes. The first 2^+ and proposed deformed 0^+ state in Ni are shown.

state in Co corresponds to a 1 proton excitation from the $f_{7/2}$ to the $f_{5/2}$ and neutrons excitations from fp to $g_{9/2}$.

3.3 Shape Isomers and deformed state in the vicinity of ⁶⁸Ni

The quest for a $\pi(2p-2h) 0^+$ intruder state in ⁶⁸Ni is a long story. The first speculation for a shape isomer in this nuclei was published in [117, 100]. Using the ⁷⁰Zn(¹⁴C, ¹⁶O)⁶⁸Ni reaction, excited states have been populated in a pure 2 protons transfer. Two excited states at 1.77 MeV and 2.20 MeV have been identified and interpreted by differential cross section to be the 0^+_2 and 2^+_1 states respectively. The 2^+_1 was later proposed at 2033 keV. The presence of a 0^+ state as first excited state triggered discussion on its nature and the mean-field calculations using the Gogny interaction display a spherical minimum, but a second minimum exists at large deformation β = 0.3. This minimum could correspond to the observed 0^+_2 isomeric state. The excitation energy of the 0^+_2 isomeric state has been recently remeasured and placed at an energy of 1604 keV (instead of 1770 keV) based on the direct detection of the $0^+_2 \rightarrow 0^+_1$ E0 transition [118] and was interpreted as a neutron state with 2 particles scattered from the $p_{1/2}$ into the $g_{9/2}$ of moderate oblate deformation ($\beta \sim 0.2$) [118, 111]. The ⁶⁶Ni(t,p)⁶⁸Ni reaction was performed recently and 2 neutron transfer cross sections have shown a strong population of the 0^+_1 state and a weak population of the 0^+_2 state [119, 120]. This result is consistent with the $\nu g^{9}_{9/2}$ interpretation of the 0^+_2 state.

Despite the large number of experiments dedicated to 68 Ni, no evidence for a 0⁺ state based on a proton-pair excitation could be found. In [121], W. F. Mueller et al. proposed a (0^+) state at 2511 keV with a unique γ decay to the 2^+_1 state, populated in the decay of a low spin (3⁺) β isomer (T_{1/2}=1.6(3) s.) in ⁶⁸Co. The state in ⁶⁸Ni was not populated in the β -decay of the (7^{-}) ground state. The proposed spin/parity for the (3^{+}) isomer leads to an interpretation of a neutron state based on a pair excitation from the $f_{5/2}$ into the $g_{9/2}$ for the 2511 keV state. In [102], D. Pauwels et al. reported the observation of a $J^{\pi} = (1/2^{-}) \beta$ isomeric state at 491 keV in 67 Co and interpreted as a deformed proton intruder state (1p-2h) across the Z=28 shell gap. The proposed wave function for the proposed $(1/2^{-})$ state is a $\pi(f_{7/2}^{-2}p_{3/2}^{+1})$ deformed configuration with the Nilsson $1/2^{-}[321]$ orbital. The coupling with neutron excited from the pf to the intruder gd shell give rise to the reduction of the Z=28 effective shell gap and to the low excitation energy of the intruder state. This isomeric state associated to the 7/2⁻ π (2p-1h) in ⁶⁹Cu at 1711 keV allows the prediction of a $\pi(2p-2h)$ deformed state at 491+1711 = 2202 keV in ⁶⁸Ni [3]. In [116], we reported the observation of delayed 168(1) keV γ -ray transition with $T_{1/2} = 216^{+65}_{-50}$ ns in ⁶⁸Ni populated by the two proton transfer reaction ²³⁸U(⁷⁰Zn, ⁶⁸Ni)²⁴⁰Pu. We proposed as assignment for this $E_{\gamma}=168(1)$ keV transition the decay from a new 0_3^+ state at 2202(1) keV to the 2_1^+ . Because of the very limited statistics, it was unfortunately not possible to use γ - γ coincidences to build the level scheme. The non-observation of the $2^+_1 \rightarrow 0^+_1$ decay is consistent both with the very low cross-section to populate this new isomer and with the low efficiency at the focal plane at 2 MeV. Weisskopf estimates for the half-life of an E1, E2, M1 and M2 168 keV transition are 0.1 ps, 300 ns, 6 ps and 20 μ s respectively. The measured half-life indicates therefore an E2 character. The corresponding very low $B(E2: 0_3^+ \to 2_1^+) = 25.3(4)e^2 fm^4$ (1.5(2) W.u.) (assuming a pure E2 transition), indicates a single particle character.

The position of the proposed 0^+_3 was in a remarkable agreement with the 2202 keV π (2p-2h) 0⁺



Figure 3.3: Shape coexistence at N=40. Evolution of the 2p-2h proton *intruder* and *normal* $\nu g_{9/2}$ states in ⁶⁸Ni, ⁶⁷Co and ⁶⁶Fe.

intruder state deduced by D. Pauwels et al. [3]. In [116], we performed large shell model (LSSM) calculations with the most recent interaction tailored for the mass region (LNPS) and in an extended valence space for 68 Ni [92]. In these calculations, a third 0^+_3 state is found at 2.4 MeV for which the dominant configuration is a pure $\pi(2p-2h)$ i.e. $(f_{7/2})^{-2}$. This *intruder* state has only pure *normal* parity neutron states as neighbours. The 2^+_1 state wave function is calculated to be a pure $\nu(2p-2h)$ configuration and hence with a negligible overlap with the proposed state. Therefore the B(E2) is expected to be very small, possibly leading to an isomeric character as observed. The calculations show that the strong deformation in the Cr chain is the result of the proton-neutron correlations between the neutrons populating the $0g_{9/2}$ - $1d_{5/2}$ orbitals and the valence protons in the pf shell. The calculated deformation for the $0^+_{1,2,3}$ states are $\beta_2 \simeq 0.18$, 0.24 and 0.4 respectively. The shell model calculations using LNPS published in [116] give for the first time the microscopical description of the deformed intruder proton state. The 0^+_2 is a pure neutron state with 2 neutron in the $g_{9/2}$ from the pf shells. The 0_3^+ is a strict excitation of proton pair from the $f_{7/2}$ into the $f_{5/2}$ with a fragmented neutron wave function in the $pf - g_{9/2}$ orbitals as a signature of large deformation. In the calculations, a fourth 0^+ state compatible with the state observed by W. F. Mueller et al. proposed at 2511 keV [121] is not predicted.

Following the publication, several experiments focussed on the spectroscopy of 68 Ni. The new isomer was not confirmed by [122] and a delayed 168 keV γ -ray in coincidence with the 2^+_1 decay was excluded. However, the spin parity assignment of the (0^+) state at 2511 keV proposed by [121] was confirmed by the $0 \rightarrow 2 \rightarrow 0$ angular correlation measurement. Presently, 2 excited 0^+ states are confirmed in ⁶⁸Ni. The conclusion of [122] is "Particular attention was devoted to searching for the decay of the proton-2p2h, 2202-keV, 0^+ isomer. No evidence for such an intruder state was observed in the present study. Given the impact such a state would have on understanding the structure of nuclei in the ^{68}Ni region, more conclusive evidence of its location is required." . The 2511 keV 0_3^+ state is compatible with the shell model prediction for a 2p-2h proton intruder state and the results of the other experiments, even if its shape and microscopical configuration was never measured, support the shape isomer interpretation [116, 111, 118, 123, 115]. It is worth mentioning that the E0 branch from the 0_3^+ decay was not observed [123, 115] but its lifetime was established at $T_{1/2}=0.57(5)$ ns. Using upper limit for the E0 branching ratios to the 0_2^+ and 0_1^+ states, small $\rho^2(E0) \leq 5$ m.u and ≤ 25 m.u. respectively were extracted. The low $\rho^2(E0)$ could be interpreted as a signature of small differences in deformation between state or more likely as a signature of a low mixing of the respective wave functions. In [111, 123], a 2^+_2 state at 2742.6 keV is proposed as band member of the 0^+_3 intruder state. Such rotational band with a $E_{\gamma}(2^+ \to 0^+)$ = 232 keV in $^{68}\mathrm{Ni}$ leads to an approximated deformation parameter $\beta_2 \sim 0.45$ using the Ramans estimate, compatible with the LNPS calculations.

In the present understanding, the ⁶⁸Ni ground state configuration has a moderate deformation. The 2_1^+ and 0_2^+ states correspond to a neutron pair scattered into the $g_{9/2}$ orbital. The configuration of the highly deformed intruder 0_3^+ state, with valence protons holes in the pf shell, is very similar to the Fe $(0_1^+/2_1^+)$ states, i.e. with two proton holes in the $\pi f_{7/2}$ orbital. Therefore, the configuration corresponding to the 0_3^+ state in ⁶⁸Ni migrates down to 491 keV excitation energy in ⁶⁷Co (the $(1/2^-)$ isomer) and becomes the ground state in ⁶⁶Fe when removing protons to the Z=28 shell closure. The normal configuration, by opposition to the intruder one, might then appear as a low-lying 0^+ state in ⁶⁶Fe. In [124, 125, 114], a 0_2^+ state at 1414 keV was identified in the



Figure 3.4: Shape coexistence at N=40. Microscopical configuration illustration.



Figure 3.5: Systematics of proton deformed excited 0^+ state in Ni isotopes.

 β -decay of ⁶⁶Mn. The shell model calculations predict such a state at the proper energy with a normal configuration $\pi f_{7/2}^5 (p_{3/2} f_{5/2} p_{1/2})^1 \nu p_{3/2}^4 f_{5/2}^4 p_{1/2}^2 g_{9/2}^2$ similar to the $(0_2^+/2_1^+)$ states in ⁶⁸Ni [124]. As a summary, this shape coexistence scenario is illustrated in figures 3.4 and 3.3.

Recent large scale shell model calculations have provided wave function description of the 0⁺ states in ⁶⁸Ni. The LNPS [116, 123] and MCSMC [126], state-of-the-art, are compared in figure 3.6. Even if the approaches are slightly different, the proposed configurations are very similar for the 0_3^+ state with a pair excitation of proton from the $f_{7/2}$ into the $f_{5/2}$ and fragmented neutron wave function with significant increase of the $g_{9/2}$ occupancy. The p-n interaction between the $\pi f_{7/2}^{-2}$ and $\nu g_{9/2}^3$ gives rise to the deformed configuration.

It is worth mentioning that since this intense experimental work on 68 Ni, similar proton deformed configurations have been proposed in 66 Ni [113, 114, 127] and in 70 Ni [128, 129, 130] giving rise to a triple shape coexistence scenario evolution between 66 Ni and 70 Ni. The question of low lying deformed 0⁺ state in the neutron mid-shell in 72,74 Ni is therefore opened. From [113], four 0⁺ states have been identified in 66 Ni. How these 3 excited 0⁺ states evolve in 68 Ni leaves room for a possible explanation to the 168 keV delayed γ -ray transition.

Even if the experimental data show consistency in even-even nuclei, the β -decay chain for the mass A=68 deserves some attention since the spin parity of the β -isomer in ⁶⁸Co plays a major role in the possible constraint of the microscopic configuration of the proton 2p-2h intruder state in ⁶⁸Ni. After the discovery of the short lived β -isomer in ⁶⁸Co [121], several spin/parity assignments were proposed in the light of the populated excited states in ⁶⁸Ni. The claim for a proton 0⁺ 2p-2h state at 2511 keV in ⁶⁸Ni should make a coherent β -decay chain for A=68. This review is proposed below. The list of the different assumptions for the spins and parities assignment is chronological.

- (7^-) high spin ground state β -decay. In [121], the high spin ground state $T_{1/2}=0.23(3)$ s. is proposed as a 7⁻ state with the $\pi f_{7/2}^{-1} \otimes \nu g_{9/2}^{+1}$ configuration. The decay of this state doesn't populate the 0_3^+ state in ⁶⁸Ni but only pure neutron states. Indeed, the $\nu f_{5/2} \rightarrow \pi f_{7/2}$ Gamow-Teller is favoured and populates closed proton shell in ⁶⁸Ni with neutron states having the $\nu f_{5/2}^{-1} g_{9/2}^{+1}$ configuration compatible with the $(6^-, 7^-)$ states assumptions.
- (3^+) low spin isomer assumption. In [121], the low spin isomer $T_{1/2}=1.6(3)$ s. state is proposed



Figure 3.6: Proton and neutron occupancy for the 0^+ states in 68 Ni for (left) LNPS from [116, 123] and (right) MCSMC from [126]

as a (3^+) state with the $\pi f_{7/2}^{-1} \otimes \nu g_{9/2}^{+2} p_{1/2}^{-1}$ configuration. The decay of this state shows a weak population of the 0_3^+ state in ${}^{68}Ni$. Similarly to the (7^-) ground state, a (3^+) would favour a $\nu f_{5/2} \to \pi f_{7/2}$ Gamow-Teller, populating pure neutron states having the $\nu p_{1/2}^{-1} f_{5/2}^{-1} g_{9/2}^{+2}$ configuration. According to shell model calculations, this is the exact configuration of the 0_2^+ and 2_1^+ states in ${}^{68}Ni$. The decay of the 0_2^+ was not observed in [121] but the branching ratio to the 2_1^+ is the largest ($\leq 39(8)\%$). Proton states at 5 MeV in ${}^{68}Ni$ are also populated indicating that there is a significant $\nu p_{1/2} \to \pi p_{3/2}$ Gamow-Teller strength leading to proton states with the $\pi f_{7/2}^{-1} p_{3/2}^{+1} \otimes \nu p_{1/2}^{-2} g_{9/2}^{+2}$ configuration. Without the gain in pairing energy, such 1p-1h π states lies at higher excitation energy than a 2p-2h state. The weak population of the proposed 2p-2h proton state in ${}^{68}Ni$ would involve the combination of a $f_{5/2} \to \pi f_{5/2}$ Gamow-Teller with a $f_{7/2} \to f_{5/2}$ proton excitation. This could be compatible with the low branching ration to the 0_3^+ assuming the 2p-2h proton configuration.

- (1^+) low spin isomer assumption. In [131], ⁶⁸Co was populated from the ⁶⁸Fe ground state • favouring the low spin isomer. It is proposed a new (1^+) spin/parity assignment for the isomer fed by an E1 transition from a state at 45 keV above, thus a 2^{-} or 1^{-} state. The (1^{+}) assignment suggests a proton excitation into the deformed intruder orbital coupled to two neutrons in the $g_{9/2}$, ie the $\pi f_{7/2}^{-2} p_{3/2}^{+1} \otimes \nu p_{1/2}^1 g_{9/2}^{+2}$ configuration. The $\nu p_{1/2} \to \pi p_{3/2}$ Gamow-Teller should be favoured and populates states with the $\pi f_{7/2}^{-2} p_{3/2}^{+2} \otimes \nu p_{1/2}^0 g_{9/2}^{+2}$ configuration, ie a proton pair excitation above the Z=28 gap into the $p_{3/2}$ orbital. This configuration is not the exact configuration expected for the two proton intruder at low excitation energy since, according to calculations, the proton pair is predicted into the $\pi f_{5/2}$ (the gain in pairing energy is more important). But proton state should be anyhow favoured. Instead, the β feeding shows a large feeding of the neutron states in 68 Ni. As a result, the proposed (1⁺) ground state is very unlikely. The decay by an E1 transition at 45 keV could instead lead to the population of a 1^+ state that decays to a $(2^-, 1^-)$ isomeric state, is a reverse assignment to the one proposed in [131]. This $(2^-, 1^-)$ state assignment leads to a $\nu(p_{1/2}^{-2}g_{9/2}^3) \otimes \pi(f_{7/2}^{-1})$ or $(f_{7/2}^{-2}p_{3/2}^{+1})$ configuration. Such configuration with a fully depopulated $\nu p_{1/2}$ will favour the $\nu f_{5/2} \rightarrow \pi f_{7/2}$ Gamow-Teller and populate pure neutron states as experimentally observed.
- (2^-) low spin isomer assumption. In [123], the (2^-) spin/parity assignment is finally favoured. But the proposed configuration is a low-lying deformed [321]1/2⁻ proton configuration coupled to the down-sloping [431]3/2⁺ and [440]1/2⁺ neutron orbitals originating from the spherical $g_{9/2}$ orbital, thus one of the previously proposed configuration $\nu(f_{5/2}^6 p_{1/2}^{-2} g_{9/2}^3) \otimes \pi(f_{7/2}^{-2} p_{3/2}^{+1})$. Such configuration will favour the $\nu f_{5/2} \to \pi f_{7/2}$ Gamow-Teller transition. The populated states will be $\nu(f_{5/2}^5 p_{1/2}^{-2} g_{9/2}^3) \otimes \pi(f_{7/2}^{-1} p_{3/2}^{+1})$. It means proton single particle state as observed in [121] and to some extend if the excited proton in the $p_{3/2}$ decays to the $f_{7/2}$, the 0_2^+ and 2_1^+ neutron states in ⁶⁸Ni as experimentally observed. This excludes a strong population of the proton 2p-2h state by β -decay into the ⁶⁸Ni.

As a summary, neither the direct (t,p) reaction [120] or β -decay from the low or high spin states in ⁶⁸Co can strongly populate the 2p-2h deformed proton intruder state in ⁶⁸Ni. From this compilation, the (1⁺) assignment for the low spin isomer is very unlikely. The (3⁺) and (2⁺) assumptions are compatible with the current interpretation of the 0⁺₃ state in ⁶⁸Ni. Figure 3.7 illustrates the compilation of the data used in this review. If possible, the (³He, n) reaction from the deformed ⁶⁶Fe or α -transfer from ⁶⁴Fe would favour the population of the deformed state in



Figure 3.7: Multi-messenger approach for the 0^+ states in 68 Ni



Figure 3.8: The 168 keV transition mystery

⁶⁸Ni.

The coherence with the ${}^{70}Zn({}^{14}C,{}^{16}O){}^{68}Ni$ data remains puzzling. In the two experiments by [100] and [117], the 0_2^+ and 2_1^+ states were proposed first at 1770 and 2200 keV respectively. Later measurements, as mentioned before, corrected the respective values at 1604 and 2033 keV. The two corrections are 166 and 167 keV respectively (see figure 3.8), very close to the proposed delayed line at 168 keV which was never confirmed later. This systematic shift of 167(1) keV is disturbing and gives room for a possible explanation for the 168 keV γ -ray observed in [116].

The microscopical structure of ⁷⁰Zn would reveal the possibility to populate the proper state in a strict 2 proton transfer. It is proposed to discuss, from the constrained theoretical wave functions of ⁷⁰Zn, the probability to populate the deformed proton state in ⁶⁸Ni. For this purpose, we use, as constrain, the B(E2, $2^+ \rightarrow 0^+$) measured using different experimental methods. The compilation of the measured B(E2, $2_1^+ \rightarrow 0_1^+$) in ⁷⁰Zn is presented in figure 3.9 and compared to Shell Model calculations using the LNPS and JUN45 interactions [132, 104, 133]. The contributions to the ground state configurations using the JUN45 interaction are listed in table 3.3. The JUN45 calculations for the B(E2) is shown in figure 3.9 and reproduces well the experimental average value with a close Z=28 proton shell, indicating that the wave function doesn't involve any excitation from the ⁵⁶Ni core. The two main contributions are : $\pi(p_{3/2}^2 f_{5/2}^0) \otimes \nu(f_{5/2}^4 p_{1/2}^2 g_{9/2}^2)$ and $\pi(p_{3/2}^2 f_{5/2}^0) \otimes \nu(f_{5/2}^4 p_{1/2}^2 g_{9/2}^2)$. The feeding of the proton deformed state in ⁶⁸Ni in a 2 protons stripping reaction is excluded in the second configuration since the neutron $g_{9/2}$ orbital is not occupied.

The configurations with the highest amplitude involve several neutrons scattered into the $g_{9/2}$ orbital and two protons in the $p_{3/2}$ orbital. The amplitude corresponding to an occupied $f_{5/2}$ proton orbital (**) account for 7% of the wave function. Other theoretical calculations [134] propose a wave function for the ⁷⁰Zn ground state as $\pi(f_{7/2}^{7.4}p_{3/2}^{1.2}f_{5/2}^{0.7}) \otimes \nu(f_{5/2}^{3.7}p_{1/2}^{0.7}g_{9/2}^{3.3})$, ie with scattered

Table 3.3: Calculated wave function for the ground state of 70 Zn using the JUN45 interaction. The configurations indicated with (**) are compatible with the population of the deformed proton state in 68 Ni by a 2 proton stripping from 70 Zn.

Amplitude	70 Zn
0.1456	$\frac{\nu(p_{3/2}^4f_{5/2}^4p_{1/2}^2g_{9/2}^2)\pi(p_{3/2}^2f_{5/2}^0)}{\nu(p_{3/2}^4f_{5/2}^4)}$
0.1275	$\nu(p_{3/2}^4f_{5/2}^6p_{1/2}^2g_{9/2}^0)\pi(p_{3/2}^2f_{5/2}^0)$
0.0941	$\nu(p_{3/2}^4f_{5/2}^6p_{1/2}^0g_{9/2}^2)\pi(p_{3/2}^2f_{5/2}^0)$
0.0677	$\nu(p_{3/2}^2f_{5/2}^6p_{1/2}^2g_{9/2}^2)\pi(p_{3/2}^2f_{5/2}^0)$
0.0564	$\nu(p_{3/2}^4f_{5/2}^4p_{1/2}^0g_{9/2}^4)\pi(p_{3/2}^2f_{5/2}^0)$
0.0451	$\nu(p_{3/2}^2 f_{5/2}^4 p_{1/2}^2 g_{9/2}^4) \pi(p_{3/2}^2 f_{5/2}^0)$
0.0221	$\nu(p_{3/2}^4f_{5/2}^4p_{1/2}^0g_{9/2}^4)\pi(p_{3/2}^0f_{5/2}^2) \ **$
0.0212	$\nu(p_{3/2}^2 f_{5/2}^6 p_{1/2}^0 g_{9/2}^4) \pi(p_{3/2}^2 f_{5/2}^0)$
0.0203	$\nu(p_{3/2}^4f_{5/2}^4p_{1/2}^2g_{9/2}^2)\pi(p_{3/2}^0f_{5/2}^2)^{**}$
0.0200	$\nu(p_{3/2}^4f_{5/2}^2p_{1/2}^2g_{9/2}^4)\pi(p_{3/2}^2f_{5/2}^0)$
0.0167	$\nu(p_{3/2}^3f_{5/2}^6p_{1/2}^1g_{9/2}^2)\pi(p_{3/2}^2f_{5/2}^0)$
0.0149	$\nu(p_{3/2}^4f_{5/2}^5p_{1/2}^1g_{9/2}^2)\pi(p_{3/2}^2f_{5/2}^0)$
0.0141	$\nu(p_{3/2}^4f_{5/2}^6p_{1/2}^0g_{9/2}^2)\pi(p_{3/2}^0f_{5/2}^2)^{**}$
0.0130	$\nu(p_{3/2}^2f_{5/2}^4p_{1/2}^2g_{9/2}^4)\pi(p_{3/2}^0f_{5/2}^2)^{**}$



Figure 3.9: Systematics of the measured $B(E2,2_1^+ \rightarrow 0_1^+)$ in ⁷⁰Zn. Experimental data (a) [135], (b) [136], (c) [132], (d) [137], (e) SM JUN45, (f) SM LNPS. The average value is 299(11) $e^2 fm^4$ to be compared to 302 and 327 for the JUN45 and LNPS calculations respectively.

neutron in the $g_{9/2}$ and also with a very low occupancy of the proton $f_{5/2}$.

The ¹⁴C beam has a $p_{1/2}^{-2}$ proton configuration considering a ¹⁶O core. Therefore, the pair transfer should favour $\Delta l=0$, and the removal of the two protons in ⁷⁰Zn from the $p_{3/2}^2$ would be favoured. The two protons stripping from the occupied $p_{3/2}$ or $f_{5/2}$ proton orbital will give rise to the wave function of the 0_1^+ and $(0_2^+$ and $2_1^+)$ neutron states in ⁶⁸Ni according to the proposed calculations. Therefore, the main feeding in ⁶⁸Ni should populate neutron states. The remaining four configurations with two protons in the $f_{5/2}$ orbital could populate the deformed proton state if protons are removed from the $f_{7/2}$ core with a $\Delta l=2$ transfer, ie giving a similar 2 hole proton configuration into the $f_{7/2}$. This transfer would correspond to a very low cross section. As a conclusion, based on the present discussion on the ⁷⁰Zn ground state wave function, it is very unlikely that the experiments performed in [100] and [117] have populated the proton deformed state in ⁶⁸Ni. In these experiments, the ground state is populated with the highest cross section, indicating that the second calculated wave function, with empty neutron $g_{9/2}$ orbital, should be the dominant configuration in ⁷⁰Zn. The 0_2^+ and 2_1^+ states are populated with two orders of magnitude less giving a very weak chance to observe the low cross section to the proton deformed state.

One could propose two reactions compatible with the population of the proton state in ⁶⁸Ni. Obviously, starting from a deformed configuration will increase the cross section. Both ⁶⁴Fe and ⁶⁶Fe ground state have a proton $(f_{5/2})^{-2}$ configuration and the lifetime measurements show that both neutron $g_{9/2}$ and $d_{5/2}$ orbitals are already populated in ⁶⁴Fe [106]. Therefore the ⁶⁴Fe(⁷Li,t)⁶⁸Ni (α transfer) or ⁶⁶Fe(³He,n)⁶⁸Ni reactions could favor the population of the proton deformed state.

3.4 Q_s systematics

Contrary to the two previous mass regions, experimental spectroscopic quadrupole moments have not been measured in the vicinity of 68 Ni (last measured is 64 Ni), in Co (last measured is 60 Co), Fe (last measured is 61 Fe) and Mn (last measured is 56 Mn) isotopes. Therefore, the large overview of spectroscopic quadrupole moments presented in the N=Z and A=100 mass region cannot be produced. This underlines the lack of experimental data in the mass region. Ni, Co, Fe and Mn elements have chemical properties making these elements difficult to extract from an ISOL target. Nuclei lying below 68 Ni are not populated by fission of heavy elements and being neutron rich are not populated in fusion-evaporation of heavy ions. These isotopes are populated by high energy heavy ions fragmentation or Multi-nucleon transfer reaction around the Coulomb barrier using heavy ions. Such area remains a Terra incognita for such techniques.

3.5 Conclusions

A shape coexistence scenario emerges around N=40 in Ni, Co, Fe and Cr isotopes. An important experimental effort in the last five years have clarified the level schemes and spin/parity assignments. Very few relevant transition probabilities have been measured and no experimental data have been published on the direct determination of the shape (ie spectroscopic quadrupole moment) or possible constrains on the microscopical configuration of the intruder state. Presently, the direct proof for a 2p-2h proton excitation would a be milestone in the mass region.

In Co isotopes, unknown E2/M1 mixing prevent from a direct B(E2)/B(M1) comparison. In addition, based on the unique multi-polarity, the overlap between the B(E2) in Co and Ni is not as unambiguous as expected from the weak coupling model. The systematic in excitation energy of the $(1/2^{-})$ deformed state suggests an inversion beyond N=40 with the ground state. This has never been proven.

In ⁶⁸Ni isotope, one should remember that so far the E0 transition from the 0_3^+ was not observed and the spin assignment comes from the angular correlation $0 \rightarrow 2 \rightarrow 0$. The shape of this state is not confirmed by a spectroscopic quadrupole moment and no B(E2) connecting to a possible rotational 2⁺ state is measured. The safe Coulomb excitation of a high intense ⁶⁸Ni beam would be a milestone in the present discussion.

In the last section, an analysis of the different path to reach the 2p-2h proton state by β -decay and nucleon transfer is proposed. It shows that indeed, with its proposed configuration, these direct reactions cannot strongly populate such configuration. The consistency between proton, neutron transfer and β -decay is proven. The consistency between the 2 protons transfer from ¹⁴C and the delayed 168 keV line is not obvious. One could speculated that, instead, a third excited 0⁺ state, of neutron nature, similar to the one in ⁶⁶Ni, was populated.

Partie IV

Shape coexistence around N=Z=40

Chapter 4

Shape coexistence around N=Z=40

4.1 Introduction

The neutron deficient isotopes, close to the N=Z line, between Ge and Zr, are a long standing area of shape coexistence. Stable Ge isotopes have shown a competition between a spherical excited configuration and a deformed ground state. As these isotopes are stable, they have been studied by safe Coulomb excitation and spectroscopic quadrupole moment for the first 2^+ states were extracted. Low lying 0^{+m} states have been identified as evidence of shape coexistence. However, shape parameter of states built on top of this proposed shape isomers are not yet experimentally measured.

For heavier elements, from Se to Zr, Wood-Saxon calculations including a spin-orbit term [138, 139] and more elaborated mean field calculations using the Skyrme interaction [140, 141] or the Gogny interaction [142, 143, 144, 145, 146, 147] have predicted a shape coexistence between prolate and oblate configurations. Low-lying 0^{+m} states have been identified in 72,74,76 Se, 72,74,76 Kr and 80 Sr as first evidence. A specificity of the mass region is that several spectroscopic observations point to a strong mixing of the wave functions corresponding to the two configurations. In Kr isotopes, a perturbation of the ground state band moment of inertia at low spin was observed. Similarly, the reduced transition probabilities is showing a reduction of the collectivity at low spin as a result of the possible shape mixing (see figure 4.1). Combining the excitation energy of both the 2_1^+ and 4_1^+ states and the B(E2, $2_1^+ \rightarrow 0_1^+)$, the correlation between the $R_{42}=4_1^+/2_1^+$ excitation energy ratio and the B(E2)[W.u.].A⁻¹ shows that these N~Z nuclei are located far from the systematics of all other nuclei in the nuclear chart as shown in [148]. These off-systematics points are understood as the result of the mixing.

The perturbation of the 0^+ ground state and 0^{+m} isomeric state excitation energy with respect to the higher spins extrapolation was used to quantify for the first time the wave function mixing using the two levels mixing model [22]. In the case of the neutron deficient Kr isotopes, a mixing of 27% of the wave function is extracted for the ⁷⁶Kr ground state. This mixing reaches a maximum



Figure 4.1: Experimental $B(E2\downarrow)$ in the ground state band of 74,76 Kr from lifetime measurement.

of 50% in ⁷⁴Kr deduced from the maximum value of $\rho(E0)$ and minimum excitation energy of the 0^{+m} . In ⁷²Kr, the 0^{+m} excitation energy increases with a reduced $\rho^2(E0)$ leading to a mixing of 10% [22]. This parabolic trend suggests an inversion of shape from ⁷⁸Kr to ⁷²Kr with a crossing at ⁷⁴Kr where both configurations are almost degenerated [22]. So far, the evaluation of the sign of the deformation, i.e. prolate or oblate, was derived from comparison with theoretical models. For instance, using a comparison between measured B(E2; $2^+ \rightarrow 0^+$) and theoretical values, Iwasaki et al [149] proposed that the first 2^+ state in ⁷²Kr is compatible with an oblate shape and therefore suggesting a prolate shape for heavier isotopes.

All these experimental probes gave rise to *indirect* evidence of the shape coexistence. Laser spectroscopy is an experimental technique from which spectroscopic quadrupole moment in odd-mass nuclei ground state can be measured as well as for long-lived isomers. Unfortunately such systematic is rather scarce close to the N=Z line and does not involve the isotopes of interest as it will be more detailed in section 4.3.

4.2 Coulomb excitation experiment results

Direct evidence of the shape coexistence in this mass region came from the safe Coulomb excitation of radioactive beams. The Coulomb excitation cross section, via the second order effect of the reorientation [39, 40, 72], is sensitive to the E2 diagonal matrix element from which a spectroscopic quadrupole moment can be deduced. The very first safe multi-step coulomb excitation of a postaccelerated radioactive beam was performed for the neutron deficient Kr isotopes at the SPIRAL1 facility, GANIL in the framework of my PhD thesis work. Post-accelerated beams of ⁷⁴Kr and ⁷⁶Kr were delivered to the EXOGAM setup and bombarded a ²⁰⁸Pb target just below the Coulomb barrier. A charged particle detector covered scattering angles ensuring "safe" Coulomb excitation according to Cline's criteria [39]. As a result of my analysis, the first ever spectroscopic quadrupole moments in short lived excited states in unstable isotopes were measured [9]. This work represents the first direct experimental proof of the proposed shape coexistence scenario in neutron deficient Krypton isotopes, and the reorientation effect has been exploited for the first time with a radioactive ion beam. The details of the analysis and discussion can be found in [9]. To summarize the output of this rich set of E2 matrix elements both transitional (leading to B(E2)) and diagonal (leading to spectroscopic quadrupole moment) :

- 16(13) E2 transitional matrix elements have been measured in ⁷⁶Kr (⁷⁴Kr) respectively.
- The measurement has shown an inconsistent value of the 4_1^+ state lifetime in ⁷⁴Kr later confirmed and corrected by a dedicated plunger measurement [32]
- 5(5) E2 diagonal matrix elements have been measured in 76 Kr (74 Kr) respectively for the first time.
- The transition strengths from the Coulomb excitation measurement are in good agreement with the results of the lifetime measurement.
- Using the precise lifetimes as additional input in the analysis of the Coulomb excitation data enhanced the sensitivity to the reorientation effect and allowed extracting spectroscopic quadrupole moments for several excited states.
- The negative diagonal matrix elements of the states in the ground-state bands of both isotopes prove their prolate character
- The excited 2_2^+ and 2_3^+ states are interpreted as γ -vibrational and oblate rotational states, respectively.
- However, a clear classification of the states is difficult due to the mixing of prolate and oblate configurations, on one hand, and K = 0 and K = 2 configurations, on the other hand.
- The matrix elements were interpreted in a phenomenological two-band mixing model. The results confirm the earlier finding of maximum mixing between prolate and oblate configurations in the wave functions of the 0⁺ states in ⁷⁴Kr. In ⁷⁶Kr, the mixing of the ground state is 31(4)% whereas it was measured at 27(1)% from the E0 spectroscopy. In ⁷⁴Kr, the mixing is found equal (52%) in both experiment.

- The purity of the wave functions increases rapidly with spin, explaining the increase in the transition strength and at the same time the very strong inter-band transitions between the different 2⁺ and 0⁺ states (see figure 4.1).
- The two level mixing model fails to describe consistently the band mixing in 76 Kr, probably due to the strong coupling with the presumed γ -vibrational band.
- The quadrupole sum rule formalism was applied to derive shape parameters for the 0⁺ states.
- The first rotational invariant, Q^2 (similar to $|\beta_2|$), is found large for both states in both isotopes confirming their deformed character
- The second rotational invariant, $cos3\delta$ (similar to γ) shows that the ^{76,74}Kr ground state are axially prolate. The 0_2^+ state in ⁷⁶Kr is oblate with a large triaxial contribution. The second 0^+ state in ⁷⁴Kr reaches a maximum of triaxility.
- The transition strengths and quadrupole moments were compared to theoretical calculations going beyond the static mean-field approach using the Gogny D1S interaction and the Skyrme SLy6 interaction. The present SLy6 interaction is limited to axial symmetry. The comparison emphasize the importance of including the triaxial degree of freedom to describe the shape coexistence in the light krypton isotopes.

4.3 Q_s systematics

The diagonal E2 matrix elements extracted from the Coulomb excitation analysis can be translated into spectroscopic quadrupole moments and compared to the ground states quadrupole moment in odd-mass neighbouring nuclei. Data used for this comparison are compiled from [70, 46] and completed by quadrupole moments of the 2^+_1 states in 70,72 Se[150, 149, 151] and 72 Ge [152].

In figure 4.2,the compilation of the absolute deformation amplitude ($|\beta_2|$) is restricted to Z>31 and N<50. First, it is worth mentioning that such measurement did not yet reach the N=Z line above the Ni chain and are far from ¹⁰⁰Sn.

From the Z=32, a clear onset of deformation is observed between Ge and Se isotopes, i.e. when the proton $f_{5/2}$ is populated and with at least four neutrons holes in the $g_{9/2}$. From this systematics in neutron number, the shell closure at nucleon number 40 does not present a stabilizing effect toward spherical shape and the maximum of deformation is reached at Z \simeq N \simeq 38-39.

In figure 4.3, the algebraic deformation is deduced (β_2). The blue (green) squares indicate a prolate (oblate) deformations respectively. The first matrix shows the deformation for the ground states in odd-nucleon isotopes and for the 2_1^+ state in even-even isotopes. Beyond Se, deformation are large and prolate. Approaching the N=Z line, a prolate to oblate transition occurs in ⁷⁰Se. No other shape inversion as a function of the neutron number was proven so far. The second panel highlights the shape coexistence scenario. This matrix shows the same algebraic deformation (β_2) measured for isomers, which are living long-enough to allow a laser spectroscopy, and for $2_{2,3}^+$ states built above a low-lying 0_2^+ from safe Coulomb excitation measurements. The 2_2^+ state is not chosen by default. Indeed, in this mass region, K=2 γ -band are frequent at low excitation energy. Therefore, for the compilation, the 2_2^+ or 2_3^+ state is chosen from the largest B(E2) value to the



Figure 4.2: Absolute deformation amplitude $(|\beta_2|)$ in the (N,Z) plan from measured spectroscopic quadrupole moments. The spherical Nilsson orbitals are indicated, the blue squares size are proportional to the absolute deformation amplitude $(|\beta_2|)$ for ground state in odd-nucleon isotopes and for the 2^+_1 state for even-even isotopes.

 0_2^+ state.

The comparison between the two matrices illustrates the shape coexistence in a given isotope. In Ge isotopes, the ground state presents a very small deformation and the 2_3^+ state has a large one illustrating the shape coexistence between a spherical ground state and a deformed excited configuration. So far, the shape change in Ge isotopes was not proven. In light odd-mass Kr, the Q_s in long-lived isomers are not established. Large systematics also exists for the Rb isotopes (Z=37). The ground states have large prolate deformation. The isomers have an oblate or prolate character. Beyond Rb, available data are almost non-existing.

4.4 Probing the deformation from the first excited states in even-even isotopes

In ⁶⁸Se, the systematics of the momentum of inertia suggests that the ground state band is oblate as the band built on top of the 2_2^+ at 1594 keV has a prolate character (see figure 4.4) [153]. The B(E2, $2_1^+ \rightarrow 0_1^+$) was first measured during my PhD [154] and later with more precision by [155]. The value is found compatible with theoretical prediction of an oblate, γ -unstable, deformed state. The spectroscopic quadrupole moment remains un-measured.

In 70 Se, the systematics of the momentum of inertia suggests that the ground state band at low



Figure 4.3: Algebraic deformation amplitude (β_2) in the (N,Z) plan from measured spectroscopic quadrupole moments. *Top*: for ground or 2_1^+ states. *Bottom*: for isomers or $2_{2,3}^+$ states

spin is oblate and turn to be prolate at higher spin [156]. Combined Safe Coulomb excitation using post-accelerated beams by A. Hurst [150] and high precision lifetime measurement by J. Ljungvall [149] have proven the oblate character of the 2_1^+ state. Finally, J. Henderson performed the safe Coulomb excitation of ⁷²Se [151]. A negative spectroscopic quadrupole moment for the 2_1^+ state was measured, supporting its prolate character [151]. Previous measurements in stable Se isotopes by [157, 158] have also proved a prolate 2_1^+ state. There is therefore a shape change for the 2_1^+ state.

Recently, the first spectroscopy of ⁷⁰Kr was performed and suggests, by comparison with the oretical calculations, a shape coexistence scenario [159]. In ⁷²Kr, using a comparison between measured B(E2; $2^+ \rightarrow 0^+$) and theoretical values, Iwasaki et al [149] proposed that the first 2^+ state in ⁷²Kr is compatible with an oblate shape. The safe Coulomb excitation of ^{74,76}Kr provides the first evidence for a shape coexistence scenario with a prolate ground state band and an oblate


Figure 4.4: Proposed shape assignment in neutron deficient Se isotopes

excited configuration with large mixing of the wave functions. Finally, the safe Coulomb excitation of 78 Kr was performed in [160]. The prolate character of the 2^+_1 is established as for the 2^+_2 and 2^+_3 state triaxial prolate and oblate deformation are determined.

Experimental data are even more scarce in Sr isotopes. Its chemical properties makes Sr element very difficult to extract from an ISOL target. In addition, the N \sim Z ⁷⁶Sr isotopes becomes very exotic and produced with very low cross section. Neutron deficient Sr isotopes have a large deformation as deduced from the observation of rather regular rotational ground states band and corresponding large B(E2). In [148], A. Lemasson et al., have proposed a low mixing between the configurations from the measured B(E2, 2⁺ \rightarrow 0⁺) placed in the systematics of the correlation between the R₄₂=4⁺₁/2⁺₁ excitation energy ratio and the B(E2)[W.u.].A⁻¹. Even if the experimental point follows the systematics of the Kr isotopes, assuming that the R₄₂ is perturbed by the mixing, a unique feature of ⁷⁶Sr arises. Indeed, with a typical mixing strength of 0.2 MeV and a 0⁺₂ excitation energy at around 1 MeV, a mixing of 5% is extracted. This apparent contradiction between weak mixing and perturbation of the collectivity calls for more detailed spectroscopic data as the search for the 0⁺₂ state and related electromagnetic strength.

The systematics of the 2_1^+ excitation energy and B(E2) for N=Z nuclei are presented in figure 4.5 extracted from [148]. The systematics show a clear onset of collectivity between ⁷²Kr and ⁷⁶Sr at the N=Z line. It is proposed, at the mean-field level, that an increase of axiallity explains the onset of collectivity [155]. A similar systematics of spectroscopic quadrupole moment and quadrupole invariant would confirm the proposed symmetry effect.



Figure 4.5: $B(E2;2_1^+ \rightarrow 0_{\pm 1})$ systematics at the N=Z line. Figure from [148].

4.5 Probing the role of the $g_{9/2}$ from laser spectroscopy

One could expect that this increase of collectivity as a function of the proton number could be related to an increase of the $g_{9/2}$ occupancy acting as an intruder. Its role can be evaluated from the experimental point of view from the systematics of the spin and parity of the ground states. In figure 4.6, the spin and parity measured for the ground states of odd-masses by laser spectroscopy is presented from Se to Sr. The most probable Nilsson orbital is indicated with the spherical orbit parentage. The deformation character is extracted from laser spectroscopy or deduced from neighbouring even-even nuclei using Coulomb excitation experiment.

In ⁶⁹Se (Z=34, N=35), an oblate (from ⁷⁰Se shape) $1/2^{-}$ spin/parity is assigned. It is proposed that the configuration is dominated by the deformed $1f_{5/2}$ orbit with the 1/2[310] oblate orbital. In ⁷³Se, the $9/2^{+}$ assignment suggests the 9/2[404] orbits, i.e. an oblate deformation, with a $g_{9/2}$ orbital acting as an intruder. From Coulomb excitation measurement, ⁷²Se has a prolate character and the shape inversion to oblate occurs for ⁷⁰Se. Therefore, one could assume that ⁷³Se has a prolate deformation and the local oblate character is surprising. At N=41, the $g_{9/2}$ is naturally populated and the $5/2^{+}$ spin/parity is compatible with the deformed 5/2[422] orbital. In Br isotopes, with a spin parity $1/2^{-}$ and $3/2^{-}$ assigned to the 1/2[310] and 3/2[301] deformed $f_{5/2}$ orbital, the proton $g_{9/2}$ is not yet involved. In Kr isotopes, ⁷³Kr with a spin/parity $3/2^{-}$ points again to the $f_{5/2}$ orbital, hence suggesting an oblate shape. Beyond, at N=39 and 41 with a proposed prolate shape as for N=38 and N=40, the $5/2^{+}$ spin/parity supports the occupation of the prolate deformed $g_{9/2}$ similarly to the Se and Kr isotopes, the ground state suggests the occupation of the neutron deformed $g_{9/2}$. Surprisingly, ⁷⁹Sr would correspond to a ground state involving the $\nu f_{5/2}$ orbital.

From the present systematics, the odd-proton isotopes do not point for the occupation of the $\pi g_{9/2}$ as in intruder, at least until Z=37, and underline the role of the deformed $\pi f_{5/2}$ orbitals. In even-proton isotopes, similarly, until N=37, the deformed neutron $f_{5/2}$ is favoured. From N=39,



Figure 4.6: Top : Systematics of spin/parity assignment from laser spectroscopy in N~Z nuclei. The shape mentioned next to the configuration is determined from measured Q_s .

the neutron $g_{9/2}$ orbital appears as an intruder. But its role in the increase of collectivity when the proton number increases is not established from this systematics. For the Se and Kr isotopic chains, the transition from prolate to oblate deformation when approaching the N=Z line seems to be correlated to the change from the neutron $g_{9/2}$ to $f_{5/2}$ occupation. ⁷⁷Sr and ⁷⁹Sr remain prolate with however a change of the neutron orbit. The systematics underline that the shape coexistence, before N=Z, from Se to Sr, seems to be dominated by the competition between the deformed gaps at 34, 36 and 38 in the pf shells.

4.6 Short overview of the theoretical approaches

4.6.1 Mean Field calculations in Se, Kr, Sr and Zr isotopes

In this section, spectroscopic data such as B(E2) and excitation energy of the first excited states are compared to mean-field calculations using the Gogny D1S or Skyrme SLy6 interactions. This formalism is expected to perform well in these deformed nuclei having complex mixing of configurations.



Figure 4.7: Calculated shape evolution in Se isotopes. Picture from [161].

The Se isotopes have been investigated using the Gogny D1S interaction in the mean-field framework in [161]. The known B(E2) are in good agreement with theoretical calculations for the first excited states. These calculations predict a shape change for both 70,72 Se from oblate to prolate at spin 4 and 6 respectively as 68 Se remains oblate as illustrated in figure 4.7. Unfortunately the spin evolution was not probed so far experimentally. Low lying 0_2^+ states are calculated at high excitation energy (≥ 1 MeV). All three 68,70,72 Se are predicted oblate in the 2_1^+ state by the Gogny D1S interaction as a shape change occurs experimentally between 72 Se and 70 Se from prolate to oblate deformation.

In Kr isotopes, the experimental B(E2) and Q_s were then compared to mean field calculations using the Gogny D1S or Skyrme SLy6 interactions in [9]. In the publication, the later is restricted to axial symmetry. Along the rotational prolate and oblate bands, both calculations reproduce the trend as a function of angular momentum as illustrated in figure 4.8. The Skyrme calculation slightly overestimates the collectivity while the Gogny calculations reproduce well the collectivity as a results of the restriction to axial symmetry in the Skyrme calculation. We proposed that the large overlap between K=0 bands and K=2 γ -band, reflected by large inter-bands B(E2), leads to triaxility and reduces in-band B(E2) and Q_s .

In figure 4.9, the experimental level scheme is compared to the Skyrme calculations. As they are restricted to axial symmetry, no K=2 states are calculated. The oblate configuration is predicted to be the ground state by opposition to the experimental conclusions. Finally, inter-band B(E2) are underestimated. Using the Gogny D1S interaction, the agreement is remarkable for excitation energy and matrix elements (see figure 4.10). The K=0 prolate band is predicted to the ground state band as the 0_2^+ and 2_3^+ are part of the oblate rotational band. A K=2 γ band is predicted with a 2_2^+ band head located below the 2_3^+ as experimentally proposed. A strong mixing of K=0 and K=2 components for the 2_2^+ and 2_3^+ states is obtained, as strong indication of triaxility. E0 transition strengths, $\rho^2(E0)$, are also compared (see figure 4.11). Calculated values are far from



Figure 4.8: Experimental B(E2, \downarrow) and Q_s compared to both Gogny D1S or Skyrme SLy6 calculations.

the experimental one. The reason is not yet established.

In [162], potential energy surfaces were published in details following the convincing comparison with the experimental spectroscopic data. As it can be seen, the potential energy surfaces using the Gogny GCM+GOA approach show two minima for prolate and oblate deformation. The $J^{\pi}=0^{+}_{1}$ projection shows a prolate minima in ⁷⁶Kr with an increased spread into the γ triaxial degree of freedom in ⁷⁴Kr and more axial oblate ground state in ⁷²Kr. It was therefore proposed a shape inversion at ⁷⁴Kr with a maximum triaxiality as suggested by the experimental data even if the oblate character of the ground state of ⁷²Kr is not yet firmly established. In the calculations, increasing the angular momentum leads to a narrow minimum in the wave function distribution with less γ -spread and increased axiality. This increased of axiality in the wave function is correlated to the increase of the measured B(E2) therefore reflecting a weaker mixing as proposed from the experimental data. One can finally notice that there is a shape change between spin 2 and 6 in ⁷²Kr which remains to be proven experimentally. Triaxility seems to be the key ingredient to reproduce the shape coexistence in Kr isotopes. Later on, the Skyrme calculations were developed. Meanfield states generated by triaxial quadrupole constraints that are projected on particle number and angular momentum and mixed by the generator coordinate method on the quadrupole moment were



Figure 4.9: Level Scheme comparison between experimental data and Skyrme SLy6 calculations



Figure 4.10: Level Scheme comparison between experimental data and Gogny D1S calculations

proposed in [163]. This method is equivalent to a seven-dimensional GCM calculations, mixing all five degrees of freedom of the quadrupole operator and the gauge angles for protons and neutrons. The deduced level schemes cancel the previous disagreement. When triaxiality is not authorized, the same results than the axial Skyrme are obtained suggesting that triaxiality seems to be the key to describe prolate-oblate shape coexistence in this region.

Large systematics for the Sr isotopes are not yet available due to the lack of experimental data. The B(E2) and excitation energy of the first 2^+ state are well reproduced by the calculations using the Gogny D1S interaction.

Along the N=Z, the calculations reproduce well the onset of collectivity between 68 Se and 76 Sr as shown in figure 4.5. The corresponding potential energy surfaces are also systematically calcu-



Figure 4.11: Theoretical (Gogny) and experimental E0 systematics. Figure from [162].



Figure 4.12: HFB potential energy surface for N=Z isotopes. Figure from [162].

lated for N=Z nuclei from ⁶⁰Zn to ⁷⁶Sr in the (β, γ) plan in [162]. Figure 4.12 shows an increased of axiallity toward the heaviest masses with a very triaxial and γ -soft minima for ⁶⁴Ge, a γ -soft oblate minima in ⁶⁸Se, more axial in ⁷²Kr as the ground state in ⁷⁶Sr has a large and axial prolate deformation. This increase of axiality is associated to the increase of B(E2) experimentally observed and emphasizes the role of the degree of freedom in the mass region [148].

4.6.2 Shell Model calculations in Se, Kr, Sr and Zr isotopes

Mean field calculations have shown a remarkable agreement with experimental data. However, they do not provide a microscopical description. One could wonder for instance, if this clear onset of collectivity associated to axiality between Se and Sr is related to an increase in occupancy of a specific orbital for both proton and neutron. Shell model calculations can provide a more fundamental basis or quantitative influence of a specific orbital or valence space. However, there are severe limitations due to the large valence space needed for such calculations. Monte-Carlo Shell Model Calculations (MCSMC) could in principle be applied like in mass A=100 [8, 58], in the Ni chain [6] or in the light Hg isotopes [7]. However, this has never been done for N=Z nuclei.



Figure 4.13: Theoretical $g_{9/2}$ occupancy systematics for Kr, Sr, and Zr isotopes. Figure from [164]

Few exact shell model calculations have been performed in truncated valence space and compared to experimental spectroscopic data. Shell model calculations have been performed by K. Langanke et al. with a complete 0f1p0g1d2s model space for Kr, Sr and Zr isotopes [164]. Only experimental B(E2; $2_1^+ \rightarrow 0_1^+$) are compared with the calculations and do not reproduce the onset of collectivity between ⁷²Kr and ⁷⁶Sr. In figure 4.13 taken from [164], the $g_{9/2}$ occupancy is shown for proton and neutron separately. N=Z nuclei are highlighted with brown squares. The increase of collectivity, in the calculations, from Kr to Zr, is correlated to an increase of the $g_{9/2}$ occupancy for both proton and neutron. In the Kr chain, the neutron $g_{9/2}$ occupancy increases with mass as expected with a calculated shape change between 72 Kr and 76 Kr. The increased number of neutron in the $g_{9/2}$ favours the prolate deformation.

Later, M. Hasegawa et al. have performed calculations in a truncated $f_{5/2}p_{1/2}g_{9/2}d_{5/2}$ valence space (ie no active $p_{3/2}$) [165]. A strong p-n correlation is proposed since they occupy similar orbitals. Calculations are only available for N=Z nuclei. For the ground state bands, the level schemes are rather well reproduced for ⁶⁸Se and ⁷²Kr but the calculations underestimate the onset of collectivity in ⁷⁶Sr.



Figure 4.14: Theoretical N=Z isotopes orbits occupancies from [165]

The $f_{5/2}p_{1/2}g_{9/2}d_{5/2}$ occupancies, assuming that there are equal for proton and neutron, are shown for ⁶⁸Se, ⁷²Kr and ⁷⁶Sr in figure 4.14. It is known experimentally that ⁶⁸Se has a lower collectivity [154, 155]. In the calculations for ⁶⁸Se, the $g_{9/2}d_{5/2}$ occupancy is very low (~ 0.2) for the $0_{1,2}^+$ and 2_1^+ states, ie equivalent for both ground states band and excited configuration. One could speculate that this similarity favours large mixing and consequently lower collectivity. For ⁷²Kr and ⁷⁶Sr, the $f_{5/2}$ occupancy increases as expected. The $g_{9/2}d_{5/2}$ occupancy increases between ⁶⁸Se and (⁷²Kr,⁷⁶Sr) but remains still low (~ 1 particle) for the ground state band and ~ 0.5 particle for the 0_2^+ . Contrary to ⁶⁸Se, there is a clear difference between the ground states band and the excited configuration which could reduce the mixing inducing a more clear shape coexistence scenario with two 0⁺ states. The increase by a factor 2 of the experimental B(E2)'s between ⁶⁸Se and ⁷²Kr is reproduced supporting the contribution of the $g_{9/2}$ orbital. Experimentally, there is a second step by a factor 2 between ⁷²Kr and ⁷⁶Sr in B(E2) which is also reproduced in the calculation associated to a small increase of the $g_{9/2}$ occupation (~ 0.5 particle).

More recent shell model calculations using the JUN45 and the LNPS interactions have been performed by B. Bounthong in [166]. In the calculations, the B(E2; $2_1^+ \rightarrow 0_1^+$) increases by only 20% between ⁷²Kr and ⁷⁶Sr as the $g_{9/2}$ occupation remains almost identical, supporting again the role of this orbital in the collectivity of the N=Z nuclei.

In summary, some shell model calculations have investigated the 2^+ state in N=Z nuclei and in the Kr, Sr and Zr chain leading to incomplete conclusions. The Kr chain remains the case where lot of experimental informations are known (energies, B(E2)'s, g-factors, Qs) and should be an ideal case for SM-like calculations to investigate the microscopical origin of the shape evolution in this mass region. Mean-field calculations work well but only highlight the role of the triaxial degree of freedom. Shell model calculations which should provide a more microscopical description are scare and very limited in their comparison with experimental data but highlight the role of the $g_{9/2}$ orbital.

4.7 Conclusions

For the future, several systematics are needed :

- The Kr case remains unique a decade after the publication of the present results. Postaccelerated radioactive beam of Se, Sr and possibly Zr are of great interest. Especially ^{74–70}Se, ⁷²Kr and ^{76–80}Sr beams must be Coulomb excited to provide similar set of E2 matrix element as for the Kr case. These will provide a systematic of the shape evolution and mixing along the N=Z line.
- 0_2^+ states remain unknown in $^{68-70}$ Se, 76,78 Sr.
- Yrast band's B(E2) have been measured at very low spin up to ⁷²Kr, ⁶⁸Se, ⁷⁶Sr.
- no non-yrast E2 matrix elements is known in these isotopes.
- There is a lack of spectroscopic quadrupole moment in the ground state beyond Sr and long-lived isomeric states (particularly $9/2^{+m}$ state) in odd-masses by laser spectroscopy.

The electromagnetic probe provides a shape description and possible link between them. However, it does not provide any direct experimental contribution to the microscopical description in particular the role of the $g_{9/2}$ orbit. Direct nucleon transfer reactions are the standard technique to identify effective single particle energy and occupancy number. Direct reaction such as (d,p) or $(d,^{3}He)$ to probe neutron and proton orbits were never attempted in these isotopes. Furthermore, similarly to the A~100 region (see chapter II), two nucleon transfer to probe pair excitation origin across the gap 40 are not yet reachable. Two proton transfer from Mo or Zr isotopes to probe the multi pair excitation across Z=40 would have a significant impact in the shape coexistence scenario description. However, the shape identification (prolate or oblate) in the daughter nuclei must be identified first by safe Coulomb excitation. Two neutrons transfer from ⁷⁴Kr to ⁷²Kr would also give more insight in the shape inversion. Single nucleon transfer would be the cornerstone of these studies to tentatively match the shape inversion and mixing with a microscopical description. Partie V

Conclusion

Conclusion

The study of the shape coexistence at the 40 sub-shell closure was revolutionized in the last decade thanks to the availability of new instruments, new accelerators, the re-born of relevant experimental techniques and the Monte-Carlo Shell Model Calculations. Shape coexistence remains a spectacular effect in nuclear structure involving strong proton-neutron quadrupole correlation and pairing against the sub-shells gap. The cross comparison between experimental data from in-beam data and laser spectroscopy brings the pieces for a global understanding. Comparison with theoretical calculations remains today partial. Around the 40 sub-shell closure, large set of experimental data can be used as benchmark for large scale comparisons.

In this manuscript, I have reviewed the state-of-the-art of the shape coexistence phenomena involving the sub-shell closure 40 in three mass regions with N=40 around ⁶⁸Ni, with Z~40 in neutron rich isotopes with A~100 and in self-conjugated nuclei with N~Z~40. The two later have benefited in the recent years of experimental results using post-accelerated radioactive ions beam for Coulomb excitation and single nucleon transfer. The Coulomb excitation technique gives access to the spectroscopic quadrupole moment. This is a major step in the description of the shape coexistence in these mass region and reinforces the relevance of the comparison with the most advanced theoretical models. In this manuscript, a particular care was taken in bringing together the different probes such as spin/parity and spectroscopic quadrupole moment from laser spectroscopy in the ground state and the isomers, Coulomb excitation, lifetime measurements, nucleon transfer, E0 spectroscopy and β -decay. The three mass regions have very different level of description.

It is clear that the A~100 region is characterized by a large amount of experimental data with all techniques showing a clear coherence. Nothing unexpected arise so far. The MCSMC have described for the first time the microscopical mechanism. However, few questions remain. The N=59 isotone evolution as a function of the proton number from Ru to Kr would highlight the role of pairing and proton occupancy. The microscopical description of the Kr isotopes should explain why the offset of deformation is delayed and smoother. Even if the agreement between the MCSMC and some experimental data is remarkable, a global comparison with all spectroscopy data would push forward the constrains to the models. The microscopical description reproduces well the data but this particle-hole scheme coupled to intruder orbital is not experimentally verified. Contrary to the N~Z nuclei, the agreement between the mean-field calculations and the experimental data is not as good.

In the N~Z nuclei, large set of experimental data are available for excited states until the Kr isotopes. Clearly, there is a lack of experimental data in the elements from Sr and for all elements in the ground state using laser spectroscopy to draw a global experimental picture. In addition,

these nuclei have never been investigated by direct reaction. The agreement between spectroscopic data and the mean-field calculation is remarkable. Microscopical description using conventional or MCSMC have not reached the level of accuracy obtained in the $A \sim 100$ region. Further studies using shell models calculations are the next millestone.

Around ⁶⁸Ni, the level scheme have been recently clarified. Shell model calculations using conventional approached (LNPS) or Monte-Carlo give a remarkable agreement with the proposed level scheme and first B(E2). The lack of experimental data such spectroscopic quadrupole moments in excited or ground states limits the comparison. The absence of data using laser spectroscopy is evidenced. Similarly to the A~100 region, the proposed microscopical description is not experimentally verified and systematics studies by direct reaction are needed.



Figure 4.15: Experimentally established area of shape coexistence. Figure from [1]

In figure 4.15, the archipelago of experimentally proven shape coexistence area is shown. In many aspects, the shape coexistence in $A \sim 100$ and around ⁶⁸Ni follow the mechanism proposed by K. Heyde and collaborators without any surprises. All new experimental data are successfully compared to state-to-the-art calculations which reinforce the statements of [1]. So far, along the nuclear chart, the shape coexistence area can be characterized by this mechanism and experimen-

tally proven shape coexistence close to closed shells which do not follow this picture are scarce (see for instance ⁸⁰Ge [15]). However, the N \sim Z nuclei around mass A=80 should draw a particular attention. Indeed, as discussed in this manuscript, they do not really follow this description and should attract the future experimental and theoretical works.

This universal mechanism was never used to predict new area of shape coexistence. Global calculations in the mean-field approach are available and can be used to predict the next area of shape coexistence. In [167], P. Möller proposed a global survey of the phenomena. In figure 4.16 from [167], the number of minima in the potential energy surface is shown for a given nucleus as soon as the depth is greater than 200 keV and its excitation energy lower than 2 MeV. Beyond Z=20, areas with a second minimum overlap very well the figure 4.15. New areas of shape coexistence are predicted in very neutron rich Sn isotopes around N=100 and in super heavy elements around Z=100.



Figure 4.16: The shape coexistence archipelo: calculated number of minima in the potential energy surface of even-even nuclei. Figure from [167].

Similar comparison can be done using the Gogny D1S interaction using the published calculations from [168]. In figure 4.17, stable nuclei are represented with large blue squares. Nuclei indicated with a brown square have a second 0^+ state with an excitation energy below 1.0 MeV and a degree of triaxiality lower than $\gamma = 30$ degrees. Below Z=28, 0^+_2 states are calculated close to 2 MeV. From Ni isotopes, the calculations overlap very well the figure 4.15. Similarly, next area of shape coexistence in the vicinity of the closed Z=50 and Z=82 proton shells are predicted for very neutron rich isotopes. In the Sn isotopes, 0_2^+ at about 0.9 MeV excitation energy are expected in $^{146-150}$ Sn. In Pb isotopes, shape isomer at 0.5 MeV excitation energy are predicted in $^{224-230}$ Pb. Finally, several areas are anticipated in the super heavy elements in very neutron rich Fm-No isotopes. In the scheme proposed by K. Heyde, the Sn and Pb isotopes can be anticipated. Indeed, the N=100 shell closure lies between the $1h_{9/2}$ and $2f_{5/2}$ neutron orbital. With proton promoted from the $1g_{9/2}$ to $1g_{7/2}$, the p-n interaction is similar to the mass A~100 region.

One can also notice than both calculations do not predict an area of shape coexistence near 78 Ni as proposed in 80 Ge [15].



Figure 4.17: The shape coexistence archipelo: calculated low lying 0^+ states in even-even nuclei. Figure from [168].

So far the shape coexistence in atomic nuclei was theoretically investigated by effective theories. In a longer perspectives, with the development of Ab - Initio calculations, one could wonder if the phenomenom and its present effective description emerge naturally from the *first - principles* ?

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