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0_2^+ band in ^{102}Ru and the evolution of nuclear deformation in Ru isotopes

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Two measurements of γ rays from the slow-neutron capture reaction on a ^{101}Ru target were performed at the PF1B cold-neutron facility and the DuMond GAMma Spectrometer GAMS5 of ILL Grenoble, to study in detail excitations in the transitional nucleus ^{102}Ru. A band structure on top of the 0_2^+ level has been identified and its nature is discussed. Mixing between the 0_1^+ and 0_2^+ levels in Ru isotopes with $50 < N < 66$ neutrons and its role in the development of deformation in the ground state of the Ru nuclei have been studied.

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The emergence of collective effects in complex systems is of general interest in physics, in particular when observed in systems governed by laws of quantum mechanics, like atomic nuclei. In certain regions of the nuclear chart rapid changes of nuclear deformation are observed when the number of nucleons changes just by a few percent. Such pronounced effects offer deeper insight into the appearance of the nuclear deformation.

The mechanism of the sudden onset of deformation at $N \approx 60$ neutrons in mass $A \sim 100$ nuclei has been studied in numerous works. It was first proposed [1,2] that the effect is due to the proton-neutron interaction between the spin-orbit partner (SOP) orbitals, $\nu g_{7/2}$ and $\pi g_{9/2}$, causing a gradual increase of the deformation of the initially spherical ground state when neutrons are added to the $\nu g_{7/2}$ orbital. Although likely, this process alone could not reproduce the scale and the rate of the observed deformation change. Therefore, it has been proposed that the sudden deformation change is due to a lowering of a deformed structure based on the third 0^+ level, which becomes the deformed ground state in nuclei with $N \geqslant 60$ [3,4]. However, such deformed structures on top of the 0_3^+ level were not confirmed [5]. Instead, weakly deformed bands based on the *second* 0^+ level have been reported in Sr and Zr isotopes, with the deformation growing with the neutron number [6]. Further deformation increase, at $N \geqslant 60$, has been attributed to the population of the deformation-driving $\nu h_{11/2}$ intruder [7–9]. It has been proposed that this process is reinforced by vacating, at the same time, the spherical-driving $\nu 9/2[404]$ extruder orbital [10,11]. The subshell closures at $Z = 40$ and $N = 56$ most likely enhance the sudden nature of this deformation change [12].

It is interesting to ask about the nature of the weakly deformed bands at $N = 58$ on top of the 0_2^+ level, which are the "seeds" for the deformed bands at $N > 58$. The evolution of such bands should be seen more clearly away from the $Z = 40$ closure, which preserves the noncollective structures up to $N = 56$. Furthermore, it is expected that at $Z \geqslant 42$ the $\nu 9/2[404]$ extruder departs from the Fermi level and does not contribute to the deformation change. Therefore, Ru isotopes are good candidates to study the evolution of a collective structure based on the 0_2^+ level.

With excitations suggesting both spherical and deformed configurations, ^{102}Ru marks the point on the $N = 58$ and $Z = 44$ lines, which separates spherical from deformed nuclei [13]. A good knowledge of the structure of this transitional nucleus is essential for understanding the evolution of deformation in the $A \sim 100$ region. However, the nature of the crucial cascade on top of the 0_2^+ level in ^{102}Ru is still not clear [13]. In this work we report on experimental studies of nonyrast excitations in ^{102}Ru, performed in order to determine the properties of this cascade as well as its role in the development of nuclear deformation.

With spin $I^\pi = 5/2^+$ of the ground state in ^{101}Ru, the decay of the neutron-capture level in ^{102}Ru at 9219.7 keV has sufficient spin and energy to populate levels with spins from $I = 0$ to $I = 6$. This allows a search for bands in ^{102}Ru. In this work excited levels in ^{102}Ru were populated in the ^{101}Ru$(n,\gamma)^{102}$Ru reaction at the PF1B cold-neutron facility of the Institut Laue-Langevin. A neutron beam, collimated to about 1 cm in diameter, irradiated a 4-mg target of ^{101}Ru, isotopically enriched to 99%. The γ radiation from the reaction has been measured using 8 Ge detectors of about 60% relative efficiency each, placed in a plane perpendicular to the neutron-beam direction. The angle between two neighboring detectors was 45 deg, which has enabled angular correlation measurement at three angles of 90, 135, and 180 deg. Details of the experimental setup are given in Ref. [14].

About 2×10^9 *triggerless* events, consisting of a γ energy and the time of its registration (by a 40-MHz clock), have been collected. Out of this data we sorted 4×10^8 double-γ and 5×10^6 triple-γ coincidence events, applying a 600-ns time window to define a coincidence event. Various two- and three-dimensional histograms were sorted and used to build the level scheme of ^{102}Ru.

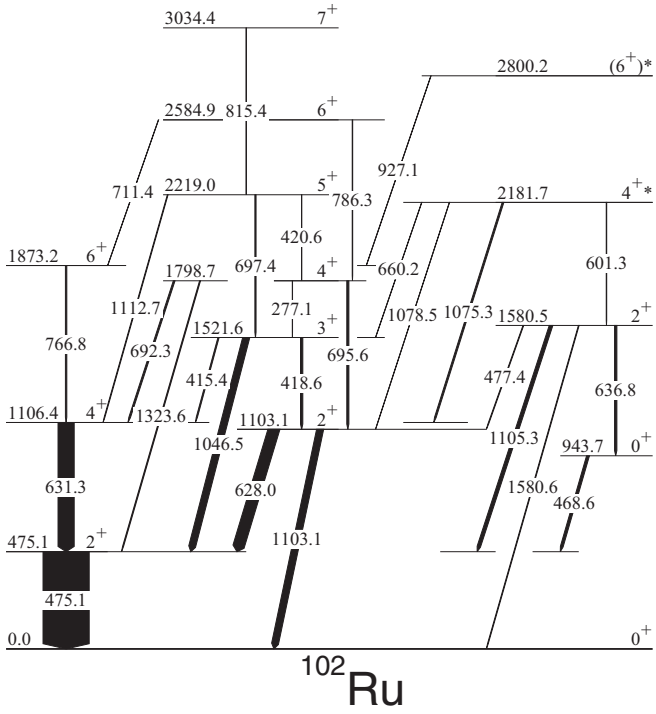

FIG. 1. Partial level scheme of ^{102}Ru as observed in the present work. New levels are marked with an asterisk. More accurate level and transition energies are given in Table I.

We have identified and placed in the decay scheme of ^{102}Ru more than 10^3 γ transitions. A full report on these results will be given elsewhere [15] while in this paper we present the data relevant to the discussion of the cascade on top of the 0_2^+ level. A partial level scheme of ^{102}Ru as observed in the present work is shown in Fig. 1, and the properties of the γ lines are collected in Tables I and II.

TABLE I. Energies and intensities γ transitions in ^{102}Ru, as measured in the present work in the $^{101}\mathrm{Ru}+n_{th}$ reaction, using the Ge array.[a] Uncertainties of γ intensity include a 3% systematic error due to the efficiency calibration.

E_γ (keV)	I_γ (rel.)	E^i_{exc} (keV)	E_γ (keV)	I_γ (rel.)	E^i_{exc} (keV)
277.1(1)	0.07(1)	1798.68(2)	697.41(2)	1.7(1)	2219.03(4)
415.36(5)	0.71(7)	1521.62(3)	711.4(1)	0.04(1)	2584.90(5)
418.60(3)	3.8(2)	1521.62(3)	766.84(3)	1.6(1)	1873.21(3)
420.6(2)	0.03(1)	2219.03(4)	786.27(4)	0.42(5)	2584.90(5)
468.58(1)	4.8(2)	943.68(2)	815.4(2)	0.02(1)	3034.4(3)
475.10(1)	100(3)	475.10(1)	927.05(8)	0.11(1)	2800.3(1)
477.41(6)[a]	1.3(4)	1580.48(4)	1046.48(2)	10.4(5)	1521.62(3)
601.30(5)	0.27(2)	2181.74(4)	1075.34(3)	1.9(1)	2181.74(4)
627.99(1)	16.9(6)	1103.09(2)	1078.51(9)	0.08(1)	2181.74(4)
631.27(1)	28.9(7)	1106.37(2)	1103.09(2)	10.0(5)	1103.09(2)
636.84(1)	3.3(1)	1580.48(4)	1105.31(3)	5.6(4)	1580.48(4)
660.15(9)	0.15(2)	2181.74(4)	1112.74(4)	0.63(9)	2219.03(4)
692.29(3)	2.4(2)	1798.68(2)	1323.60(3)	1.1(21)	1798.68(2)
695.60(2)	3.6(2)	1798.68(3)	1580.57(3)	0.9(1)	1580.48(4)

[a]Energy taken from the GAMS5 measurement.

TABLE II. Angular correlation coefficients for γ-γ cascades in ^{102}Ru populated in the $^{101}\mathrm{Ru}+n_{th}$ reaction in this work.

$E_{\gamma1}$–$E_{\gamma2}$ cascade	A_2/A_0 exp.	A_4/A_0 exp.	$\delta^{exp}(E_{\gamma1})$
468.6–475.1	0.364(13)	1.156(26)	
628.0–475.1	−0.074(11)	0.337(21)	−287(−∞,+234)
631.3–475.1	0.104(8)	0.009(15)	
1105.3–475.1	0.117(9)	0.014(19)	0.17(2)
1046.5–475.1	−0.301(6)	−0.087(11)	−7.8(6)
636.8–468.6	0.001(9)	0.018(18)	
601.3–638.8	0.05(11)	0.12(21)	
601.3–1105.3	0.27(8)	−0.16(18)	0.6(−0.4,+0.6)
1075.3–631.3	0.177(18)	0.022(36)	0.06(6) or −1.02(12)

The energies of levels and transitions in Table I agree with the literature values [16] and are, in most cases, more accurate. Two examples illustrate the high quality of the angular correlations shown in Table II: (i) the measured A_2 and A_4 values for the very anisotropic, 468.6- to 475.1-keV cascade agree well with the theoretical values of 0.357 and 1.143, respectively, and (b) the δ values of +0.25(3) and −5.7(3) reported previously [16] for the 1105.3- and 1046.5-keV transitions, respectively, agree with our values, both in amplitude and sign.

Decays of the 1580.5-keV level to the 1103.1- and 1106.4-keV levels, not observed until now, are expected in the vibrational limit [13]. In a fit to the singles spectrum we have found a new line at 477.32(9) keV. Its energy agrees with the 1580.48- to 1103.09-keV difference between the corresponding level energies (Table I). Because of the particular location of the 477.3 decay branch, double-γ coincidences involving this line are obscured by the strong 475.1-keV line. More helpful are triple-γ coincidences. The presence of the 477.3-keV decay is supported by a spectrum doubly gated on the 475.1- and 628.0-keV lines, in which there is a doublet around 476 keV with two components fitted at 475.0(2) and 478.1(3) keV. However, the intensity of the 475.0(2)-keV component is more than twice that of the 478.1(3)-keV component, while they should be equal. This may be due to chance self-coincidences of the very strong 475.1-keV line or due to another line in this multiplet.

To clear these doubts we have measured a singles spectrum from the neutron capture on ^{101}Ru, using the high-resolution Bragg spectrometer GAMS5 of Institut Laue-Langevin (ILL), equipped with a curved crystal in DuMond diffraction geometry [17,18]. Figure 2 shows a fragment of a spectrum around the 475-keV multiplet. In the inset one sees a line at 477.41(6) keV. Its intensity is 0.013(4) as compared to the 475.095-keV reference line [16]. The decay to the 1106.4-keV level, expected at 474.11 keV is not observed. The error bar for the intensity of the 477.41-keV line may serve as the estimate of the upper intensity limit of the unobserved 474.11-keV line.

Our coincidence data indicate the existence of a 2181.7-keV level. Its decay branches limit the possible spin for this level to 2^+, 3^+, 3^-, and 4^+. The 3^- and 3^+ options are eliminated by angular correlations. The 4^+ solution is consistent with angular correlations (all fits with $\chi^2 \leqslant 1.0$). Spin 2^+ is less

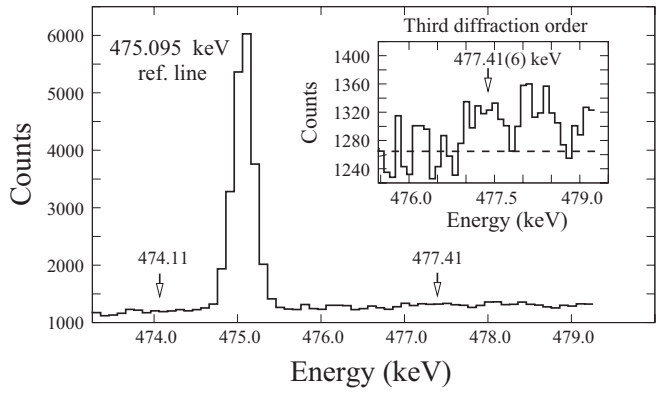

FIG. 2. Singles-γ spectrum from GAMS5, measured in the third diffraction order. In the picture one channel corresponds to 100 eV. The energy scale has been adjusted to reproduce the energy of the 475.095(1)-keV reference line [16], with accuracy of 0.003 keV.

likely due to $\chi^2 = 5.4$ for the 1075.3- to 631.3-keV cascade and the absence of any decay to the ground and the 475.1-keV states. Therefore, we propose that the 2181.7-keV level has spin 4^+ and is the next excitation in the band on top of the 943.7-keV level. The 2800.2-keV level, which decays only to the 6^+ level at 1873.2 keV, is a candidate for the 6^+ member of the band on top of the 943.7-keV level. The nonobservation of a decay to the 2181.7-keV level may be due to the detection limit.

In Ref. [13] it was suggested that the decay properties of the 1580.5-keV level are what one expects in a rotor. However, the present data are in favor of a vibrational picture. The selection rules for a vibrator predict $B(E2)$ branching ratios of 1.4:0.57:1.03 for the decay of the 2_3^+ level to the 0_2^+, 2_2^+, and 4_1^+ levels, respectively [19]. With the upper limit of γ intensity for the 474.1-keV decay to the 1106.4-keV level, the upper limit for the $B(E2)$ rate for this pure $E2$ branch is 11.5 W.u. This value is similar to what is observed in ^{100}Ru [20], a nucleus which is closer to the vibrational limit. A similar analysis for the 477.4-keV is more difficult because we do not know the mixing ratio, δ, for this $M1 + E2$ transition. We have calculated the δ value using the formalism of the generalized Bohr Hamiltonian [21,22] (the full account of these calculations will be published elsewhere [15]). The resulting $B(E2)$ value for the 477.4-keV decay branch is 6.8 W.u., compared with the expectation of 8.8 W.u. for the vibrator. This result is again close to the vibrational limit.

To learn more about the vibrational cascade on top of the 843.7-keV, 0_2^+ level, we have drawn the systematics of 0_2^+ bands in Ru isotopes, shown in Fig. 3. In ^{108}Ru, at the neutron number $N = 64$, the cascade on top of the 0_2^+ level has level spacing characteristic of a rotational band. This changes gradually with the decreasing neutron number towards vibrational spacings in ^{102}Ru. The vibrational character and a low deformation of the 0_2^+ band in ^{102}Ru may be a surprise. With eight valence neutrons one would expect high collectivity in this state. Below we present a simple picture, which may explain the low collectivity in this cascade and the role of the 0_2^+ configuration in the rapid deformation change in this region.

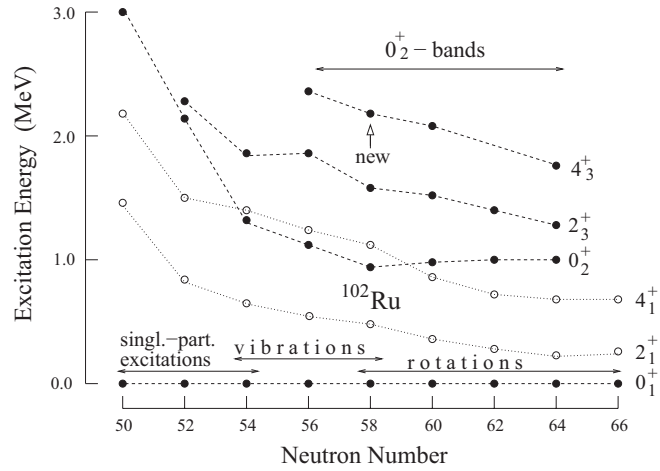

FIG. 3. Systematics of 0_2^+ bands in Ru isotopes. The 2_1^+ and 4_1^+ levels are also shown to assist further discussion (see the text). Lines are drawn to guide the eye. The data are taken from Refs. [16,20,23–28].

In spherical nuclei one observes a collective excitation, a phonon state with spin and parity 2^+, corresponding to a quadrupole vibration of the nuclear surface. At twice its energy, the 0_2^+, 2_2^+, and 4_1^+ triplet of collective two-phonon states is formed. The 4_1^+ and 2_2^+ level of the triplet are progenitors of the 4^+ and 2^+ excitations of the ground state and γ cascade, respectively (see Fig. 1). The role of the 0_2^+ level is less clear. One expects that due to its two-phonon character this level should be more collective than the ground state.

Figure 4 shows positions of the 0_1^+ and 0_2^+ levels in Ru isotopes (filled circles) relative to the 2_1^+ phonon excitation. In the neutron range from $N = 50$ to $N = 58$ the 0_1^+ and 0_2^+ levels display a characteristic, two-level mixing pattern as a function of neutron number. The symmetry seen in the figure supports the two-phonon contribution to the 0_2^+ level, because in vibrational nuclei this level is expected at twice the excitation energy of the 2_1^+ phonon excitation. More interesting

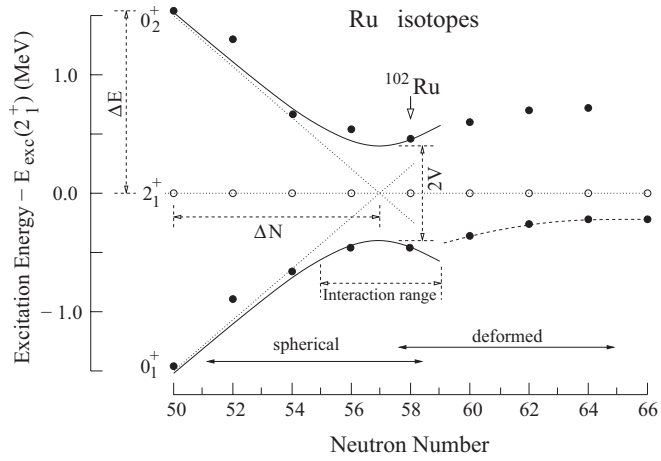

FIG. 4. Positions of 0_1^+ and 0_2^+ levels (full dots), relative to the 2_1^+ phonon excitation (open dots) in Ru isotopes. Solid lines represent a two-level mixing calculation. Symbols shown in the figure are explained in the text.

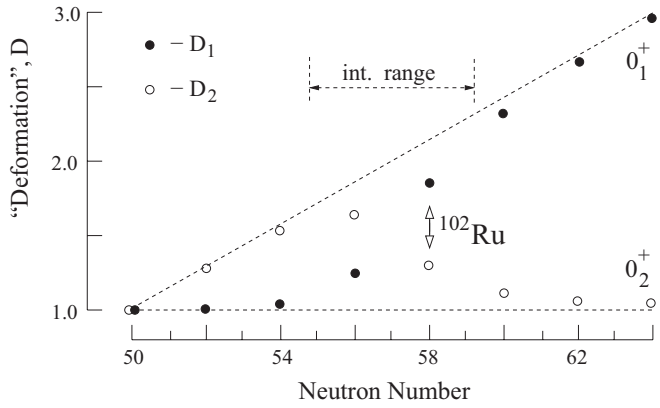

FIG. 5. Exchange of properties between the crossing 0_1^+ and 0_2^+ levels, as simulated in two-level mixing calculation. See text for the explanation of D_1 and D_2 and further comments.

is the fact that the two levels interact strongly in a limited range, $N = 55$ to 59.

We have performed a two-level mixing calculation (see, e.g., Ref. [19]) for the data points in Fig. 4, under simple assumptions that the excitation energy of the unperturbed 0^+ level, 0_{1u}^+ (or 0_{2u}^+), raises (or falls) linearly with the increasing number of valence neutrons, $n = N - 50$, as $E(0_{1u}^+) = \Delta E(-1 + n/\Delta N)$ [or $E(0_{2u}^+) = \Delta E(1 - n/\Delta N)$], as shown by dotted lines in Fig. 4 (ΔN is the distance to the crossing point as shown in Fig. 4). The perturbed energies, calculated using $\Delta E = 1500$ keV (which is half of the separation energy at $n = 0$), $\Delta N = 7$, and the interactions strength of $V = 400$ keV, are shown in Fig. 4 by solid lines. The calculation reproduces the experimental points surprisingly well, supporting the picture of the 0_1^+ and 0_2^+ states crossing.

When crossing, the two levels exchange their properties [19]. One may expect that the ground state in ^{102}Ru will gain collectivity, while the collectivity in the cascade on top of the 0_2^+ will decrease. A schematic simulation shown in Fig. 5 illustrates this process. Taking the ΔE, ΔN, and V parameters, as found above, we have simulated the exchange between the 0_1^+ and 0_2^+ levels of their certain property, D. We assumed that for the unperturbed 0_2^+ state, 0_{2u}^+, its D value increases linearly with n from 1.0 to 3.0 arbitrary units over the distance $2\Delta N$, $D_{2u} = 1.0 + n/\Delta N$, while the D value of the unperturbed 0_1^+ state, 0_{1u}^+, stays constant, $D_{1u} = 1.0$ (the D parameter could be seen as a measure of a collectivity). The expectation value of D in the 0_1^+ perturbed level is calculated as

$$D_1 = \langle 0_1^+|D|0_1^+\rangle = \alpha D_{1u} + \beta D_{2u},$$

where α and β are the usual mixing amplitudes in the perturbed level, $|0_1^+\rangle = \alpha|0_{1u}^+\rangle + \beta|0_{2u}^+\rangle$ [19], while $D_{1u} = \langle 0_{1u}^+|D|0_{1u}^+\rangle$, $D_{2u} = \langle 0_{2u}^+|D|0_{2u}^+\rangle$, and $\langle 0_{1u}^+|D|0_{2u}^+\rangle = 0$. The analogous formula for the 0_2^+ level reads $D_2 = -\beta D_{1u} + \alpha D_{2u}$.

The simulation shows that the D_1 value (collectivity of the ground state) increases rapidly over a small range of neutrons $54 < N < 60$, where, effectively, the interaction takes place. This result, obtained with realistic ΔE, ΔN, and V parameters, coincides with the four to six neutron range of the deformation change observed in Sr and Zr isotopes [6,11].

TABLE III. Truncated quadrupole shape invariants for 0_1^+ and 0_2^+ levels in Ru isotopes. The data are taken from Refs. [16,20,21,27,28]. See text for further explanation.

	^{98}Ru	^{100}Ru	^{102}Ru	^{104}Ru
$q_2(0_1^+)$	0.43(1) e^2b^2	0.52(9) e^2b^2	0.65(2) e^2b^2	0.89(2)[a] e^2b^2
$q_2(0_2^+)$		0.61(14) e^2b^2	$<$0.98 e^2b^2	0.62(14) e^2b^2

[a]Without Ref. [21]; $q_2(0_2^+) = 1.41(6)e^2b^2$ if Ref. [21] is used only.

In contrast, the D_2 value falls and at $N = 58$ has low D value, coinciding with the low collectivity in the cascade on top of the 0_2^+ level in the heavier Ru isotopes.

An analogous crossing between 0_1^+ and 0_2^+ states has been reported in Gd nuclei [29] of the $A \approx 150$ region, another place where a rapid change of nuclear deformation occurs. In Ref. [29] the crossing has been traced with the help of the quadrupole shape invariant, q_2, which measures quadrupole collectivity (or deformation, due to its proportionality to $\langle\beta_2\rangle$) [30]. Interestingly, the behavior of the q_2 invariants for the 0_1^+ and 0_2^+ states in Gd nuclei is similar to the behavior of D_1 and D_2 values in Fig. 5. As shown in Fig. 1 of Ref. [29], the $q_2(0_1^+)$ invariant increases quickly over a range of four neutrons while the $q_2(0_2^+)$ invariant, which first grows faster than $q_2(0_1^+)$, starts to drop just before the crossing point and after the crossing is smaller than $q_2(0_1^+)$.

The available $B(E2)$ rates in Ru isotopes allow one to construct truncated $q_2(0_1^+)$ and $q_2(0_2^+)$ values, as described in Ref. [29]. We used the $B(E2)$ rates between the 0_1^+, 0_2^+, 2_1^+, 2_2^+, and 2_3^+ levels in the $^{98-104}$Ru isotopes [16,20,21,27,28]. The q_2 values obtained are shown in Table III. One observes that the $q_2(0_1^+)$ invariant increases with the increasing neutron number. In contrast, the $q_2(0_2^+)$ does not increase and in ^{104}Ru is significantly lower than $q_2(0_1^+)$. It is of great interest to improve the accuracy of the experimental $B(E2)$ values in the Ru isotopes, especially those used to construct $q_2(0_2^+)$ in ^{100}Ru and ^{102}Ru.

It is worth noting that the truncated $q_2(0_1^+)$ and $q_2(0_2^+)$ values in Table III are dominated by the $B(E2; 2_1^+ \to 0_1^+)$ and the $B(E2; 2_3^+ \to 0_2^+)$ rates, respectively. Therefore, they reflect the "in-band" $B(E2)$ rates and thus collectivity in the two cascades.

It is interesting to ask about the mechanism which creates deformation in the 0_2^+ bands in the $50 < N < 60$ range. The proposed two-phonon excitation may be only one of the contributing effects. While the ground states at $N \leqslant 56$ should be dominated by the $d_{5/2}$ pair of neutrons, at the excitation energy of the 0_2^+ level one expects the $(g_{7/2})^2_{0^+}$ configuration, which may contribute to the 0_2^+ level. With the $\pi g_{9/2}$ proton orbital well populated in the Ru isotopes, the SOP mechanism proposed in this region for the ground state [1,2] may actually become the source of the collectivity in the 0_2^+ level. Such a possibility has been suggested for Sr isotopes [31] and recently supported by calculations of various 0^+ levels in ^{96}Sr and ^{98}Zr isotones of ^{102}Ru [32].

Finally, we note that above $N = 58$ the picture changes. As shown in Fig. 4, the 2_1^+ excitation energy (the difference between the 0_1^+ points and the 2_1^+ line) drops significantly.

This suggests that another deformation-generating mechanism starts around $N = 60$, which is most likely due to the population of low-Ω, deformation-driving orbitals, originating from the $\nu h_{11/2}$ intruder [7–9]. It has been proposed in Ref. [13] that the rather low energy of the 2_1^+ level in ^{102}Ru and the $B(E2; 2_1^+ \rightarrow 0_1^+)$, about 50% higher in ^{102}Ru than in lighter Ru isotopes, may indicate a rotational nature for the ground-state band in ^{102}Ru. In fact, the 0_1^+ point for ^{102}Ru in Fig. 4 might be seen as the lowest-N family member of deformed Ru isotopes. However, its deformation is probably of a dynamic nature in contrast to the heavier Ru isotopes, which have static deformation. Therefore, it is more likely that ^{102}Ru is the highest-N, spherical Ru isotope. Figures 3 and 4 show that the 0_2^+ band in ^{102}Ru can be seen as a "seed" for the 0_2^+ deformed bands at $N > 58$. It would be interesting to study the mechanism which generates the deformation in 0_2^+ bands at $N > 58$. One possibility is that the SOP mechanism is still at work at $N > 58$. Considering the fact that the energy separation between the 0_1^+ and 0_2^+ levels is not larger there than in ^{102}Ru, it is also possible that the two levels still interact and the strong deformation generated in the 0_1^+ ground state by the $\nu h_{11/2}$ intruder population is passed to the 0_2^+ level.

Finally, let us remark that the role of the third 0^+ level, proposed earlier as a band head of a deformed structure, is not clear. In ^{94}Ru, ^{100}Ru, and ^{102}Ru the 0_3^+ level scatters at 750 keV above the 0_2^+ level. At $N = 60$, it drops to be found only 347 keV above the 0_2^+ in ^{104}Ru but at $N = 62$ it rises suddenly to 1643 keV above the 0_2^+ level. It is of interest to search for the unknown 0_3^+ excitation energy in ^{96}Ru, ^{98}Ru, and ^{108}Ru to see if any interaction pattern is emerging between the 0_2^+ and 0_3^+ levels.

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