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### <sup>102</sup>Ru, A Pivotal Nucleus in the $A \sim 100$ region

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The nucleus  ${}^{102}$ Ru was studied with the GRID lifetime technique and an upper limit on the  $B(E2: 2_3^+ \to 0_2^+)$  of 21.6 W.u. was obtained. This nucleus is pivotal in the  $A \sim 100$  region as it marks the boundary, in Z, between nuclei that do, and do not, become deformed with increasing neutron number, and, in N, between those that exhibit characteristics of a phase transition mediated by changes in subshell structure and those that do not. The measurements are discussed primarily in terms of vibrational structure.

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#### I. INTRODUCTION

The nucleus  $^{102}$ Ru is an important isotope in the  $A \sim 100$  transitional region. This is seen in Fig. 1 (left) where the ratio  $R_{4/2}$  of the energy of the first  $4^+$  state to the first  $2^+$  state is plotted against neutron number for various elements in the  $A \sim 100$  region. The Ru isotopes are a fulcrum for this region separating those elements, Sr, Zr, and, to a lesser extent, Mo which drive towards deformation near N = 60, from the Pd and Cd nuclei, which show much less collectivity as Sn is approached. This is shown by the crossing pattern [1], where  $R_{4/2}$  values for Z < 44 cross from below those with  $Z \ge 44$  to above between N = 58 and 60.

Another measure of the development of collectivity is the energy of the first  $2^+$  state itself. While  $R_{4/2}$  increases from closed shell nuclei into deformed regions,  $E(2_1^+)$  decreases. It is often more illustrative, therefore, to plot  $1/E(2_1^+)$  since this quantity behaves similarly to  $R_{4/2}$ . This is shown in the bottom half of Fig. 1 (left).



FIG. 1: (Color online) Top:  $R_{4/2} == E(4_1^+)/E(2_1^+)$  values for the  $A \sim 100$  region, against N (left) and against Z (right). Bottom: Similar except for  $1/E(2_1^+)$  in units of keV<sup>-1</sup>.

Again, one can see the classic crossing pattern typical of regions of rapid shape change driven by the dissolution of a magic number (Z = 40 at N = 60 in this case). See refs. [2–4].

In this region, N = 58 (corresponding to  $^{102}$ Ru for Z = 44) itself plays a central role. This is seen in Fig. 1 (right panels, near  $Z \sim 40$ ). Nuclei with  $N \leq 58$  show vibrational structure at low energy ( $R_{4/2} \leq 2.4$ ). Above N = 58 the structure tends towards a rotational character. Figure 1 (top right), in fact shows that N = 58 is the last concave curve, while N = 60 plays the same role of transitional character as N = 90 does in the rare earth region and nuclei with N > 60 have convex behavior. The values of  $1/E(2_1^+)$  show similar behavior. The "bubble" pattern [5] again points to a breakdown of Z = 40 as a magic number at N = 60. Finally, Ru (Z = 44) is the terminus (with increasing Z) of the bubble. For larger Z values, the  $R_{4/2}$  and  $1/E(2_1^+)$  curves for different elements either merge or show a regular progression.

These remarks point to  $^{102}$ Ru as central to understanding this region. Interestingly, <sup>102</sup>Ru is itself somewhat schizophrenic showing an evolution of structure as a function of spin. This is most easily shown in an EGOS-plot in which <sup>102</sup>Ru was used as the example of that technique because of the interesting variation of structure it shows. An EGOS plot of  $E_{\gamma}/J$  vs. angular momentum J for  $^{102}$ Ru based on data from [6] is shown in Fig. 2. In such a plot, a vibrational structure gives a curve that continually decreases with increasing spin, asymptotically towards zero. In contrast, a rotor shows a much flatter curve which increases, from J = 2 to large J values, by a factor of 4/3. Nuclei with  $R_{4/2}$  values less than ~ 3.0 exhibit the down sloping behavior of a vibrator, albeit with flatter trajectories with spin as  $R_{4/2}$  approaches 3.0. Nuclei with  $R_{4/2}$  values above 3.0, and approaching the rotor value 3.33, show the slightly increasing behavior. An anharmonic vibrator with  $R_{4/2} = 3.00$  is a flat line in an EGOS plot.

The  $^{102}$ Ru data decrease at first, typical of a vibrator, but then, at  $J \sim 12$ , start to increase, resembling the rotor behavior. The interpretation [6] is of a change in structure around spin 10 when two maximally aligned



FIG. 2: (Color online) EGOS plots: The EGOS plots (arbitrary normalization) of  $^{102}$ Ru based on data from ref. [6] are compared to EGOS plots of the harmonic vibrator and the axially symmetric rotor.

 $h_{11/2}$  neutrons start to dominate the yrast structure and polarize the nucleus along the rotation (deformed) axis.

Despite the importance of  $^{102}$ Ru, its structure is rather poorly known. The lowest levels (as noted above) resemble a vibrator, although inspection of the B(E2) values suggests considerable anharmonicity and loss of collectivity. Above the 2-phonon triplet no B(E2) values are known except for the 3-phonon candidate 6<sup>+</sup> level. But that level is the least informative of the 3-phonon states (since it is difficult to make a low lying 6<sup>+</sup> level with alternate structure in a spherical nucleus) and it is important to test the vibrational structure for other candidates for 3-phonon levels.

#### **II. EXPERIMENT**

Fortunately, a good set of potential 3-phonon levels exists at around 1500 keV. In particular, the 1581 keV  $2^+$  level has three known decay branches but its lifetime is unknown. As a first step in unraveling the structure of  $^{102}$ Ru, we have therefore measured the lifetime of this level, using the Gamma Ray Induced Doppler broadening technique (GRID) [7] and obtained a lower limit of  $\tau >$ 4.4 ps. We first describe the measurements and then comment on their consequences.

The GRID technique uses the recoil of a nucleus following the emission of a  $\gamma$ -ray as a means of Doppler broadening the spectrum of a subsequent  $\gamma$ -ray, provided that the  $\gamma$ -ray is emitted while the recoiling nucleus is still in flight. These stopping times are on the order of a few hundreds of fs and so the technique is best used for lifetimes less than, at most, a few ps. The key challenge stems from the fact that, in medium mass and heavy nuclei, the recoil energies, and therefore the Doppler broadening, are typically only in the few 10 eV range. This in turn implies that the second  $\gamma$ -ray must be detected with a FWHM on



FIG. 3: (Color online) Line profiles showing the contributions from instrumental, thermal and lifetime broadening and the data for the 1105 keV transition from the 1581 keV state to the  $2_1^+$  level. The left insert shows the dependence of the  $\chi^2$  as function of lifetime. Lifetimes shorter then  $\tau = 4.4$  ps (increased broadening of line profiles) can be excluded, while longer lifetimes (convergence towards thermal broadening) are statistically compatible with the experimental data. The right insert, a magnification of the lines for  $\tau = 4.4$  ps, indicates that for this value lifetime contribution just starts to show some additional broadening.

the order of 1-2 eV itself. Such extraordinary resolving power requires the use of a flat crystal spectrometer so that the gamma ray energy is determined from the angle of diffraction. For a sufficiently perfect crystal, and a stable environment, the required energy precision and resolution can be obtained provided the diffraction angle can be measured to accuracies of  $\sim 10^{-10}$  radians. With appropriate laser measurement techniques this is possible. The GAMS spectrometers [8] at the ILL at Grenoble are the primary example of such instruments and are routinely used for such lifetime measurements with the GRID technique [9–11].

The present experiment was carried out at the ILLreactor. The target of 12 g. natural Ru was placed near the reactor core into a thermal neutron flux of  $\sim 5~{\rm x}$  $10^{14} \text{sec}^{-1} \text{cm}^{-2}$ . The thermal neutron capture cross section on <sup>101</sup>Ru is 5 b which is adequate for such an experiment. The product nucleus,  $^{102}$ Ru, is formed at the neutron separation energy and decays by complex cascades of  $\gamma$  rays. <sup>101</sup>Ru has a ground state spin of  $5/2^+$  so that s-wave neutron capture (dominant at thermal neutron energies) leads to capture state spins of  $2^+$  and  $3^+$ . Sequences of a few decay cascade dipole transitions then readily populate low lying final states in the spin range  $0^+$  to  $5^+$  rather indiscriminately. The 1581 keV has J  $= 2^+$  and is therefore easily populated. The strongest (in intensity)  $\gamma$  ray de-exciting this level is the 1105 keV  $\gamma$  ray and it is this transition whose Doppler profile was measured here, using the GAMS4 spectrometer. Figure 3 shows the instrumental profile of this device along with

the profile for the 1105 keV line. Within statistical sensitivity no visible widening is observed.

In order to convert this null result to a lifetime limit, one needs to consider the  $\gamma$ -feeding of the 1581 keV level since that affects the average nuclear recoil velocity. One could obtain estimates of this feeding distribution from measured spectra (that means the experimentally known feeding) and the missing part from statistical model calculations [12] and/or from extreme feeding assumptions. In the present case the missing feeding was replaced by a two step cascade, where lifetime  $\tau_i$  and energy  $E_i$  of the intermediate level are varied within the  $\chi^2$  minimization procedure. Independent of the assumed  $\tau_i, E_i$  parameter combination the  $\chi^2$  converged to a minimum value  $\chi^2_{min}$  with increasing lifetime ( $\tau > 10$  ps). This indicates that the dominant contribution to the measured Doppler-broadening results from thermal motion and consequently that the lifetime of the 1581 keV state must be much larger than the typical slowing down time and consequently one can only deduce a lower limit on  $\tau$ . The convergence of  $\chi^2(\tau)$  towards the value of  $\chi^2_{min}$ depends weakly on the chosen parameters of  $E_i$  and  $\tau_i$ . Combinations of high  $E_i$  and low  $\tau_i$  values tend to give slower convergence, while low  $E_i$  and high  $\tau_i$  converge faster. Obviously a conservative estimate for the lower limit is based on a combination of a low  $E_i$  and high  $\tau_i$ , with the constraint that  $E_i$  should be sufficiently high to still produce some recoil motion. We found that  $E_i$ should be about 750 keV above the 1581 keV level and  $\tau_i$  larger than 100 fs. A  $\chi^2(\tau)$  curve for the combination of  $E_i=2331$  keV and  $\tau_i=4$  ps has been calculated. The lower limit was extracted from the crossing of this curve with a value of  $\chi_{min} + 2$  (this adds  $1\sigma$  errors of determining  $\chi_{min}$  and of the lifetime itself). Additionally, to provide a consistency check for the experimental parameters such as thermal broadening and instrument response function, we re-measured the known lifetime of the 1362 keV  $2^+$  level in  $^{100}$ Ru via the 823 keV transition. Here the feeding is very well known (about 50%). We obtained a value of 0.415 ps  $< \tau < 0.914$  ps, which is somewhat shorter than the value of  $\tau = 1.47\binom{35}{23}$  ps previously measured via Coulomb excitation [13].

Thus the net result obtained in the present experiment is  $\tau > 4.4$  ps. There are three known  $\gamma$  ray decays of the 1581 keV level. See the partial level scheme in Fig. 4. Using the experimentally known branching ratios, one gets the limits on B(E2) values in Table 1.

#### **III. INTERPRETATION**

Given the rather clear multiplets of levels (e.g., the 0<sup>+</sup>,  $2^+$ ,  $4^+$  triplet near 1100 keV) and the  $R_{4/2}$  value of 2.3 for  $^{102}$ Ru the most obvious comparison of its level scheme is to the vibrator. Figure 5 summarizes the transition rate data and compares it to that model. The  $2^+_1$  to ground state transition of 45 W.u. is typical of, but somewhat larger than, such transitions in vibrational nuclei in this



FIG. 4: Partial level scheme for <sup>102</sup>Ru showing the levels and transitions of interest for this work. Based on ref. [14].



FIG. 5: Schematic level scheme showing the one-phonon and two-phonon levels of the harmonic vibrator along with the  $2_3^+$ and  $6_1^+$  three-phonon states. The B(E2) values in black are those of the vibrator model, normalized to the experimental value of 45 W.u. for the  $2_1^+$  to  $0_1^+$  transition. Experimental values (with uncertainties) or limits are shown in red. Data from the Nuclear Data Sheets [14] and the present experiment.

region [15–21]. The B(E2) values from the triplet of  $0^+$ ,  $2^+$  and  $4^+$  levels around 1100 keV also resemble what is found for typical vibrational nuclei near  $A \sim 100$ , namely a  $4^+_1 \rightarrow 2^+_1$  transition that exhausts the majority of the two-phonon to one-phonon strength while the decays of the two-phonon candidate  $2^+_2$  and  $0^+_2$  levels are substantially weaker.

As for higher multi-phonon levels, in <sup>98</sup>Ru [22], no further states were found with any resemblance whatsoever to the three-phonon vibrator levels. Here in <sup>102</sup>Ru the situation may be somewhat closer to the vibrator model. The  $B(E2: 6_1^+ \rightarrow 4_2^+)$  value of 68(25) W.u., despite the large uncertainty, nevertheless exhibits a high degree of collectivity. The situation for the  $2_3^+$  level at 1581 keV is

TABLE I: B(E2) values for the decay of the 2<sup>+</sup> 1581 keV level in <sup>102</sup>Ru.

Transition $(keV)$	Final Energy $(keV)$	Final Spin	B(E2) value (W.u.)
1580.5	0	$0^{+}$	< 0.05
1105.7	475.1	$2^{+}$	< 0.5
636.8	943.7	$0^{+}$	< 21.6

murkier since our measurements only give an upper limit to the B(E2) values. Nevertheless, the relative B(E2)values, with the  $2^+_3$  to  $0^+_2$  transition orders of magnitude stronger than the forbidden transitions to the first  $2^+$ state and the ground state, are consistent with the vibrator selection rules. Further, if the actual B(E2) values are close to the limits obtained here, then the  $2^+_3$  to  $0^+_2$  transition has at least a substantial portion of the expected collectivity. In any case, one clear conclusion from our measured upper limit is that the phonon strength at the three phonon level is severely fragmented.

Comparison to  ${}^{98}$ Ru on the one hand, where vibrational structure above the two phonon level seems completely destroyed, and the Cd nuclei from  ${}^{110-118}$ Cd [15– 21] where good candidates for three and higher phonon levels with substantial fractions of the vibrator collectivity are known, suggests that  ${}^{102}$ Ru has an intermediate structure.

An alternate way of looking at  $^{102}$ Ru is in terms of the transition to a rotor behavior in this region. As noted in the introduction,  $^{102}$ Ru is pivotal, lying at the tipping point from vibrational to transitional and rotational structure in this region. Its  $2_1^+$  energy is lower than the lighter Ru isotopes (e.g.,  $^{98}$ Ru has  $E(2_1^+) = 652$  keV) and its  $B(E2: 2_1^+ \rightarrow 0_1^+)$  value is, for example, about 50 % higher than in both the lighter Ru and the Cd isotopes). Finally, inspection of differential operators [23] also slightly separates the behavior of  $^{102}$ Ru from both the lighter Ru isotopes and the higher Z elements en route to Sn, in the direction towards (but still far from) the rotor.

While, with  $R_{4/2} \sim 2.3$  and with strong transitions from each of the three levels at ~ 1100 keV to the  $2_1^+$  state, <sup>102</sup>Ru cannot in any way be described as a deformed rotational nucleus, the difficulties of interpretation are exhibited in an interesting way by the decay of the 1581 keV level studied here. We noted above that, at least in broad terms, its decay seemed to reflect the vibrator selection rules. It is worth noting, however, that they are also what one would expect in a rotor. In such a model the  $2^+_3$  level would be a rotational excitation of the  $0^+_2$  level and its B(E2) value to that level would far exceed those to the yrast levels. This, in fact, is the case, as seen in Fig. 5.

#### IV. CONCLUSION

Clearly, then, no obvious picture of the structure of  $^{102}\mathrm{Ru}$  yet emerges, both due to the lack of absolute B(E2) values for the higher levels, and to its structural complexity. Given its pivotal point in this mass region, however, further data would be very valuable. The present results provide some guidance in that direction but not nearly enough to reliably determine its structure. The most important would be to obtain absolute values rather than limits for the B(E2) values for the 1581 keV level as well as for the other possible candidate for 3phonon structure, the  $3_1^+$  level at 1521 keV and the  $4_2^+$ level at 1799 keV (The *relative* B(E2) values from each of these levels [evident in Fig. 4 after accounting for the  $E_{\gamma}^{5}$  factor] resemble the vibrator selection rules). Also important is to search for transitions (or limits) from the 1581 keV level to the  $2^+_2$  and  $4^+_1$  states near 1100 keV.

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