β^- decay of $^{102}Tc^g$

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The $^{102}Tc^g$ activity has been obtained by chemical separation of the parent nucleus ^{102}Mo from an $UO_2(NO_3)_2 \cdot 6\ H_2O$ sample, irradiated with 20-MeV bremsstrahlung. Four successive Ge(Li) spectra were measured. Several new γ rays were found and the energy values and relative intensities of the known γ lines are compared to the existing data. Levels at 0, 475.07, 943.65, 1103.15, 1580.83, and 1837.10 keV were found to be fed in the β^- decay of $^{102}Tc^g$. Based on the well-known, spectrometrically determined, yield for mass 102 in the thermal neutron fission of ^{235}U the absolute intensity for the 475.0 keV γ ray has been determined. A value of $(6.25 \pm 1.0)\%$ was found. Also the absolute intensities of the other observed γ lines could be obtained. Intensities for all β branches and corresponding logft values were calculated. The results differ considerably from those given in the literature. The systematics of the energy spectra of the doubly even Ru isotopes in the mass region $94 < A < 110$ are discussed. The total potential energy surface for quadrupole deformation is calculated. Energy spectra for ^{102}Ru are calculated within the interacting boson model and also treating cubic anharmonicities in perturbation theory. They are compared with the experimental results.

[RADIOACTIVITY $^{102}Tc^g$ [from $^{235}U(n_{th},f)$ and $^{238}U(\gamma,f)$, $E_{\gamma_{max}} = 20$ MeV, chemical separation of Mo]. Measured E_γ, I_γ. $^{102}Tc^g$ deduced logft ^{102}Ru deduced levels, j, π.]

I. INTRODUCTION

Until now very little experimental information was available concerning the decay of $^{102}Tc^g$ ($T_{1/2} = 5.28$ s; $J^\pi = 1^+$) (Ref. 1). The most recent and complete β-decay study was performed by Blanchot *et al.*[2] This study reveals unexpected results concerning the intensities of the different β branches. Indeed three intense β branches ($E_{\beta 1} = 2200$ keV, $I_{\beta 1} = 20\%$; $E_{\beta 2} = 3400$ keV, $I_{\beta 2} = 39\%$; $E_{\beta 3} = 4150$ keV, $I_{\beta 3} = 41\%$) were observed, in contradiction with the known β decay of similar doubly odd Tc isotopes, especially $^{100}Tc^g$, where only one very intense (93%) allowed β transition ($^{100}Tc^g \rightarrow \beta^- {}^{100}Ru^g$) is observed. However, the study of the β decay of $^{102}Tc^g$ is a complex problem because it is practically impossible to prepare pure $^{102}Tc^g$ sources. Indeed, if ^{102}Tc is produced by a neutron or a charged-particle induced reaction on Mo or Ru targets, besides the disturbing Mo, Ru, and Tc isotopes, one always produces a mixture of $^{102}Tc^g$ ($T_{1/2} = 5.28$ s) and $^{102}Tc^m$ ($T_{1/2} = 4.35$ min). Practically, this means that, due to the fact that most of the γ rays of $^{102}Tc^g$ are common to those of $^{102}Tc^m$, it becomes almost impossible to study both decays separately.

In fission studies it is possible, by combining γ spectrometric methods with catcherfoil techniques, to calculate from the absolute intensity of a γ ray of the decay of a given isotope, the yield of this isotope (see, e.g., Ref. 3). In the fission of uranium, $^{102}Tc^g$ is indirectly formed from the decay of ^{102}Mo ($J^\pi = 0^+$), which has a half life of 11.1 min. A value of 58% for the absolute intensity of the 475.0 keV γ ray in the decay of $^{102}Tc^g$ is deduced by Auble[1] from the β intensities given by Blachot *et al.*[2] This value gives, for the total chain yield of mass 102 in the post neutron mass distribution for the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung (which we studied in detail previously—Ref. 3—), a value which is about 10 times too small, compared to the expected value obtained by interpolation between the measured yields for masses 101 and 103.

In the experiments described in this paper, we determined the absolute intensity of the 470.5 keV γ ray from measured catcherfoil γ spectra for $^{235}U(n_{th}, f)$ (Ref. 5). This can be done since for the thermal neutron induced fission of ^{235}U, the post neutron mass yield for mass 102 (4.19 ± 0.06, the mass distribution is, as usually, normalized to a total yield of 200%) was measured directly.[4] On the other hand, in order to study the relative intensities of the different γ rays and β branches for the $^{102}Tc^g$ decay we separated the Mo fraction from the other fission products obtained in the photofission of ^{238}U with 20-MeV bremsstrahlung. The γ spectra obtained will be essentially a mixture of the well-known γ spectra of the decay of ^{101}Mo and ^{101}Tc, and of ^{102}Mo and ^{102}Tc. The re-

sulting β intensities are in contradiction with the results of Blachot *et al.*[2] but are very similar to those of Berzins *et al.*[6] for the decay of $^{100}Tc^g$. Blachot *et al.*[2] separated the Tc isotopes from the Mo fraction using a "milking" procedure. However, due to the short half-life, the separation is incomplete and its efficiency is not very high. The γ sources thus obtained are impure and have a very low intensity. Moreover, even with this procedure one cannot eliminate the presence of ^{101}Tc. Consequently, the β-decay results obtained with this method will be unreliable. Therefore we decided not to perform such milking experiments.

In this paper also the systematics of the energy spectra of ^{102}Ru and the surrounding doubly even Ru isotopes are discussed.[1,7-20] We studied more in detail the total potential-energy surface[21] for the quadrupole deformation degree of freedom in ^{102}Ru. Indications of anharmonicities result. Subsequently, the anharmonic quadrupole vibrational spectrum is calculated using phenomenological approaches to transitional nuclei: the interacting boson model (IBA) (Refs. 22–26) and second-order perturbation theory with cubic anharmonicities in the collective Hamiltonian.[27] Finally the quadrupole anharmonicities for ^{102}Ru are established by the experimental evidence for nonzero crossover ($\Delta N = 2$) $E2$ transitions.

II. EXPERIMENTAL TECHNIQUES

For the study of the decay of $^{102}Tc^g$ samples of 2 g $UO_2(NO_3)_2 \cdot 6H_2O$ were irradiated with 20-MeV bremsstrahlung. The irradiation time was 30 minutes and 20 irradiations were performed. Immediately after the irradiation the Mo fraction was extracted from the uranylnitrate, using the method of Cuninghame.[28] Starting 17 minutes after the irradiation, four successive γ spectra, each of 10 minutes, were taken, in order to be able to distinguish between the γ rays belonging to the decay of ^{101}Mo ($T_{1/2} = 14.62$ min) and ^{101}Tc ($T_{1/2} = 14.2$ min), or to the decay of ^{102}Mo ($T_{1/2} = 11.1$ min) and $^{102}Tc^g$ ($T_{1/2} = 5.26$ s). The apparatus used is the same as described earlier.[29] Due to the low intensity of most of the observed γ rays, no coincidence experiments were performed.

In order to determine I_γ^{abs} (475.0 keV), irradiations, with thermal neutrons, of a ^{235}U target, followed by an Al catcherfoil, were performed on the *T7* beam of the BRII reactor of the SCK-Mol (Belgium). The Cd ratio of this beam was about 30. The irradiation, cooling, and measuring times for the catcherfoil spectra were 15 minutes each. Because of the sufficiently high intensity of the 475.0 keV γ line and the absence of any observable disturbing contribution of γ lines in this energy region of the measured spectra, no chemical separations were needed in these experiments. More details concerning the apparatus used have been published in a previous paper.[5]

III. EXPERIMENTAL RESULTS

Part of the γ spectrum of the Mo fraction, taken with a 19 cm^3 Ge(Li) detector 17 minutes after the irradiation and for a period of 10 minutes, is given in Fig. 1. This spectrum is a summation of 20 individual runs. Besides the γ rays belonging to the decay of $^{102}Tc^g$, the most prominent γ rays of the decay of ^{101}Mo and ^{101}Tc are indicated.

The intense low-energy γ rays at 135.9 and 148.0 keV, and partly those at 211.7 and 223.8 keV (both contaminated with well-known γ rays of ^{101}Mo), could not unambiguously be identified. They have a decay period of (11 ± 1) min, but do not fit into the well-known level scheme of ^{102}Ru. They probably belong to the decay of ^{102}Mo, for which no γ rays are known at the moment. Indeed one does not expect excited states in the doubly even nucleus ^{102}Ru at the energy of these observed γ rays. On the other hand, if the above mentioned γ rays would fit between higher lying unknown levels of ^{102}Ru, the corresponding deexciting higher-energy γ rays should have been seen. This is certainly not so in our experiments. Also the fact that Blachot *et al.*[2] did not mention those intense γ rays in his study points towards this hypothesis.

Another rather intense γ ray at 865.6 keV, with a decay period of (9 ± 2) min was observed. Such a γ ray was also mentioned by von Baeckmann,[30] but not by Blachot *et al.*[2] It is very unlikely that this γ ray should belong to the decay of ^{102}Mo because if one admits that the above mentioned γ ray should correspond to a ground-state transition of a level at 865.6 keV, the β branch feeding such a level would have a logft value of 3.76, which is very improbable. On the other hand, this γ ray does not correspond to any known transition in ^{102}Ru. So if it belongs to the decay of $^{102}Tc^g$ its position is not fixed at the moment.

In Table I the energies and relative intensities of the γ rays belonging to the decay of $^{102}Tc^g$ are given. Because up to now no γ rays of the decay of ^{102}Mo are given in the literature,[1] our decay study of $^{102}Tc^g$ will be limited to the β branches feeding known levels in ^{102}Ru. Only the γ rays having the decay period of ^{102}Mo and corresponding to the observed transitions in ^{102}Ru are assigned to the decay of $^{102}Tc^g$. Due to better statistics in our spectra compared with the data of Blachot *et al.*[2] we were able to find several new γ rays. For the γ rays which were also seen by Blachot *et al.*[2] a better accuracy for the energy as well as the rel-

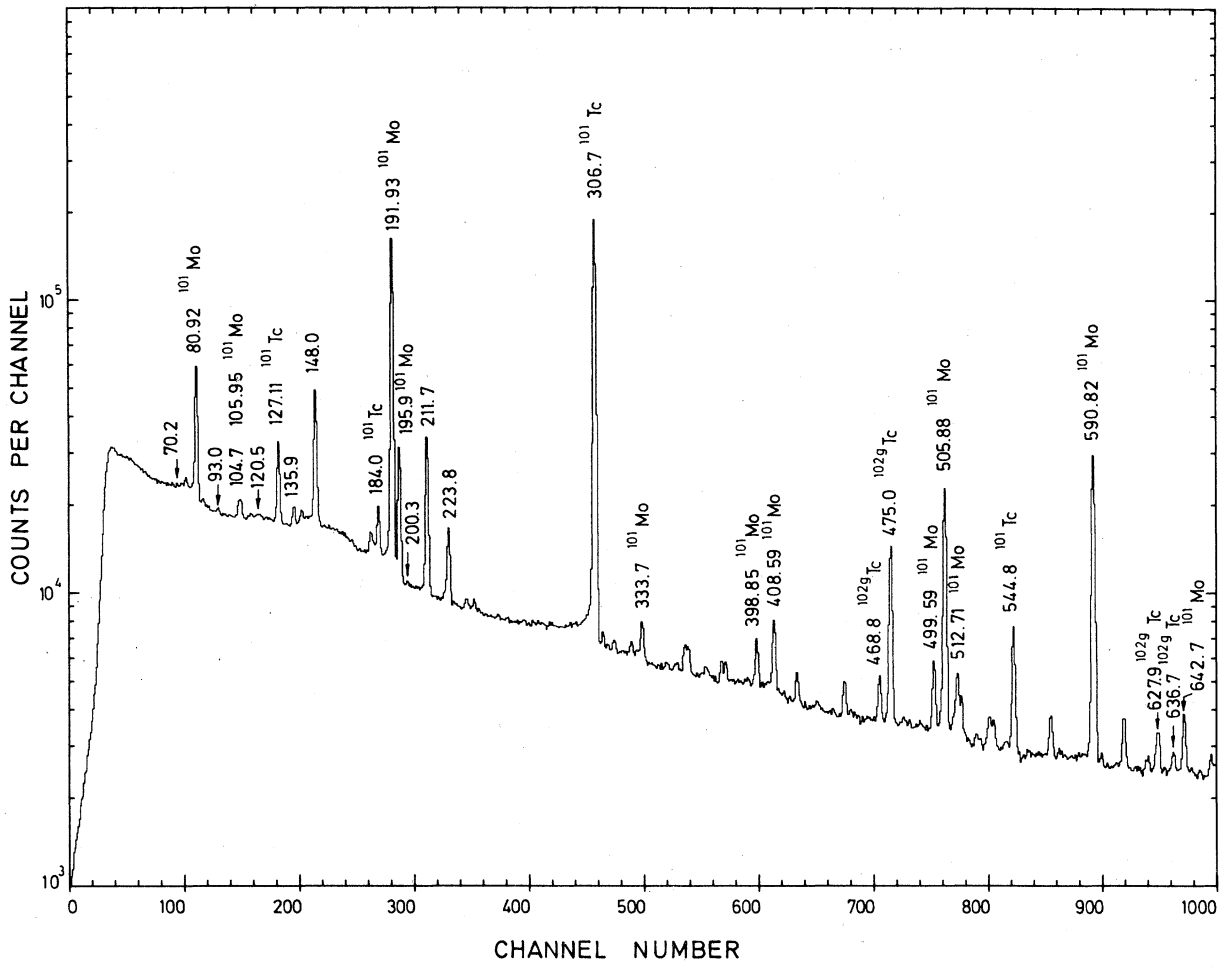

FIG. 1. Part of the γ-ray spectrum of the Mo fraction extracted from uranylnitrate, irradiated with 20-MeV bremsstrahlung. Besides the γ rays belonging to the decay of ^{102}Tc^{g} only the most prominent γ lines of the decay of ^{101}Mo and ^{101}Tc are given.

ative intensity of the observed γ rays was achieved.

In addition, weak γ transitions with a half life of 10 min$< T_{1/2} <$20 min and an energy of 70.2, 93.0, 120.5, 200.3, 742.6, 786.4, 2200.9, and 2434.6 keV were also observed, none of which fit into the existing level scheme of ^{102}Ru.

The determination of the absolute intensity of the 475.0 keV γ ray was based on the knowledge of the post neutron mass yield for mass 102 in the thermal neutron induced fission of ^{235}U and on the fact that the independent yield of ^{102}Tc is negligible for that fission process,[31] so that the total yield of mass 102 is given by the number of ^{102}Mo nuclei and consequently by the number of ^{102}Tc^{g} nuclei in the measured sample. From the measured catcherfoil spectra a value of (6.25 $\pm$1.0)% for $I_{\gamma}^{\mathrm{abs}}$ (475.0 keV) could be obtained. This value differs considerably from the value of 58% as given by Auble *et al.*[1]

Using the value $I_{\gamma}^{\mathrm{abs}}$ (475.0 keV) = 6.25% we find that the yield of mass 102 is 4.0 $\pm$ 1.2 for ^{252}Cf spontaneous fission and 3.1 $\pm$ 0.6 and 5.2 $\pm$ 1.1 for

TABLE I. Energies and relative intensities of the γ rays assigned to the decay of ^{102}Tc^{g}.

Blachot *et al.* (Ref. 2)		Our experiments	
E_{γ} (keV)	$I_{\gamma}^{\mathrm{rel}}$	E_{γ} (keV)	$I_{\gamma}^{\mathrm{rel}}$
468	15	468.8	13.2 $\pm$1.1
475	100	475.0	100
628	19	627.9	11.7 $\pm$1.2
•••	•••	636.7	5.8 $\pm$0.7
•••	•••	733.8	1.9 $\pm$1.0
1105	$\simeq$11	1103.5	5.6 $\pm$1.0
1105	$\simeq$ 9	1105.8	11.0 $\pm$1.6
•••	•••	1362.1	5.4 $\pm$1.2

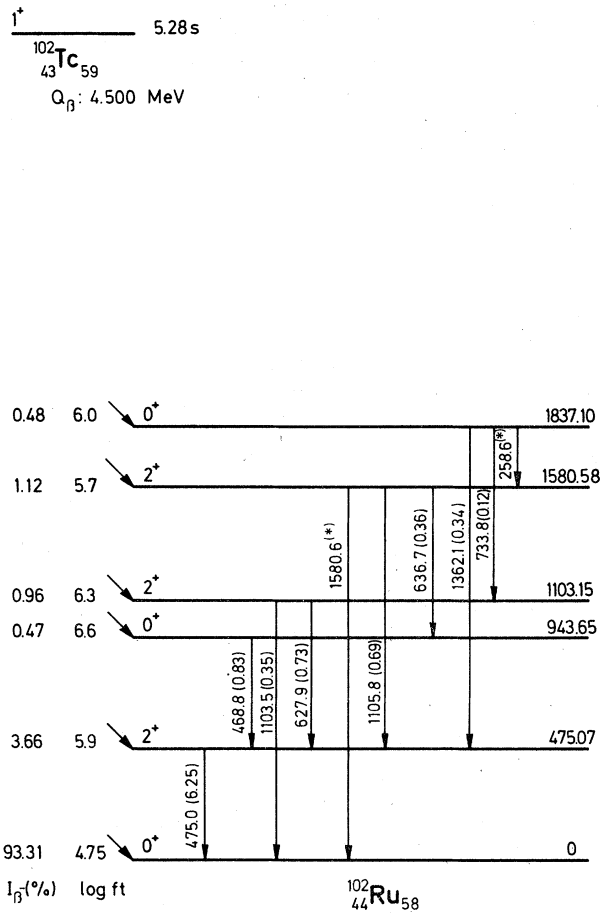

FIG. 2. Decay scheme of $^{102}Tc^g$. The γ-ray intensities are given per hundred $^{102}Tc^g$ decays. The intensities of the γ lines marked with (*) were taken from the literature[1] and could not be observed in our experiments. The level energies (in keV) and the J^π values are taken from Ref. 1.

the photofission of ^{235}U and ^{238}U with 25-MeV bremsstrahlung. This is in agreement with the expected values deduced from the interpolation between our earlier obtained yields of masses 101 and 103 for the same fissioning systems (see Refs. 3 and 5).

The decay scheme of $^{102}Tc^g$ as it could be obtained from our measurements is given in Fig. 2. The absolute intensities of the γ rays were calculated based on our value of 6.25% for the 475.0 keV γ ray. The energies and J^π values of the levels were obtained from reaction work and from the decay of ^{102}Rh.[1] The intensities and corresponding $\log ft$ values of the different β branches observed are given in Table II and are compared to those of Auble *et al.*[1] from which they differ considerably. As can be seen from Table II the very important fragmentation of the β branches,

TABLE II. Energies, intensities, and corresponding $\log ft$ values for the β transitions in the β^- decay of $^{102}Tc^g$.

	R.L. Auble *et al.*		Our experiments	
E_β (keV)	I_β (%)	$\log ft$	I_β (%)	$\log ft$
4500	≃41	≃5.0	93.31	4.75
4025	≃27	≃4.9	3.66	5.9
3557	≃ 9	≃5.1	0.47	6.6
3396	≃18	≃4.8	0.96	6.3
2920	≃ 5	≃5.0	1.12	5.7
2663	•••	•••	0.48	6.0

[a] The given I_β and $\log ft$ (Ref. 1) are deduced from the combined β-γ experiments of J. Blachot *et al.* (Ref. 2).

as given by Auble *et al.*,[1] disappears, and a strong β branch between the ground states of ^{102}Tc and ^{102}Mo is observed. This is also observed in the very similar $^{100}Tc^g \rightarrow \beta^- {}^{100}Ru$ decay.

IV. DISCUSSION

The energy spectrum of the nucleus ^{102}Ru can be situated between the pure vibrational and rotational limit. In Fig. 3 we plot the energy ratio $E_{J_i^\pi}/E_{2_1^+}^+$ for the $J_i^\pi = 4_1^+$, 2_2^+, 0_2^+, 6_1^+, and 8_1^+ levels for the doubly even Ru isotopes with $50 \leq N \leq 66$ (Refs. 1, 7–20). One can observe a smooth change starting from the seniority $v = 2$ states originating from the $(1g_{9/2})^4$ proton configuration in $^{94}_{44}Ru_{50}$ (single closed shell nucleus) towards anharmonic vibrational spectra in the mass region 98–102 ($54 \leq N \leq 58$). For the heaviest doubly even Ru isotopes for which the $J_i^\pi = 2_1^+$, 4_1^+, 6_1^+, 8_1^+ levels are known, rotational properties are clearly established.

The anharmonicities corresponding to the quadrupole degree of freedom (ϵ_2), of the nucleus ^{102}Ru itself, can be studied in more detail by calculating the total potential-energy surface for this nucleus.[21] Therefore we have made use of the macroscopic-microscopic Strutinsky renormalization procedure[32] using the Nilsson modified harmonic-oscillator potential[33]

$$V = \tfrac{1}{2}\hbar\omega_0(\epsilon_2)\rho^2[1 - \tfrac{2}{3}\epsilon_2 P_2(\cos\theta)] - \kappa\hbar\omega_0^\circ 2\vec{l}_t \cdot \vec{s}\mu(\vec{l}_t{}^2\langle 1_t{}^2\rangle_N),$$

and the modified parameters corresponding with the $A = 100$–110 mass region[21,34] $\kappa_P = 0.069$, $\mu_P = 0.45$, $\kappa_N = 0.066$, $\mu_N = 0.35$ (full line in Fig. 4). The macroscopic part of the energy was obtained using the liquid-drop formula with Meyers-Swiatecki parameters.[35] The pairing strength as well as the number of levels taken into account in the pairing calculations have been discussed by

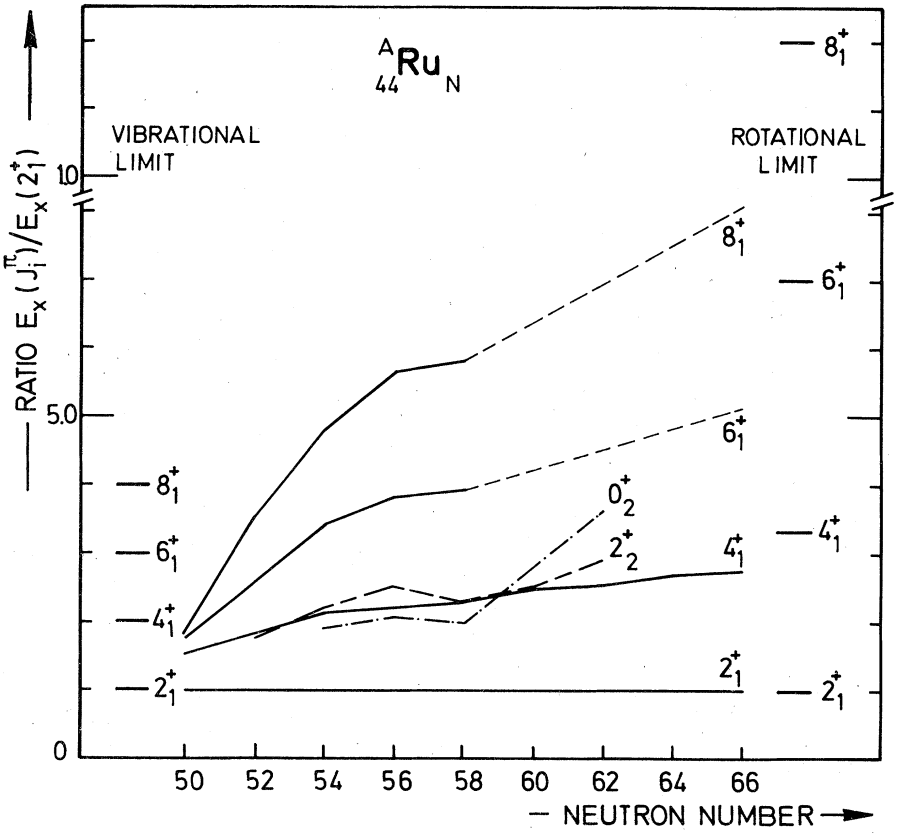

FIG. 3. The energy ratio $E_{J_i^\pi}/E_{2_1^+}$ for the $J_i^\pi = 4_1^+, 2_2^+, 0_2^+, 6_1^+$, and 8_1^+ levels for the doubly even Ru isotopes with $94 \leq A \leq 110$ (if available). Both the vibrational and rotational limit for this ratio are given on the extreme left and extreme right of the figure, respectively.

Ragnarsson[34] and Heyde *et al.*[21] We have also studied the influence of small variations of the most sensitive parameters, i.e., $\mu_P = 0.40$, $\mu_N = 0.30$, same κ_P, κ_N (dashed line in Fig. 4); only relatively small changes result. From these potential-energy surfaces, anharmonicities are clearly established which, however, due to the zero-point oscillator motion $\frac{5}{2}\hbar\omega_2$, will not lead to stable quadrupole deformation effects. The anharmonicities as observed in the experimental spectrum (see Fig. 5) are thus clearly in line with the results from the potential-energy calculations. Although this is a purely qualitative argument these anharmonicities can be discussed by treating explicitly a higher-order collective quadrupole Hamiltonian. Many attempts have thus far been undertaken, and one of the most successful approaches stems from the interacting boson model (IBA) making simple exact solutions possible in the vibrational and rotational limit.

The IBA Hamiltonian in the vibrational limit reads

$$H = \epsilon \sum_m d_m^\dagger d_m + \sum_L c_L [(d^\dagger d^\dagger)^L (dd)^L]^0 ,$$

with

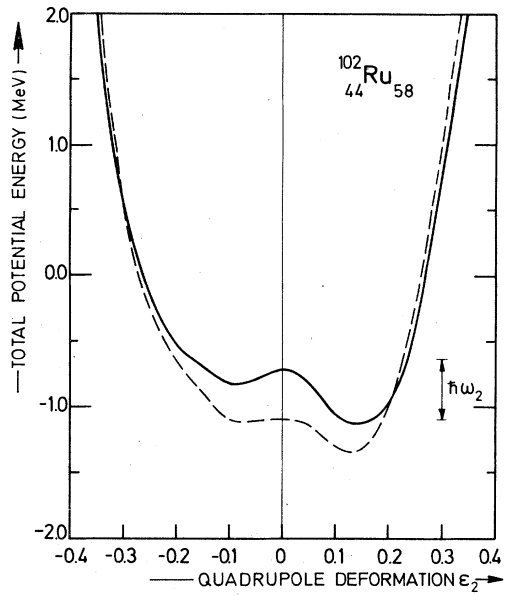

FIG. 4. The total potential-energy curves for ^{102}Ru corresponding to the quadrupole degree of freedom. The Nilsson modified harmonic-oscillator-model parameters are, respectively, $\kappa_P = 0.069$, $\kappa_N = 0.066$, $\mu_P = 0.45$, $\mu_N = 0.35$ (full line) and $\mu_P = 0.40$, $\mu_N = 0.30$ for the same κ_P and κ_N values (dashed line—see text).

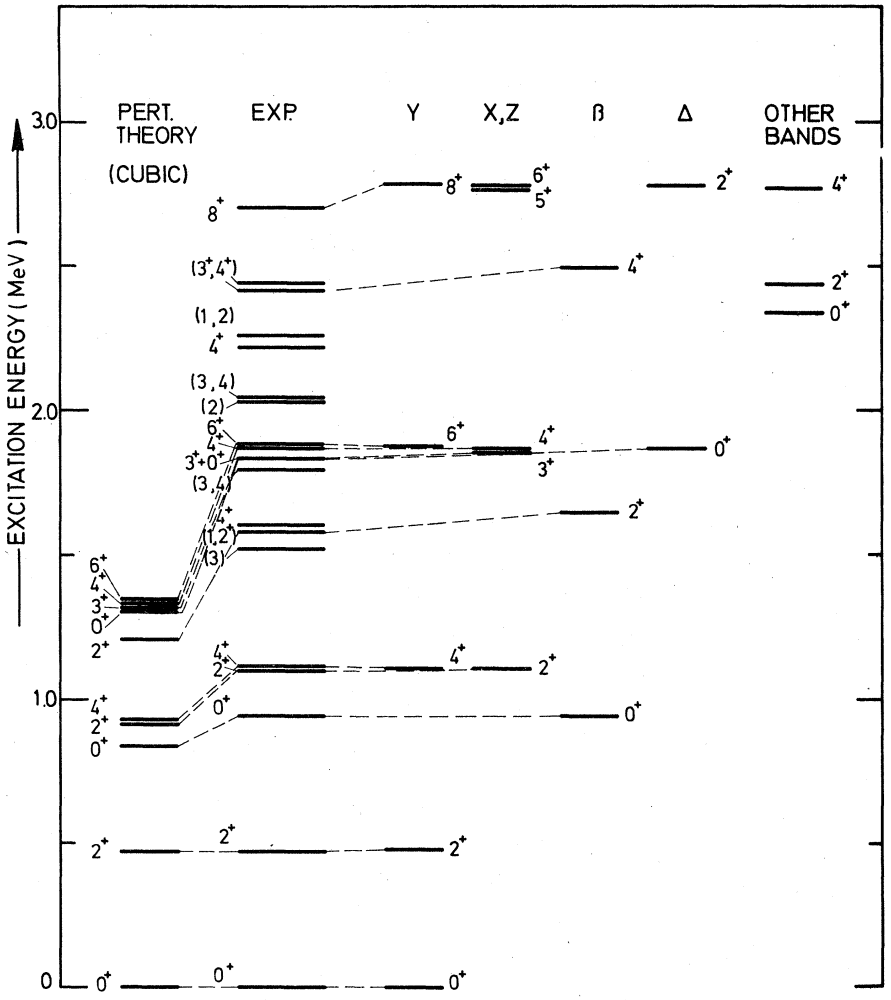

FIG. 5. Comparison between the experimental spectrum of ^{102}Ru, the calculation within the IBA, and a perturbation theory calculation using a collective Hamiltonian with cubic anharmonicities. In the IBA, the different bands (Y, X, Z, β, Δ, etc.) are drawn separately to facilitate comparison with experiment.

$$E(n_d, v, n_\Delta, L, M) = \epsilon n_d + \alpha \frac{n_d(n_d-1)}{2} + \beta(n_d - v)(n_d - v + 3) + \gamma[L(L+1) - 6n_d]$$

as eigenvalue, where n_d, v, n_Δ, L, and M denote the number of quadrupole bosons, the seniority, the quantum number counting the boson triplets coupled to zero angular momentum, the angular momentum, and its Z projection, respectively. The relation between (α,β,γ) and (c_0, c_2, c_4) is discussed extensively by Arima and Iachello.[25]

In the specific case of ^{102}Ru we obtain a best fit for $\epsilon = 481$ keV, $\alpha = 142.28$ keV, $\beta = -15.77$ keV, $\gamma = 0.214$ keV. It is shown in Fig. 5 where the theoretical levels are classified in the different bands (Yrast, β-band, etc.). The agreement is striking although some experimental levels remain unexplained. However, since vibrational nuclei exhibit other excitation modes such as two quasiparticle and collective octupole modes, special care must be taken in comparing with experiment.

The importance of the fourth-order anharmonicities as used in the IBA can be studied if the collective Hamiltonian discussed by Kerman and Shakin,[27] containing up to cubic terms, is used. It reads

$$H = H_{\text{harmonic}} - \frac{BB'}{\alpha'}\left(\tfrac{35}{2}\right)^{1/2} \sum_{\alpha\beta\gamma} \begin{pmatrix} 2 & 2 & 2 \\ \alpha & \beta & \gamma \end{pmatrix} \dot{\alpha}_\alpha \alpha_\beta \dot{\alpha}_\gamma + \frac{h\omega C'}{(\alpha')^3}\left(\tfrac{35}{2}\right)^{1/2} \sum_{\alpha\beta\gamma} \begin{pmatrix} 2 & 2 & 2 \\ \alpha & \beta & \gamma \end{pmatrix} \alpha_\alpha \alpha_\beta \alpha_\gamma ,$$

with $\omega = (C/B)^{1/2}$, $\alpha' = (\hbar\omega/C)^{1/2}$, and $\alpha = C'/B'$. The best agreement with ^{102}Ru, for a calculation up to second-order perturbation theory, has been obtained for $(B')^2 = 0.06$ and $\alpha = 0.01$. In this calculation, we made an exact fit for the $J^\pi = 2_1^+$ level. One observes, however, for the two and three quadrupole phonon states, a theoretical excitation energy that is too low compared with the experiment, although the ordering in each multiplet is in good agreement with the experiment and with the IBA.

Other possibilities for testing the anharmonicities of the experimental vibrational spectrum can be obtained by considering reduced $E2$ transition probabilities and comparing them with the pure vibrational intensity limit. If one reduces the experimental ratios from the $J_i^\pi = 2_2^+, 2_3^+, 0_3^+$ levels to ratios of reduced $E2$ transition probabilities one obtains

$$\frac{B(E2; 2_2^+ \to 0_1^+)}{B(E2; 2_2^+ \to 2_1^+)} = 0.029 \pm 0.008 , \tag{1}$$

$$\frac{B(E2; 2_3^+ \to 2_1^+)}{B(E2; 2_3^+ \to 0_2^+)} = 0.125 \pm 0.03 , \tag{2}$$

$$\frac{B(E2; 0_3^+ \to 2_1^+)}{B(E2; 0_3^+ \to 2_2^+)} = 0.13 \pm 0.09 , \tag{3}$$

as a measure of the anharmonicities producing nonzero $\Delta N = 2$ $E2$ transitions. The values in (1) and (2) are lower and upper limits, respectively, because of the possibility of non-negligible $M1$ contributions in the $2_2^+ \to 2_1^+$ and $2_3^+ \to 2_1^+$ transitions. The ratio (1), calculated with the best fit parameters α and $(B')^2$, is 0.029 in very good agreement with the experimentally observed value.

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