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NEUTRON CAPTURE GAMMA-RAY SPECTROSCOPIC MEASUREMENTS IN THE ACTINIDE REGION\*

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ABSTRACT. From recent neutron capture gamma-ray measurements, experimental data for states involving quasiparticle-vibrational admixtures in  $^{227}\text{Ra}$ ,  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{237}\text{U}$ , and  $^{239}\text{U}$  have been compared with theoretical calculations by Soloviev's group. This analysis shows the experimental level structure is more complex than that calculated. In the levels of  $^{250}\text{Bk}$ , four Gallagher-Moszkowski pairs are observed. The moment of inertia for each band with antiparallel alignment of odd-nucleon momenta is systematically larger than for its parallel-aligned mate.

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A general review of nuclear structure of the odd-mass nuclei in the actinide region has been published by Chasman *et al.* (1977). The results of neutron capture gamma-ray spectroscopic measurements in actinide nuclei have been summarized by von Egidy (1979a). The intent of this paper is to review recent neutron capture spectroscopic measurements, mostly those published since the 1978 conference at Brookhaven National Laboratory, and, for selected actinide nuclei, to make detailed comparisons between experimental level structures and model calculations, especially those that address the question of mixed quasiparticle-vibrational excitations.

The recently-published results of neutron capture gamma-ray and conversion-electron measurements involve the study of excited levels in both odd-neutron nuclei,  $^{227}\text{Ra}$  (von Egidy 1981),  $^{231}\text{Th}$  (White 1979),  $^{233}\text{Th}$  (Jeuch 1979),  $^{235}\text{U}$  (Almeida 1979),  $^{237}\text{U}$  (von Egidy 1979b),  $^{239}\text{U}$  (Börner 1978), and  $^{249}\text{Cm}$  (Hoff 1981a), and odd-odd nuclei,  $^{232}\text{Pa}$  (Hoff 1981b),  $^{238}\text{Np}$  (Ionescu 1979),  $^{244}\text{Am}$  (Hoff 1981d), and  $^{250}\text{Bk}$  (Hoff 1981e). For all of these nuclei, secondary gamma rays have been measured at the ILL by use of the GAMS spectrometers. For most, conversion electron data have also been taken with the BILL spectrometer at Grenoble; exceptions are  $^{227}\text{Ra}$  and  $^{249}\text{Cm}$ . A particularly important set of studies are those of levels in  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{239}\text{U}$ ; these odd-neutron nuclei are closely related with neutron number varying in a narrow range,  $N = 141$  to  $147$ . What sets this group of measurements apart is that primary gamma-ray transitions from average resonance capture have been measured at Brookhaven National Laboratory for each. These results, combined with the GAMS and BILL data, provide an essentially complete measurement of the low-spin levels,  $I^\pi = 1/2^-, 3/2^-, 1/2^+, 3/2^+$ , and  $5/2^+$ , in these nuclei (Casten 1980).

The  $^{227}\text{Ra}$  paper (von Egidy 1981) presents a detailed level scheme of this nucleus for the first time. Rotational bands built on seven different quasiparticle excitations were observed. Two levels, tentatively assigned as  $[770^+]$ , are the first experimental observation of this configuration. The decoupling constant for the  $[631^+]$  band is positive,  $a = +0.6$ , a continuation of the trend with decreasing  $A$ ; theoretical calculations suggest a negative value for the decoupling constant, as found experimentally at higher mass numbers. Possible bands with octupole vibrational mixing have been assigned in  $^{227}\text{Ra}$ ; these are reviewed later in our discussion of octupole vibrations.

In the  $^{237}\text{U}$  investigation (von Egidy 1979b), members of rotational bands assigned to six Nilsson states have been identified. A  $K^\pi = 1/2^+$  band is observed at 847 keV. For this, persuasive arguments can be made for a configuration assignment of  $\{[622^+]-2^+\} + [620^+]$ . The experimental evidence includes strong population by the  $(d, p)$  reaction and rotational parameters similar to those found for this configuration in neighboring nuclei. The 687-keV spacing between this band and its  $[622^+]$  base state is the same as in  $^{235}\text{U}$  and  $^{239}\text{U}$ . A second  $K^\pi = 1/2^+$  band at 906 keV has been assigned  $\{[631^+]+0^+\}$  character, based largely upon its excitation above the base state and rotational parameters. The energy of this configuration fits very well in a series of observations that include  $^{235}\text{U}$  and  $^{239}\text{U}$ ; for these latter measurements, the experimentally-observed  $E_0$  components of the depopulating transitions are indicative of  $K^\pi = 0^+$  vibrational admixtures. Several unexpected levels have been observed in  $^{237}\text{U}$  below 800 keV. Of these, two  $K^\pi = 1/2^-$  bands are proposed to include octupole vibrations based upon the  $[631^+]$  and  $[622^+]$  states.

In the  $^{249}\text{Cm}$  study (Hoff 1981a), new quasiparticle bands assigned were  $[622^+]$ , 530 keV,  $[752^+]$ , 773 keV, and  $[501^+]$ , 917 keV. Each of these appears experimentally at energies much below those calculated. Much of this energy decrease appears to be due to mixing of the quasiparticle state with vibrational components. A definitive measure of these admixtures will require conversion electron data. It is interesting that the  $[501^+]$  state, a hole configuration in all of the actinide nuclei and one with strongly uploping energy in a Nilsson diagram, has been observed at excitation energies below 1 MeV in fifteen nuclei, from  $^{227}\text{Ra}$  ( $N = 139$ ) to  $^{249}\text{Cm}$  ( $N = 153$ ).

Collective excitations in the form of quadrupole and octupole vibrations have been well-studied experimentally in even-even actinide nuclei. Levels corresponding to single-phonon excitation of quadrupole vibrations with  $K^\pi = 0^+$  and  $2^+$  and of octupole vibrations with  $K^\pi = 0^-$  have been assigned in many of the even-even thorium, uranium, plutonium, and curium nuclei. The energies of these vibrational levels are shown in Figs. 1, 2, and 3 for selected thorium and uranium nuclei. In odd-mass actinide nuclei, one finds vibrational excitations that involve collective excitation of the even-even core and that are coupled to the various intrinsic states of the odd nucleon. An odd-mass nucleus exhibits three distinct forms of excitation: a) quasiparticle excitations of the odd-nucleon, b) vibrational excitations built upon various quasiparticle excitations, and c) rotational bands built upon both quasiparticle and vibrational excitations. It is necessary to consider interactions between these forms of excitation. Thus, the nonrotational excitations of an odd-mass nucleus can be described in terms of a mixture of quasiparticle and vibrational states. We will review the experimental data for excited levels in  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{239}\text{U}$  for information regarding these admixtures; some information on this question has been presented in an earlier publication (von Egidy 1979c).

In previous experimental studies, vibrational states in odd-mass actinides were determined by means of Coulomb excitation studies ( $K^\pi = 2^+$  vibrations coupled to the ground state quasiparticle configuration), by means of the (d,d') reaction ( $K^\pi = 0^-$  vibrations coupled to the g.s. quasiparticle configuration), by means of the (p,t) reaction ( $K^\pi = 0^+$  vibrations), and certain decay scheme studies ( $K^\pi = 2^+$  vibrations). Chasman et al. (1977) list 19 vibrational state assignments in odd-neutron actinide nuclei, including four in  $^{235}\text{U}$ . In the (n, $\gamma$ ) experiments being discussed in this paper, the presence of quadrupole and octupole vibrational admixtures is deduced from experimental information on the internal conversion coefficients of de-exciting transitions and the predominant de-excitation paths for various levels.

Recent (n, $\gamma$ ) investigations have produced evidence for appreciable E0 strength in transitions de-exciting levels in  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{239}\text{U}$ . These transitions have conversion coefficients that are markedly larger (as much as 5 to 10 times) than theoretical M1 (or E2 or E1) coefficients. This E0 strength is ascribed to the presence a  $K^\pi = 0^+$  vibrational admixture in the wave function of the upper state. This phonon excitation is coupled to the single-particle state most predominant in the wave function of the lower level. In an analogous way, the observation of intense E2 transitions de-exciting bands to predominantly single-particle states is a manifestation of a quadrupole,  $K^\pi = 2^+$ , vibrational admixture in the wave function of the parent level.

Another type of vibrational admixture, octupole vibrations with  $K^\pi = 0^-$ , has been identified in the levels of these odd-mass nuclei. Octupole states in even-even nuclei have been characterized by large BE3 matrix elements between the ground state and vibrational level. Although the corresponding BE3 matrix elements should have large values in odd-mass nuclei, it is usually likely that the more rapid E1 transitions will be predominant in the transitions between a state containing octupole phonon-quasiparticle coupling and its corresponding base state. This third type of de-excitation which identifies the vibrational admixtures in the parent state is more difficult to distinguish unequivocally in secondary gamma transitions following neutron capture than either of the first two.

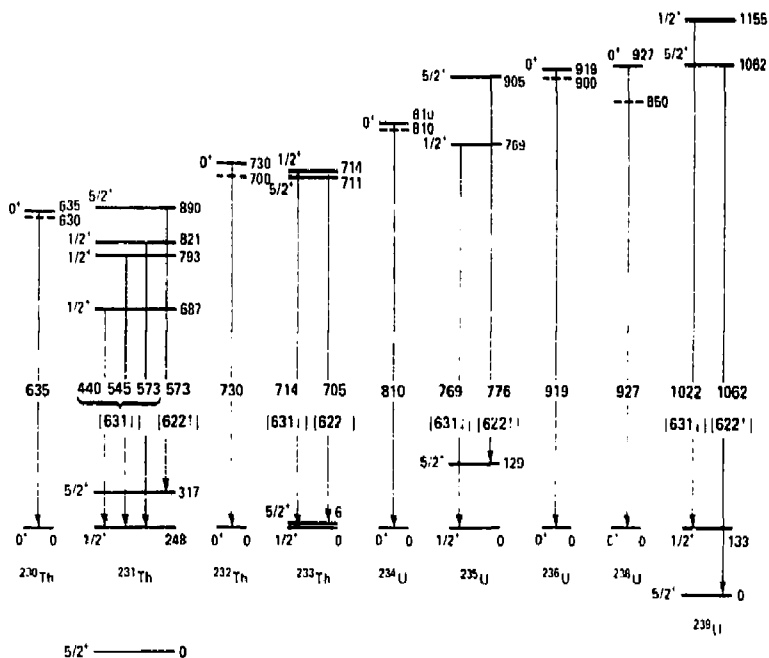


Fig. 1. Experimental levels in  $^{230}\text{Th}$ - $^{239}\text{U}$ ;  $K^\pi = 0^+$  phonon vibrations.

Of those transitions in  $^{231}\text{Th}$ ,  $^{233}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{239}\text{U}$  that exhibit distinct EO strength, the majority are interpreted as de-excitation modes of levels involving  $K^\pi = 0^+$  vibrational components coupled to just two low-lying single-particle states,  $[631^+]$  and  $[622^+]$ . The pattern of observation has been to find EO strength in transitions depopulating the  $I = 1/2$  and  $3/2$  (and sometimes  $5/2$ ) levels of the  $\{[631^+]+0^+\}$  configuration and in transitions depopulating the  $I = 5/2$  level of the  $\{[622^+]+0^+\}$  configuration. The selectivity of the  $(n,\gamma)$  reaction with even-even targets limits these observations to levels with  $I \leq 7/2$ . Excluding  $^{231}\text{Th}$  from this discussion for the moment, the energy spacings for these vibrational configurations above the corresponding single particle states range from  $\sim 700$  keV in  $^{233}\text{Th}$  to  $\sim 1060$  keV in  $^{239}\text{U}$ . These energies compare favorably with the energies of the first-excited  $0^+$  states in neighboring even-even nuclei, as shown in Fig. 1. A clear trend of decreasing excitation energy with

decreasing neutron number is seen. In  $^{231}\text{Th}$ , we find the  $\{[631+]+0^+\}$  configuration exists with observable strength in three  $K^\pi = 1/2^+$  bands at 687, 793, and 821 keV. This is in distinct contrast to the situation in  $^{233}\text{Th}$ ,  $^{235}\text{U}$ , and  $^{239}\text{U}$  where only one state with these characteristics has been found for each nucleus.

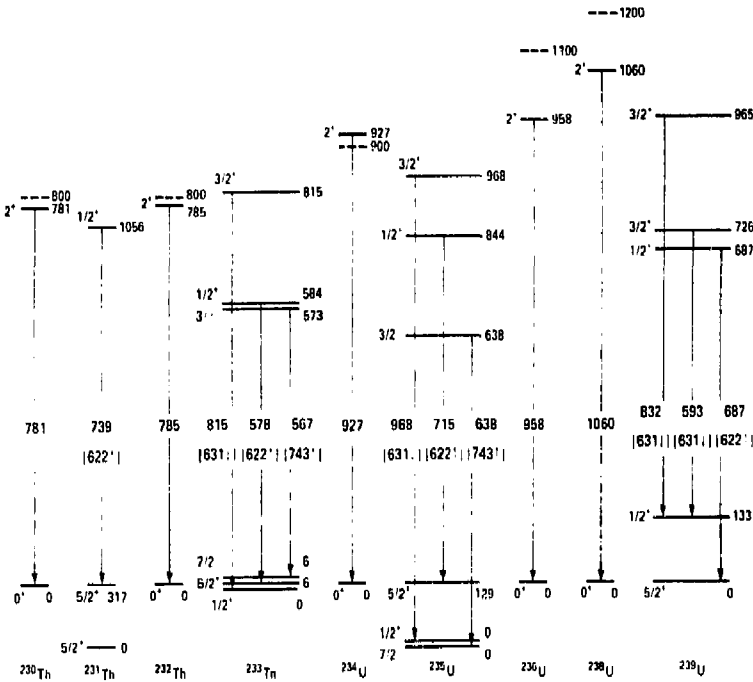


Fig. 2. Experimental levels in  $^{230}\text{Th}$ - $^{239}\text{U}$ ;  $K^\pi = 2^+$  phonon vibrations.

Experimental data for vibrational states with admixtures of  $K^\pi = 2^+$  phonons coupled to quasiparticle configurations are given in Fig. 2. The observed trend of energy spacing for the first excited  $K^\pi = 2^+$  level in even-even nuclei is that of decreasing energy with decreasing neutron number, as for the  $K^\pi = 0^+$  levels. The quasiparticle states involved in gamma-vibrational coupling in the odd-mass nuclei are three,  $[631^+]$ ,  $[622^+]$ , and  $[743^+]$ . The energies of gamma vibrational states range from 575 to 970 keV above the base states. A 638-keV level in  $^{235}\text{U}$ , already assigned vibrational character in studies of Coulomb excitation and the  $(d, d')$  reaction, is observed to decay predominantly via an E2 transition to the  $[743^+]$  configuration, confirming the previous configuration assignment,  $\{[743^+]-2^+\}$ . There appears to be no very clear trend of energy spacing of gamma-vibrational states with mass number for the nuclei shown in Fig. 2.

Octupole vibrational states with  $K^\pi = 0^-$  phonons are summarized in Fig. 3. For the even-even nuclei, energies of the  $I^\pi = 1^-$  levels of the  $K^\pi = 0^-$  octupole bands are shown. This state occurs at rather low energies in these nuclei; for the even-even nuclei in Fig. 3, the  $I^\pi = 1^-$  level lies

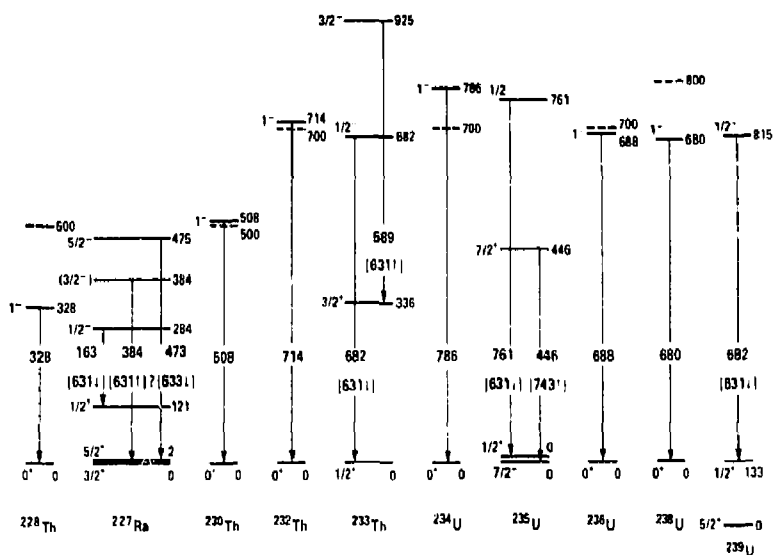


Fig. 3. Experimental levels in  $^{228}\text{Th}$ - $^{239}\text{U}$ ;  $K^\pi = 0^-$  phonon vibrations.

lower than either the first  $K^\pi = 0^+$  excited state or the first  $K^\pi = 2^+$  state. The smallest energy spacings for  $K^\pi = 0^-$  vibrational admixtures are observed in the level scheme of  $^{227}\text{Ra}$ . The octupole state built on the  $[631+]$  configuration is observed in all of the odd-neutron nuclei shown in the figure. The 446-keV level in  $^{235}\text{U}$  has been identified in an earlier study of the  $(d,d')$  reaction as having a  $\{[743^+]+0^-\}$  configuration. No clear evidence has been found yet for states with octupole vibrational admixtures in  $^{231}\text{Th}$ .

A number of theoretical calculations of vibrational state energies in even-even actinide nuclei have been reported (Ragnarsson 1976, Griffin 1971). The results of Soloviev (1976) are shown as dashed lines in Figs. 1-3. The agreement between experiment and theory is generally good; the average deviation is approximately 50 keV. For odd-mass nuclei, we can compare the experimental data with the calculational results of the Soloviev group (Gareev 1971, Ivanova 1975, Komov 1971); these are the only published calculations of quasiparticle-phonon coupled states in odd-mass thorium and uranium nuclei of which we are aware.

For these calculations, the interaction of quasiparticles with quadrupole and octupole phonons in deformed odd-mass nuclides was considered in the framework of the method of approximate second quantization. Pairing and multipole-multipole interactions have been included. As an assumption for the basic model, the phonons of the odd nucleus  $A + 1$  have been assumed to be identical to the phonons of the even-even nucleus  $A$ . The quasiparticle and phonon operators were treated as fully-determined entities. The values for multipole-multipole interaction constants were taken from studies of even-even nuclei. Thus, there were no free parameters in this analysis of odd- $A$  nuclei. A number of modifications to these basic assumptions were made to improve the model; these included a) an approximate treatment of the blocking effect improved the description of energies; b)





at 815 keV and assigned a  $\{[631+] + 0^-\}$  configuration, correlates well with a theoretical level at 700 keV that is calculated to be largely collective, 71%  $\{[631+] + 0^-\}$ .

While for  $^{235}\text{U}$  and  $^{233}\text{Th}$  we will not make a detailed comparison between theory and experiment, it is clear that many of the experimental features are not predicted by the calculations. The two  $K^\pi = 0^+$  vibrational bands that have been observed in both of these nuclei,  $\{[631+] + 0^+\}$  and  $\{[622+] + 0^+\}$  (see Fig. 1), generally do not appear in the calculations of vibrational admixtures for  $^{235}\text{U}$  and  $^{233}\text{Th}$ . An exception is a  $K^\pi = 1/2^+$  band at 1030 keV in  $^{235}\text{U}$  with the configuration,  $\{[622+] - 2^+\}$  37% and  $\{[631+] + 0^+\}$  22%. The corresponding experimental bands appear separately with the  $K^\pi = 0^+$  vibrational state at 769 keV and the  $K^\pi = 2^+$  vibrational state at 844 keV. Other vibrational configurations identified in  $^{235}\text{U}$  such as  $\{[743+] - 2^+\}$  at 638 keV and  $\{[631+] + 0^-\}$  at 761 keV have energies that correspond to theoretical bands with the appropriate value of  $K^\pi$ . On the other hand, the calculated configurations include very little of the observed vibrational admixture (<5%). In  $^{233}\text{Th}$ , the correspondence between the three observed bands with  $K^\pi = 2^+$  vibrational admixtures (Fig. 2) and calculated level structure is generally poor. The most significant calculated band with gamma vibrational character, one at 1080 keV which is almost entirely collective, 95%  $\{[631+] - 2^+\}$ , has its experimental counterpart at 815 keV; since the transitions de-exciting the experimental band are M1 + E2 mixtures with appreciable amounts of M1 multipolarity, the collectivity of this band may be somewhat less than that of the calculation.

In interpreting the results from our experimental study of  $^{231}\text{Th}$  level structure (White 1979), we have made configuration assignments by comparing experimental level energies, spins, and parities with the model calculations of Ivanova *et al.* (1975). Of the rotational bands below 630 keV, good agreement between theory and experiment ( $\Delta E < 80$  keV) is found for many of the bandhead energies. The largest discrepancy is the observation of the  $[622+]$  bandhead at 470 keV below its predicted energy (see Fig. 5). The evidence for the existence of levels in this rotational band is quite satisfactory and includes population of levels at 317, 380, and 449 keV by the (d,p) reaction.

We have already mentioned the three  $K^\pi = 1/2^+$  bands at 687, 793, and 821 keV which have some component of  $\{[631+] + 0^+\}$  in their wave functions. For  $^{231}\text{Th}$ , the calculations predict just one  $K^\pi = 1/2^+$  state at 610 keV that is above the  $[631+]$  band (at 247 keV) and below 1020 keV. This state is reported to consist of quasiparticle components,  $[640+]$  50%,  $[631+]$  5%,  $[620+]$  1%,  $[651+]$  1%, and a vibrational component,  $\{[640+] + 0^+\}$  30%. Since no mention is made of the fraction of  $\{[631+] + 0^+\}$  configuration in this state, we can assume it is less than 13%.

Since these observed  $K^\pi = 1/2^+$  bands (and a fourth at 1056 keV) are not predicted by the calculations, it is of interest to consider the nature of these states. In order to derive the proportion of E0 strength for the transitions leading to the  $[631+]$  band, each transition will be assumed to consist of an M1+E0 mixture, neglecting any E2 component. On this basis, the transitions depopulating the 687-keV band contain 10-20% E0 admixture; for the 793-keV band, the E0 admixture is 50%; and for the 820-keV band, the E0 admixture is 40%. All but one of these new  $1/2^+$ ,  $3/2^+$  levels decay to the ground-state level,  $[633+]$ . The presence of a  $\{[633+] - 2^+\}$  configuration is indicated by E2 strength observed in the transitions from these





$1/2[620]_n$  bandhead at 115.4 keV (as compared with 131.9 keV before) and defines a new level at 148.6 keV as the  $I = 4$  member of that rotational band. A new rotational band is defined by two new levels at 146.5 and 180.0 keV; it is assigned the configuration,  $3/2[521]_p - 7/2[613]_n$ . We have extended the ground-state rotational band by adding  $I = 4$  and 5 members at 80.3 and 137.3 keV, respectively. We have extended the rotational band at 103.8 keV by adding  $I = 3$  and 4 members at 157.4 and 203.6 keV, respectively. A new rotational band is defined by two new levels at 316.5 and 369.6 keV; it is assigned the configuration,  $7/2[633]_p + 3/2[622]_n$ . We assign a new level at 215.9 keV to the  $I = 0$  member of the  $7/2[633]_p - 7/2[613]_n$  rotational band.

Since the level energies of  $^{250}\text{Bk}$  are known with much greater precision than before, the experimentally-measured moment of inertia for each rotational band can be compared with a calculated moment which is derived by adding to the moment of inertia of the even-even core an incremental increase for each of the odd nucleons, as determined from experimental data for odd-mass nuclei. In Fig. 7a, the experimental data are plotted in the form of a value for the rotational parameter derived from each level spacing versus  $I^2$ . The calculated rotational parameters are shown as dashed lines. Agreement between experiment and calculation is good. The data shown are for combinations involving  $3/2[521]_p$ ,  $1/2[620]_n$ , and  $7/2[613]_n$  configurations. Experimental data for these same configurations in certain odd-mass nuclei are shown in the upper part of Fig. 7a.

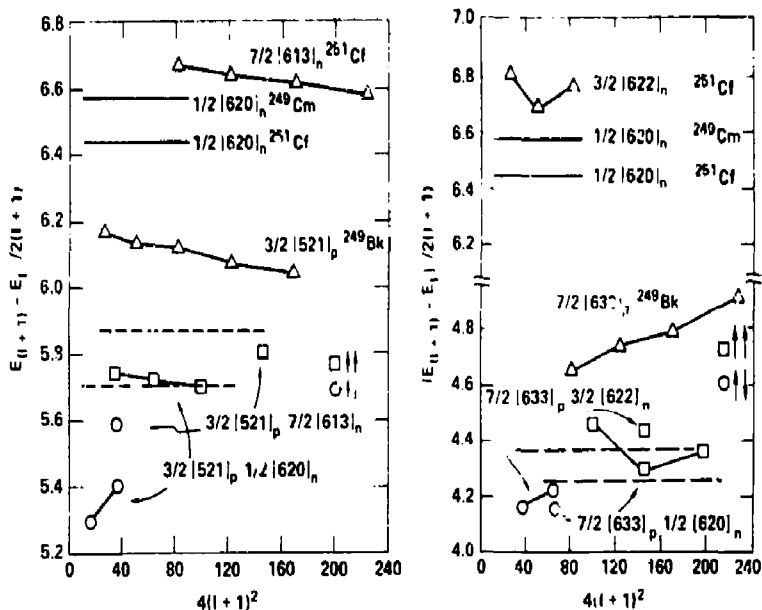


Fig. 7. Rotational parameters for single-particle configurations in  $^{250}\text{Bk}$  and associated odd-mass nuclei.

Corresponding data for a different proton state,  $7/2[633]$ , are shown in Fig. 7b. Again, there is good agreement between experiment and calculation. Since the  $7/2[633]$  proton state exhibits a low value for the

rotational parameter in  $^{249}\text{Bk}$ , configurations in  $^{250}\text{Bk}$  involving this proton state can be identified by a similar low value ( $A = 4.1-4.5$  keV). This is in distinct contrast to the configurations involving the  $3/2[521]$  proton state. Here, the larger value of the rotational parameter,  $\sim 6.2$  keV, when combined in various configurations with the odd neutron orbitals, results in rotational parameters for odd-odd configurations in  $^{250}\text{Bk}$  with rotational parameters in the range,  $A = 5.3-5.8$  keV. Thus, it is possible to make a straightforward experimental determination of the proton state in all low-lying rotational bands in  $^{250}\text{Bk}$ . The  $3/2[521]$  and  $7/2[633]$  proton orbitals are very close in energy in the Bk isotopes; the next higher proton states are  $5/2[642]$  and  $1/2[400]$  which also lie close in energy at about 330-340 keV above the first two states. Rotational bands with an odd proton in these latter states have not been identified in  $^{250}\text{Bk}$  yet.

From the results presented in Figs. 7a and 7b, we see what appears to be a systematic difference in rotational parameters between two configurations involving the same states for the odd nucleons. The rotational band with antiparallel alignment of the odd nucleons' angular momenta has a larger moment of inertia than for the corresponding band with parallel alignment. This observation holds true for the four pairs of configurations shown in Fig. 7. The general expectation has been that the moments of inertia should be the same for each of two configurations with identical odd nucleon states. Interactions such as the Coriolis force can affect the values for rotational parameters as already mentioned in our discussion of the  $7/2[633]$  proton state. Nevertheless, the effect we are considering appears quite clearly in Fig. 7a where the single-particle states involved are known not to exhibit appreciable Coriolis mixing. In other studies of odd-odd nuclei, e.g. our investigation of  $^{238}\text{Np}$  levels (Hoff 1981c), we have not seen differences in rotational parameter values for bands that are configuration pairs. Thus, it is not clear whether this is a general phenomenon or just peculiar to the  $^{250}\text{Bk}$  level scheme.

Among the configurations assigned to levels in  $^{250}\text{Bk}$  are four Gallagher-Moszkowski pairs. The energy splitting between these pairs of rotational bands is determined by the strength of the p-n residual interaction. We compare experimental and theory in Table 1. The theoretical calculations were made assuming a  $\delta$  force with a scale factor of unity for the residual interaction (Piepenbring 1978). If a scale factor of 0.75 is applied, the experimental results for the first three configuration pairs in Table 1 would be more consistent with theory. The greatest discrepancy appears for the  $7/2[633]_p-7/2[613]_n$  configuration. It is the antiparallel,  $K = 0$ , rotational band whose characteristics are most uncertain and which deserves further experimental study. The last entry in Table 1, the Newby shift, is defined as the energy offset between even-spin and odd-spin members of a  $K = 0$  rotational band.

Table 1. Experimental and calculated Gallagher-Moszkowski splittings in  $^{250}\text{Bk}$ .

Proton configuration	Neutron configuration	GM splitting (keV)		
		Exp.	Calc.	Exp./Calc.
$3/2[521]$	$1/2[620]$	110	174	0.63
$3/2[521]$	$7/2[613]$	66	91	0.72
$7/2[633]$	$1/2[620]$	85	91	0.93
$7/2[633]$	$7/2[613]$	126	71	1.79
Newby shift		-32	-88	0.36

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