Application of the multiphonon method to the study of the vibrational states with $K=0$ in heavy deformed nuclei

J. Leandri and R. Piepenbring
Institut des Sciences Nucléaires, 38026 Grenoble, France
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The multiphonon method based on $K^\pi=0^-$ and $K^\pi=0^+$ phonons is applied to heavy mass deformed even nuclei, such as Th, U, and Pu isotopes. Special emphasis is put on the location and properties of the "two phonon" states. Relative to an harmonic situation, the calculated energy spectra are systematically dilated. The octupole vibrations with $K^\pi=0^-$ present larger anharmonicities than the vibrations $K^\pi=0^+$ which, in these nuclei, are found to be mainly of pairing nature. The two modes appear to be weakly coupled. The "two phonon" states are predicted to have an energy of about 1.7 to 2.0 MeV and keep some collective character through their electromagnetic transitions to the "one phonon" states. These conclusions are at variance with those of the quasiparticle phonon nuclear model of Soloviev *et al.*

I. INTRODUCTION

In even-even deformed actinides, intrinsic states with $K=0$ appear at an energy smaller than the two quasiparticle gap. Those with negative parity are known to arise from octupole correlations which play an important role in these nuclei, while those with positive parity may be due to pairing and/or quadrupole interactions.

In even Ra and Th, with mass number $222 \leq A \leq 228$, the lowest level with $K^\pi=0^-$ appears very low in energy and well separated from the first excited $K^\pi=0^+$ state. The multiphonon method (MPM), using only the $K^\pi=0^-$ octupole phonon as the building block (hereafter noted as MPM1), has been successfully applied[1] to these nuclei. Using a basis with reflection symmetry it was possible, among others, to explain the lack of $K^\pi=0^+$ two phonon states of octupole nature in the region of twice the energy of the one phonon state $K^\pi=0^-$.

In heavier isotopes of Th, in U and Pu the first excited $K=0$ states of both parities appear with energies of the same order. The quadrupole, octupole, and pairing correlations are therefore to be treated, *a priori*, on an equal footing. In the multiphonon method one has therefore, to introduce two building phonons characterized, respectively, by the quantum numbers $K^\pi=0^-$ and $K^\pi=0^+$. Such a version of the multiphonon method (hereafter noted as MPM2) has been tested[2] in a simple model allowing an exact solution. Furthermore, an application to the realistic case of ^{234}U has been published.[3] It allowed us to study the coupling of the two $K=0$ modes with different parities and the anharmonicities of the corresponding vibrations in the nucleus. It was found that the anharmonicities obtained in ^{234}U were smaller than in the light Ra and Th isotopes studied[1] within the MPM1, and that these anharmonicities are not significantly altered by the coupling with the $K^\pi=0^+$ mode.

In the present study we apply the MPM2 to a series of Th, U, and Pu isotopes with $230 \leq A \leq 240$. A special emphasis is put on the properties of the first members 0_1^+, 0_1^-, 0_2^+, 0_3^+, 0_2^-, and 0_4^+ of the family of the intrinsic levels with $K=0$.

The general aim of this work is not to search for a fine agreement between the calculated and observed properties, but rather to study the general qualitative behavior of the lower collective vibrational states with $K=0$, when going from one nucleus to the next.

In Sec. II a brief summary of the principles of the multiphonon method[4] is given and the main ingredients of the MPM2 are recalled. Our results are discussed in Sec. III and compared to those of the quasiparticle phonon nuclear model (QPNM) of Soloviev *et al.*[5] in Sec. IV. Finally, some conclusions are drawn in Sec. V.

II. BRIEF SUMMARY OF THE MPM

The MPM is mainly an exact diagonalization of the *total* model Hamiltonian in the *collective* space spanned by some selected phonons. These are chosen of the Tamm-Dancoff type and appear as a superposition of two quasiparticles

$$Q_j^\dagger = \tfrac{1}{2} \sum_{mn} (X_j)_{mn} a_m^\dagger a_n^\dagger \ , \tag{1}$$

where the operators $a^\dagger$ and a are relative to fermions and where the matrix X_j has to be antisymmetric. Note that, with this choice, the phonon vacuum $|0\rangle$ is identical to the quasiparticle vacuum. The originality of the MPM lies in the fact that the full commutation rules

$$[Q_1, Q_2^\dagger] = -\tfrac{1}{2}\mathrm{Tr}(X_1 X_2) + \sum_{mn} (X_2 X_1)_{mn} a_m^\dagger a_n \ , \tag{2}$$

of the phonons (1), are taken into account. This means that the entities (1) are not approximated by bosons or quasibosons as in the Tamm-Dancofff approximation (TDA) or the random phase approximation (RPA). The multiphonon states

$$|a,b,c,\ldots\rangle = Q_1^{\dagger a} Q_2^{\dagger b} Q_3^{\dagger c} \ldots |0\rangle \tag{3}$$

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are then built on the phonons which are supposed to play an important role. In the version MPM2 the multiphonon states are simply given by

$$|pq\rangle=(p!q!)^{-1}Q_1^{\dagger p}Q_2^{\dagger q}|0\rangle\ , \tag{4}$$

where the $Q_1^\dagger$ and $Q_2^\dagger$ correspond each to a $K=0$ phonon, with positive parity when the index is 1, and negative parity when it is 2. The states (4) do not form an orthogonal basis. (To simplify the language we shall however, use this terminology within quotes in the forthcoming text.) The coefficient $(p!q!)^{-1}$ has been introduced in (4) to simplify some formulae obtained within the method. Since the Pauli principle is fully taken into account through the second term of Eq. (2), the overlap matrix of the states (4) and the matrix elements of the model Hamiltonian between them are not easy to calculate. It has been shown[4] that the introduction of a Wick's theorem generalized to phonons or the use of recursion formulae is required. In practical numerical situations the second choice is more convenient. The explicit expressions of the recursion formulae for the MPM2 can be deduced from the general formulae of Ref. 4 or rederivated directly. Since they can be found in earlier works[6,7] they are not given again here. It seems however, worthwhile to remind the reader that the matrix elements of any one- or two-body operator can be given in terms of the overlap matrix elements

$$F(p'q';pq)=\langle p'q'|pq\rangle\ , \tag{5}$$

the antisymmetric matrices

$$A_{mn}(p'q';pq)=\langle p'q'a_n a_m pq\rangle\ , \tag{6}$$

and the quantities

$$B_{stuv}(p'q';pq)=\langle p'q'a_v a_u a_t a_s pq\rangle\ , \tag{7}$$

all related through coupled recursion formulae. The main numerical task will therefore, be to calculate and to store, in the cheapest way, these quantities (5)–(7).

To apply the MPM2 in a realistic situation a model Hamiltonian H has to be chosen. Since the main aim of this work is to look for the general trends of the $K=0$ vibrations and not for a fine agreement between theory and experiment, we introduce the simplest H possible which takes into account the interactions known to play a major role in the region of nuclei under consideration. We therefore, describe the deformed heavy mass nuclei as being built of Z protons and N neutrons, moving in a deformed Nilsson potential, and interacting through a constant monopole pairing force and charge independent quadrupole-quadrupole and octupole-octupole forces in the following:

$$H=H_{\text{s.p.}}+H_P+H_Q+H_O\ , \tag{8}$$

where

$$H_{\text{s.p.}}=\sum_{mnj} e_m(j)C_m^\dagger(j)C_m(j)\ , \tag{9}$$

$$H_P=-\sum_j G_j L_j^\dagger L_j\ , \tag{10}$$

$$H_Q=-\tfrac{1}{2}\chi_2\sum_{ij} Q_i Q_j\ , \tag{11}$$

$$H_O=-\tfrac{1}{2}\chi_3\sum_{ij} O_i O_j\ , \tag{12}$$

with

$$\begin{aligned} L_j&=\sum_m C_{-m}(j)C_m(j)\ ,\\ Q_j&=\sum_{mn}\langle m(j)|R^2Y_{20}|n(j)\rangle C_m^\dagger(j)C_n(j)\ ,\\ O_j&=\sum_{mn}\langle m(j)|R^3Y_{30}|n(j)\rangle C_m^\dagger(j)C_n(j)\ . \end{aligned} \tag{13}$$

In these relations i and j refer to protons or neutrons whereas m and n label the single particle states. To treat the pairing, the canonical Bogoluybov-Valatin transformation is introduced to switch from the particle operators $C_m^\dagger$ and C_m to the quasiparticle operators $a_m^\dagger$ and a_m. The model Hamiltonian is then, as usual, separated in its different parts

$$H=H_{00}+H_{11}+H_{22}+H_{31}+H_{40}\ . \tag{14}$$

The explicit form of these different terms can be found in Ref. 7 and are not repeated here.

To reduce, as much as possible, the number of parameters, we fix the single particle potential parameters (κ,μ) of the Nilsson potential for the whole region as recommended by Lamm.[8] The quadrupole deformation parameter ϵ_2 is taken (but reduced to two digits), for each nucleus, from the tables of Löbner *et al.*[9] No higher order deformations are introduced. The intrinsic matrix elements of the R^2Y_{20} and R^3Y_{30} operators are evaluated according to the prescriptions of Boisson and Piepenbring.[10] The units are chosen so as to express the quadrupole χ_2 and the octupole χ_3 strength parameters in keV. The BCS gap parameters Δ_p and Δ_n are evaluated empirically from the experimental masses.[11] Their values may be obtained by use of several interpolation methods.[12–14] The spreading of the results indicate that there is a possible uncertainty on the evaluation of the Δ's of the order of ≈ 100 keV.

To limit, in a reasonable way, the number of quantities (6) and (7) to be calculated, one is forced to reduce, as much as possible, the number of active orbitals introduced in the treatment of the BCS equations. We used 20 active levels equitably distributed on each side of the Fermi surface. The sensitivity of the results to an enlargement of the single particle basis will be discussed in Sec. III. The pairing strength parameters G_j are then fixed through the usual gap equation of the BCS treatment.

To get the basic collective phonons one needs the antisymmetric matrices X_1 and X_2. These are obtained by solving the secular equations of the Tamm-Dancoff approximation for $K^\pi=0^-$ and $K^\pi=0^+$. In the first case, the secular equation has the familiar form

$$\chi_3\sum_{mnj}|\langle m(j)|R^3Y_{30}|n(j)\rangle|^2 f_{mn}^2(j)t_{mn}(j)=1\ , \tag{15}$$

where

$$t_{mn}(j)=[E_{mn}(j)-\omega]^{-1}\ ,$$

where

$$E_{mn}(j)=E_m(j)+E_n(j)$$

gives the energy of the two quasiparticle states and where $f_{mn}(j)=u_m(j)v_n(j)+u_n(j)v_m(j)$ is the usual pairing factor. In the second case, where $K^\pi=0^+$, the secular equation is more involved, due to the simultaneous effect of the pairing and quadrupole-quadrupole interactions. Since the corresponding generalized secular equation is not usual, we hereafter give its expression

$$2F(\omega)\chi_2=1 , \tag{16}$$

where

$$\begin{aligned}
&F=\sum_j [S_0(j)+S_1(j)S_u(j)+S_2(j)S_v(j)] ,\\
&S_0(j)=\tfrac{1}{2}\sum_{mn} |\langle m(j)|R^2Y_{20}|n(j)\rangle|^2 f^2_{mn}(j)t_{mn}(j) ,\\
&S_1(j)=\sum_m T_m(j)u^2_m(j) ,\\
&S_2(j)=\sum_m T_m(j)v^2_m(j) ,\\
&S_u(j)=D_u(j)/D(j) ,\\
&S_v(j)=D_v(j)/D(j) ,
\end{aligned} \tag{17}$$

with

$$\begin{aligned}
&T_m(j)=\tfrac{1}{2}\langle m(j)|R^2Y_{20}|m(j)\rangle f_{mm}(j)t_{mm}(j) ,\\
&D(j)=[G(j)S_3(j)-1][G(j)S_5(j)-1]\\
&\qquad -[G(j)S_4(j)]^2 ,\\
&D_u(j)=G^2(j)S_2(j)S_4(j)-G(j)S_1(j)[G(j)S_5(j)-1] ,\\
&D_v(j)=G^2(j)S_1(j)S_4(j)-G(j)S_2(j)[G(j)S_3(j)-1] ,
\end{aligned}$$

and

$$\begin{aligned}
&S_3(j)=\tfrac{1}{2}\sum_m u^4(j)t_{mm}(j) ,\\
&S_4(j)=\tfrac{1}{2}\sum_m u^2(j)v^2(j)t_{mm}(j) ,\\
&S_5(j)=\tfrac{1}{2}\sum_m v^4(j)t_{mm}(j) .
\end{aligned} \tag{18}$$

(To simplify these expressions we have omitted the explicit dependence in ω of the quantities F, S_i, T_m, D_i, and t_{mn}.) We note that the poles of $F(\omega)$ in (16) are no more only simple two quasiparticle energies.

To solve Eqs. (15) and (16) one has to fix the quadrupole χ_2 and octupole χ_3 strengths. In principle, the values of these *free* parameters are adjusted so as to get, *within the MPM,* the lowest 0^+ excited level at the energy $E(0^+_2)$ equal to the observed energy and the lowest 0^- excited state at an energy $E(0^-_1)$ in the neighborhood of the intrinsic band head energy $E_{\rm int}$. (This value $E_{\rm int}$ can be deduced from the observed energy of the first $I^\pi=1^-$ state by subtracting the rotational contribution roughly estimated to ≈ 10 keV.)

As is well known, the nonconservation of the number of particles introduces for $K^\pi=0^+$ a spurious state $\hat{N}|0\rangle$, where $\hat{N}=\sum_m C^\dagger_m C_m$ is the number operator. In the RPA, it corresponds to the solution $\omega=0$ of the secular equation. In the TDA, which we use here, the spurious contribution is distributed over several low lying states. We have to decide which solution of Eq. (16) to retain as the building block of our version of the MPM. In practical situations (i.e., at least for the nuclei studied in this paper), the lowest TDA solution has an overlap with the spurious state greater than 75%. Furthermore, it has the same symmetry of the wave function as the spurious RPA solution. Consequently, we prefer to use the second collective solution, which has a much smaller overlap with the spurious state (about 20%), as the building block of our approach.

In practical situations one has, of course, to truncate the space spanned by the states (4). Another numerical ingredient of the MPM2 is therefore the maximum value $n_{\rm max}$ of the number of phonons $p+q$ in the "basis" (4). This $n_{\rm max}$ is chosen so as to get the numerical stability of the energies of the states 0^-_1, 0^+_2, 0^+_3, 0^-_2, and 0^+_4. In the nuclei under consideration we used $n_{\rm max}=8$. With this choice the energies $E(0^-_1)$ and $E(0^+_2)$ were obtained within ≈ 3 keV and the energies $E(0^+_3)$, $E(0^-_2)$, and $E(0^+_4)$ within ≈ 10 keV.

Since the states (4) do not form an orthogonal basis, the eigenstates Ψ_j of H cannot be obtained by a simple diagonalization. To get them we use the method of Löwdin.[15] As a consequence, the eigenfunctions Ψ of the model Hamiltonian which we expanded in the states (4)

$$\Psi_j=\sum_{pq} D_{pq}(j)|pq\rangle \tag{19}$$

can generally not be interpreted in a simple way, e.g., their components in the "basis" (4) can even be larger than unity. Therefore, to get a better feeling concerning the nature of these eigenstates we calculate the $E1$, $E2$, and $E3$ transitions which link them. Since we restrict ourselves, as explained earlier, to 20 active single particle levels we need, of course, to introduce some effective charges in the $E\lambda$ transition operators. For the $E1$ we introduce $e_p=eN/A$ and $e_n=-eZ/A$, while for the $E2$ and $E3$ we use $e_p=e(1+\epsilon)$ and $e_n=e\epsilon$. In practical situations, calculations are done for the values $\epsilon=0$ and/or $\epsilon=0.10$. The units are chosen so that $|\langle 0^+_1|E\lambda|0^-_1\rangle|=1$ for $\lambda=1$ or $\lambda=3$ and $|\langle 0^+_2|E2|0^+_1\rangle|=1$ for $E2$ transitions. In the particular cases where some experimental results concerning $B(E2)$ and/or $B(E3)$ are known we have also used the usual single particle units (s.p.u.).

As is well known, the BCS treatment introduces some nonconservation of the particle number. To see how the MPM2 eigenstates Ψ are affected by this symmetry breaking, we have also calculated the deviations $\Delta N=|\langle\Psi|\hat{N}|\Psi\rangle-N|$ of the number of particles for the five lowest lying excited MPM2 states.

We may also remark here that there is, *a priori,* no reason to use the same values of the strength parameters χ in the model Hamiltonian and in the resolution of the TDA equation. However, to restrict as much as possible

the number of free parameters of the problem we use only one value of χ for each multipole.

III. RESULTS AND DISCUSSION

This section will be separated into two parts. In the first one we study the sensitivity of the MPM2 results versus the different ingredients of this approach in one particular nucleus. For this purpose we have chosen ^{234}U which is, from the experimental point of view, one of the best known even nuclei of this mass region and for which our theoretical results have been published earlier.[3] It corresponds to a situation where the $I^{\pi}=1^{-}$, $K=0$ and $I^{\pi}=0^{+}$, $K=0$ states are observed at energies differing by less than 30 keV. It is therefore, a case where the coupling between the two $K=0$ modes is expected, *a priori*, to be of a certain importance. The second part of this section is devoted to a systematic study of a series of nuclei of the same region: more precisely, we apply the MPM2 to ^{230}Th, $^{232-234-236}$U, and $^{238-240}$Pu. The aim of this analysis is twofold: first, we want to point out the general properties of the obtained MPM2 results within this set of nuclei. Second, we discuss these versus the experimental information, bearing in mind that our ambition is not, as discussed in Sec. I, to search for a fine agreement between theory and experiment.

A. Sensitivity of the results of the MPM2 versus the different ingredients of this approach in the case of ^{234}U

As recalled in Sec. II, the MPM2 has some "parameters" which we classify into two categories: (1) Those which we call "numerical parameters" and which are essentially related to the size of the MPM calculations. (Clearly this family includes the value $n_{\max}$ defined earlier and the number of active levels introduced in the BCS treatment.) (2) Those of dynamical nature which are, with our choice of the model Hamiltonian, the strengths of the residual pairing and multipole-multipole forces.

In Fig. 1, we recall the MPM2 results[3] obtained with these parameters fixed as indicated in Sec. II. The energy spectrum relative to the ground state clearly exhibits some anharmonicities: the 0_2^-, 0_3^+, and 0_4^+ levels appear at energies larger than twice the energy of the 0_1^- or 0_2^+ states. According to the nonorthogonality of the MPM states (4), the eigenfunctions are not easy to interpret. However, a simple glance at the $E1$ and $E2$ matrix elements demonstrate that favored electric transitions connect some definite levels: e.g., the $E1$ transitions between 0_4^+ and 0_1^- or 0_2^- and 0_2^+, the $E2$ transitions between 0_2^- and 0_1^- or 0_3^+ and 0_2^+. As a consequence, it seems natural, in this nucleus, to give some "one phonon" or "two phonon" labels to the different calculated levels. (Note the quotes used throughout the whole paper to recall that these states are not pure one or two phonons.) More precisely, 0_1^- will be called the "one 0^- phonon," 0_2^+ the "one 0^+ phonon," whereas 0_2^- will be named the "one 0^- and one 0^+ phonon," 0_3^+ the "two 0^+ phonon," and 0_4^+ the "two 0^- phonon" states. Furthermore, we will introduce some anharmonicity ratios defined as following:

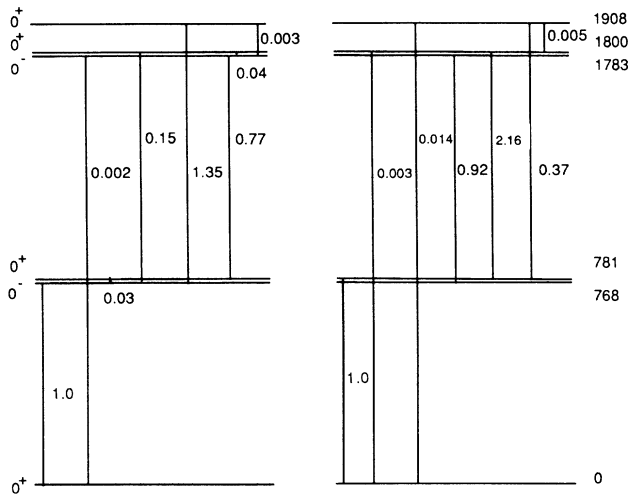

FIG. 1. Relative energy spectrum obtained within the MPM2 for ^{234}U and with the following parameters: $\epsilon_2=0.22$, $\Delta_n=660$ keV, $\Delta_p=970$ keV, $\chi_2=1$ keV, $\chi_3=9.1$ keV, and $n_{\max}=8$. The lhs and rhs give respectively the square of the intrinsic $E1$ and $E2$ matrix elements.

$$R_1=E(0_3^+)/E(0_2^+)\ ,$$
$$R_2=2E(0_2^-)/[E(0_2^+)+E(0_1^-)]\ , \tag{20}$$
$$R_3=E(0_4^+)/E(0_2^-)\ .$$

In the present nuclei, the identification of the "two phonon" states on the basis of favored electric transitions and the definitions of the R_j ratios are rather evident. We note that, in some cases, where the strength of the $E1$ transitions from the two 0_3^+ and 0_4^+ states to the 0_1^- state and the $E2$ transition from these levels to the 0_2^+ state are shared in a more equitable way, these identifications and definitions may not be so easy; they may even be meaningless.

Let us first study the sensitivity of these results vs $n_{\max}$. In Fig. 2 we give the evolution of the energies of the "one phonon" and "two phonon" states with $n_{\max}$, all other parameters being fixed. One observes on this figure that the "one" and "two phonon" states behave differently: the energies of the "one phonon" states oscillate with increasing $n_{\max}$ depending on the parity of this parameter, whereas the "two phonon" states have an energy decreasing continuously with $n_{\max}$. In the two cases, the variation of the energies decrease with increasing $n_{\max}$ leading to the required physical stability of the numerical results. These different evolutions are due to the importance of the H_{40} part of the model Hamiltonian, which connects multiphonon states $p+q$ with those having $p+q\pm2$. By the way, we note that the part H_{31}, which was completely absent in the MPM1 approach,[1] does not contribute in a significant way in the MPM2.

Now, we would like to discuss the sensitivity of the results versus the number of active levels introduced in the BCS treatment. We first remind the reader that this number must be chosen with some caution. It must be

sufficiently large to lead to stable numerical results and not too large to avoid the effects of states which would be unbound in a more realistic finite potential well (e.g., the states with a large principal quantum number for which some matrix elements of $R^{\lambda}Y_{\lambda 0}$ with bound states would become tremendously large). Earlier MPM1 calculations for $K^{\pi}=0^{-}$ states[1] or for $K^{\pi}=0^{+}$ vibrations[16] used for each kind of particles 30 active levels equitably distributed on each side of the Fermi surface. This choice seems to be a good compromise. Unfortunately, a similar choice would lead in the MPM2 version to a tremendous large number of B quantities to calculate. This would render the parameter adjustment very time consuming. We were therefore, forced to restrict ourselves to 20 active levels of each type of particles for which one needs already 30 min on a IBM 3090/200 computer for *one* given parameter set. In practice, to study the sensitivity of the results versus the number of active levels we made therefore, two types of calculation. In the first one we used the first version of the MPM build on $K^{\pi}=0^{-}$ phonons and compared, in Table I, the results obtained with 30 and 20 active levels. In the second one, we used the MPM2 and compared, in Table II, the results obtained with 20 and 16 active levels. In each case the strength parameters of the model Hamiltonian have been adjusted as indicated in Sec. II. It is noticeable that the energies of the "two phonon" states and the anharmonicity ratios remain stable. For this reason, we conclude that the restriction to 20 active levels is justified and appears, from the computational point of view, as a reasonable compromise.

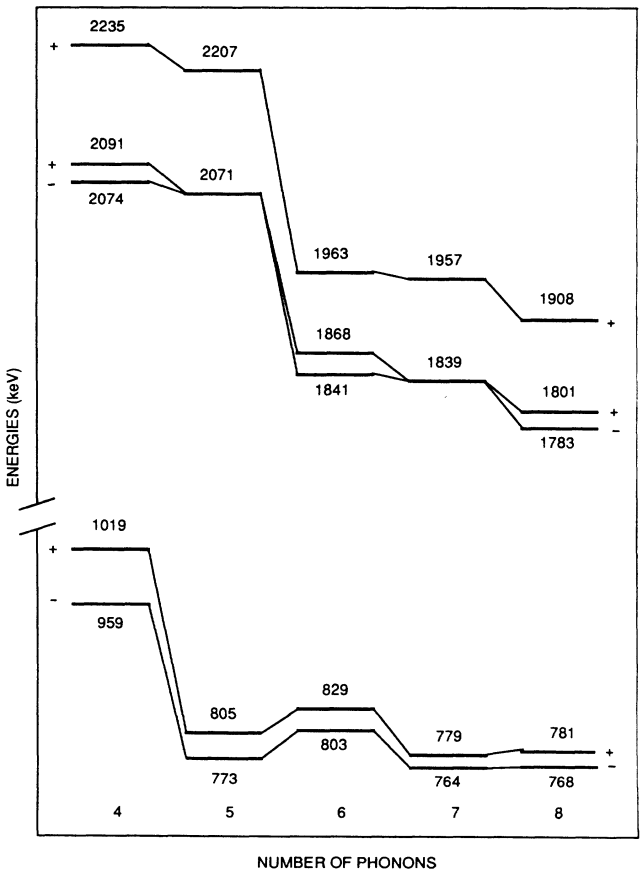

FIG. 2. Evolution of the energies, $E(0_1^-)$ and $E(0_2^+)$ of the "one phonon" states, $E(0_2^-)$, $E(0_3^+)$, and $E(0_4^+)$ of the "two phonon" states in ^{234}U vs the maximum value n_{max} of the number of phonons $p+q$ introduced in the MPM2 "basis" (4). The parameters are the same as in Fig. 1.

TABLE I. Comparison of the results obtained in ^{234}U for the MPM1 with 20 and 30 active levels. The 0_2^+ is here the "two 0^- phonon" state. All energies are given in keV.

	20 levels	30 levels
Number of X protons	18	43
Number of X neutrons	17	39
χ_3	8.84	2.65
$\omega_{TDA}(0_1^-)$	1172	1407
$E(0_1^-)$	775	775
$E(0_2^+)$	1881	1863
R_3	1.21	1.20

Now we look for the sensitivity of the MPM2 versus the variation of the dynamical parameters. In Sec. II we showed that the empirical values of the gap parameters Δ_p and Δ_n are extracted from experimental masses within ≈ 100 keV. It is therefore, interesting to see how the MPM2 results are affected by a variation of these gaps in their domain of determination. First, we note that a variation of the Δ's changes the position of the asymptotes of the TDA secular equations (15) and (16) and influences therefore, the nature of the TDA solutions used as the building blocks of the MPM2. According to the nature of these asymptotes the relevant parameter will be Δ_p or Δ_n. We made several calculations where Δ_p or Δ_n have been varied within ≈ 100 keV, and where the multipole-multipole strengths parameters are adjusted, for each choice of the Δ's, so as to reproduce the empirical "one phonon" energies. The variation observed for the energies, of the calculated "two phonon" states, were only of the order of 30–60 keV, attesting the stability of the MPM2 results and, by the way, their physical nature.

The influence of the variation of χ_3 on the MPM2 results, all other parameters being fixed, is shown in Fig. 3 and Table III. Several remarks can be done. With in-

TABLE II. Comparison of the results obtained in ^{234}U for the MPM2 with 16 and 20 active levels. All energies are given in keV.

	20 levels	16 levels
χ_3	9.1	11.3
$\omega_{TDA}(0_1^-)$	1143	1084
$\omega_{TDA}(0_1^+)$	1257	1213
$E(0_1^-)$	768	770
$E(0_2^+)$	781	712
R_1	2.30	2.37
R_2	2.30	2.33
R_3	2.48	2.51

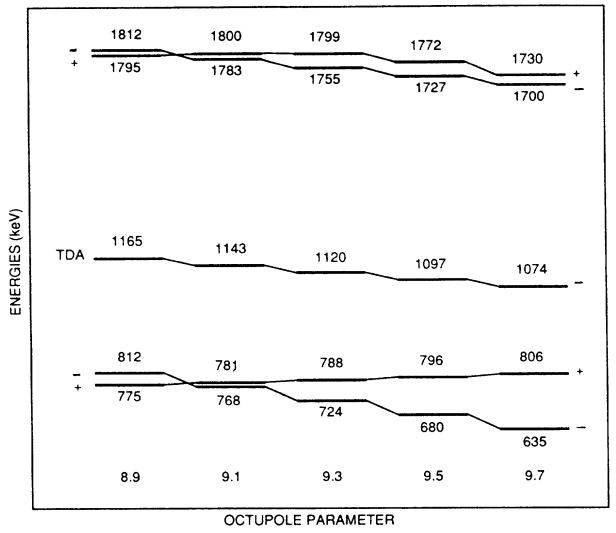

FIG. 3. Variation of the calculated MPM2 level energies in ^{234}U vs χ_3, all other parameters being those in Fig. 1.

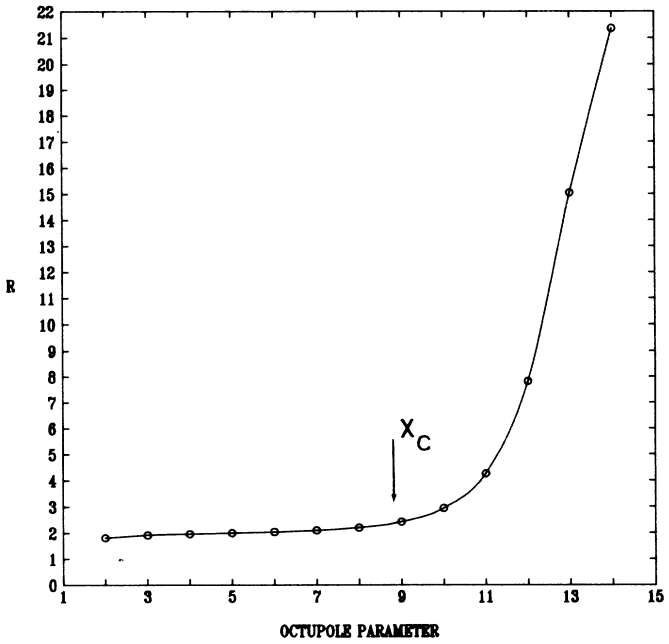

FIG. 4. Variation of the anharmonicity ratio $R_3 = E$ ("two 0^- phonon")/E ("one 0^- phonon") in ^{234}U for a MPM1 calculation versus χ_3. The arrow indicating χ_c corresponds to the critical value where the first physical solution of the RPA disappears.

creasing χ_3, the collective character of the $K^\pi = 0^-$ phonon increases, the energy $E(0_1^-)$ decreases while one observes a slight enhancement (≈ 10 keV) of $E(0_2^+)$, and the difference between $E(0_1^-)$ and the TDA energy ω_{TDA} increases. As a consequence, the difference between the harmonic TD approximation and the MPM2 results gets more important with increasing collectivity of the octupole building phonon. The evolution of the "two phonon" states is more involved. The 0_2^- keeps the character of the "one 0^+ and one 0^- phonon" and its energy follows the variation of $E(0_1^-)$. To understand the evolution of the "two phonon" states nature, it is worthwhile to consider the $E1$ transition from 0_3^+ and 0_4^+ to 0_1^- and the $E2$ transitions of the same 0^+ states to the 0_2^+. It is clear that around $\chi_3 = 9.3$ the 0_3^+ and 0_4^+ exchange their character: the 0_3^+ is mainly the "two 0^+ phonon" state for low χ_3 and the "two 0^- phonon" state for $\chi_3 > 9.3$ and *vice versa* for the 0_4^+ level. The definitions of R_1 and R_3 have to be interchanged when passing the pseudocrossing. Near this point their definition is somewhat meaningless. We note on one hand, that R_1 is quite stable, demonstrating that the variation of χ_3 does not influence in a noticeable way the behavior of the "two 0^+ phonon" state. On the other hand, R_3 increases regularly with χ_3. In Fig. 4 we give the variation of the ratio R_3 in a MPM1 calculation vs χ_3. It is seen that for small values of χ_3 the MPM1 leads practically to an harmonic situation. Around and beyond the critical value χ_c, where the first solution of the RPA disappears, R_3 gets rapidly greater than 2, and the energy spectrum appears as dilated compared to an harmonic situation.

TABLE III. Variation of the predominant electric transition probabilities linking the "two phonon" to the "one phonon" states and of the anharmonicity ratios R_j obtained in ^{234}U vs χ_3 in the MPM2. Note that for $\chi_3 > 9.3$ keV the definition of R_1 and R_3 are interchanged.

χ_3	8.9	9.1	9.3	9.5	9.7
$0_4^+ \to 0_1^+$	1.52	1.35	0.91	0.29	0.09
$0_3^+ \to 0_1^-$	0.06	0.15	0.52	1.05	1.16
$0_4^+ \to 0_2^+$	0.11	0.37	1.09	2.13	2.44
$0_3^+ \to 0_2^+$	2.37	2.16	1.44	0.41	0.10
$0_2^- \to 0_1^-$	0.93	0.92	0.92	0.91	0.91
$0_2^- \to 0_2^+$	0.78	0.77	0.75	0.74	0.72
R_1	2.28	2.30	2.32	2.32	2.32
R_2	2.32	2.30	2.28	2.34	2.36
R_3	2.42	2.48	2.58	2.60	2.72

Finally, we have studied the sensitivity of the MPM2 results to the variation of χ_2. It came out that this parameter does not play any important role. The results presented in Fig. 1 are qualitatively not changed if one varies even by 300% the value of χ_2. This finding means that the first excited 0^+ stated (noted here 0_2^+) is certainly not of a quadrupolar nature, but mainly of pairing nature. This conclusion is in total agreement with the earlier work of Chasman.[17]

B. Application of the MPM2 to several Th, U, and Pu isotopes

Apart from the ^{234}U used as a test nucleus in Sec. III A, we applied the MPM2 to five other even actinides, where some experimental information exists, and for which the energy separation of the observed 0_2^+ and 1_1^- states varies from 30 to ≈ 300 keV. For each of the studied nuclei (^{230}Th, $^{232-234-236}$U, and $^{238-240}$Pu), we give, in Table IV, the most interesting results obtained within the

TABLE IV. Results obtained in the MPM2 for the six studied nuclei. All energies are given in keV. The electric transitions are given in units where $|\langle$ ground state $|E\lambda|$ "one phonon state" $\rangle| = 1$. Experimental information on possible "two phonon" states is given for comparison where it exists. The energies marked with an asterisk correspond to states which can certainly not be interpreted as "two phonon states."

	^{230}Th	^{232}U	^{234}U	^{236}U	^{238}Pu	^{240}Pu
χ_3	8.95	9.16	9.04	9.24	7.68	7.35
$\omega_{TDA}(0^-)$	945	940	1143	1122	1055	1020
$E(0_1^-)$	510	558	768	695	605	599
$E(0_2^+)$	695	776	781	792	749	884
(E"0^-0^-")	1740	1630	1908	1775	1739	1662
(E"0^+0^+")	1606	1791	1800	1855	1708	1819
Experiment			1044*		1229*	(1411)
					1427	1526
$E(0_2^-)$	1381	1534	1783	1722	1592	1587
Experiment			1237*			
R_1	2.31	2.31	2.30	2.34	2.28	2.05
R_2	2.29	2.30	2.30	2.32	2.35	2.14
R_3	3.41	2.92	2.48	2.55	2.87	2.77
$E1$ $(0^-0^- \rightarrow 0_1^-)$	0.980	1.210	1.350	0.750	0.460	0.970
$E1$ $(0_2^- \rightarrow 0_2^+)$	0.890	0.730	0.770	0.730	0.900	0.930
$E2$ $(0_2^- \rightarrow 0_1^-)$	0.880	0.930	0.920	0.910	0.880	0.830
$E2$ $(0^+0^+ \rightarrow 0_2^+)$	2.37	2.44	2.16	1.71	1.43	2.05

MPM2. We give the energies of the fitted "one phonon" states 0_1^- and 0_2^+, the energies of the calculated "two phonon" states "0^-0^-," "0^+0^+," and 0_2^- and the anharmonicity ratios R_1, R_2, and R_3 defined in Eq. (20). Experimental information on the possible two phonon states is given for comparison when it exists. We also indicate the value of the free parameter χ_3 of the theory and the value ω_{TDA} of the solution of the TDA secular equation for $K^\pi = 0^-$. We restrict the information concerning the electric transitions to the two largest $E1$ and $E2$ linking the "two phonon" and the "one phonon" states, in the units indicated in Sec. II.

A few specific remarks can be formulated for some of the studied nuclei and some general properties of the MPM2 results become clear. The ^{240}Pu is of particular interest since it has often been considered as an example where there exists some evidence for the observation of the "two octupole phonon" states. Schmorak *et al.*[18] observed in the β^- decay of the isomeric state (7.22 min) of ^{240}Np two levels at 1411 and 1438 keV assigned to the $I=0$ and $I=2$ states of a rotational band with $K=0$. Assuming a positive parity for these levels, they found some evidence for an interpretation as "two octupole phonon" states: these levels decay mainly to levels of the one 0^- phonon band and, furthermore, they are not observed in one neutron transfer reactions (d,p) and (d,t). More recent experimental results[19,20] indicate the possibility of a negative parity for these two levels, excluding the given interpretation in favor of a two quasiproton $\frac{5}{2}^+642$ and $\frac{5}{2}^-523$ configuration. In an interpretation as a "two phonon" state the level observed at 1411 keV would have an anharmonicity ratio R_3 of 2.36 vs 2.77 in the MPM. We note that another 0^+ level has been located at 1526 keV. According to the corresponding value $R_3 = 2.56$, it may be a better candidate for a "two phonon" state. In ^{238}Pu, with the parameters fixed as indicated in Sec. II, we had some difficulties attempting to reproduce the energy of the first excited 0^+ state, which

TABLE V. Comparison of the calculated $B(E\lambda, 0 \rightarrow I_F = \lambda)$ where $\lambda=2$ and $\lambda=3$ with the experimental results taken from Ref. 21.

Nucleus	K^π	I^π	Energy	Experiment	Theory	Effective charge ϵ
^{238}Pu	0^+	2^+	983	3.8±0.5	1.0	0.0
					1.2	0.1
	0^-	3^-	661	30. ±5	17.4	0.0
^{236}U	0^-	3^-	744	23. ±3	9.4	0.0
^{234}U	0^+	2^+	852	2.3±0.3	1.2	0.0
					1.5	0.1
	0^-	3^-	849	26 ±3	8.9	0.0
^{230}Th	0^+	2^+	677	1.1±0.1	0.1	0.0
	0^-	3^-	572	29 ±3	12.4	0.0

TABLE VI. Comparison of the most interesting results obtained for the six studied nuclei in the two versions MPM1 and MPM2 of our method. The units are the same as in Table IV.

		^{230}Th	^{232}U	^{234}U	^{236}U	^{238}Pu	^{240}Pu
$E(\text{"}0^-\text{"})$		510	558	768	695	605	599
$E(\text{"}0^-0^-\text{"})$	1	1666	1620	1823	1775	1686	1649
	2	1740	1630	1908	1775	1739	1662
R_3	1	3.27	2.90	2.37	2.55	2.79	2.75
	2	3.41	2.92	2.48	2.55	2.87	2.77
$E1(2\rightarrow 1)$	1	1.020	1.336	1.690	1.456	0.800	0.996
	2	0.980	1.210	1.350	0.750	0.460	0.970

is obtained in the MPM2 at 749 keV, i.e., somewhat lower than observed (942 keV). Nevertheless, we may conclude that the 0^+ state observed at 1299 can, by no means, be interpreted as a "two phonon" state, its "experimental" anharmonicity ratio being much smaller than that predicted in the MPM. This kind of conclusion is similar to that drawn earlier[3] for the 0^+ states at 1044 keV and for the head of the $K^\pi=0^-$ band at 1237 keV observed in ^{234}U (see also Table IV).

In Table V we compare the reduced transition probabilities $B(E\lambda,0\rightarrow I_F=\lambda)$, in s.p.u., for $\lambda=2$ and $\lambda=3$ obtained in the MPM2, using $\epsilon=0$ in the effective charges, with the experimental results.[21] More precisely, we give the values of $B(E2)$ for the transition from the ground state to the rotational $I=2$ state of the first $K^\pi=0^+$ band (i.e., 0_2^+ in the MPM2 calculation) and the values of $B(E3)$ from the ground state to the rotational $I=3$ state of the $K^\pi=0^-$ band (noted 0_1^- in the MPM2). In this table we see that the order of magnitude of the calculated $B(E\lambda)$ are the same as those observed. In some cases we also give the values obtained in the MPM2 using $\epsilon=0.10$ in the effective charges, just to show that the corresponding variation goes in the right way. As mentioned in Sec. I, our aim is not to search for a fine agreement between theory and experience, but only to look for the general properties of the MPM2. The results of Table V clearly illustrate that the description of the observed electric transitions is correct.

Finally, in Table VI we compare the results concerning the "one 0^- phonon" and "two 0^- phonon" states obtained in the two versions MPM1 and MPM2 of our approach. The comparison of these data shows that, even in the case where the levels 0_3^+ and 0_4^+ have quite similar calculated energies (e.g., in ^{238}Pu) the anharmonicity ratios R_3 are not affected in a significant way when one switches from MPM1 to MPM2. The effect of the coupling between the two $K=0$ modes, of different parity, is better seen on the variation of the $E1(2\rightarrow 1)$ transition from the "two 0^- phonon" state to the "one 0^- phonon" state when one uses two building phonons instead of one. To see some noticeable effect of this coupling, it appears that the separation of the two observed 0_2^+ and 1_1^- levels and the collectivity of the two types of phonons must combine their effects so as the 0_3^+ and 0_4^+ states appear at neighboring energies.

To evaluate, in some way, the effects of the nonconservation of the number of particles induced by the BCS treatment, we have also calculated, in all the studied nuclei, the deviation ΔN of the number of particles for the five lowest excited MPM2 states. As for the ^{234}U results published earlier,[3] it was found that this deviation was generally ≈ 1 and always <2.

The analysis of the data presented in Tables IV and VI allows us to state some general properties of the MPM2 results, which were also indicated earlier[3] for ^{234}U. First, we note that the quadrupole force parameter χ_2 does not play any important role for the description of the "one phonon" state. For the "two phonon" states, we observe three cases. In ^{232}U and ^{240}Pu, the 0_3^+ is mainly the "two 0^- phonon" state and the 0_4^+ the "two 0^+ phonon" state. In ^{230}Th and ^{234}U it is the other way round. For ^{236}U and ^{238}Pu these identifications are a little less evident. But, in any case, it is observed that the "two 0^- phonon" state has an anharmonicity ratio greater than the "two 0^+ phonon" state, according to the fact that the octupole phonon is much more collective.

IV. COMPARISON BETWEEN THE MPM AND THE QPNM OF SOLOVIEV *et al.*

The two methods have in common that they aim to find an explanation of the lack of the "two phonon" states in the energy region of twice the energy of the one phonon state. On one hand, as we have seen in Sec. III, the MPM finds that the "two phonon" states appear with anharmonicity ratios ≥ 2.3 and that they keep some collective character in their electric transitions to the "one phonon" states. On the other hand, in the QPNM of Soloviev *et al.*,[5] it was concluded that the "two phonon" states are pushed to energies of the order of ≈ 3–4 MeV, where they lose completely their collectivity since they are fragmented over many levels. The aim of the present section is to try to explain this difference in the conclusions of the two models.

We therefore, remind the reader of the different steps of the QPNM. This model starts with phonons of the RPA type

$$Q_j^\dagger=\sum_{rs}\left[(X_j)_{rs}a_r^\dagger a_s^\dagger-(Y_j)_{rs}a_s a_r\right], \tag{21}$$

where the index j summarizes the quantum numbers which identify the phonon. As in (1) the $a^\dagger$ and a operators stand for quasiparticles. But, the corresponding single particle states are now eigenfunctions of the Woods-

Saxon potential, the parameters of which are tailor studied for each definite mass region. The model Hamiltonian of the QPNM includes, in addition to the constant pairing force, numerous terms of the multipole-multipole expansion of the long range force. Even in some cases, a rather sophisticated spin-multipole force is also introduced. The matrices X_j and Y_j in (21) are obtained by solving, for each multipole $\lambda\mu$, the secular equation of the RPA. The strengths $\chi_{\lambda\mu}$ are fixed, for each multipole, so as to fit, *within the RPA,* the lowest solution of the RPA to the observed level energy. The trial wave function of an excited nuclear state in an even nucleus is written in the form

$$|n\rangle = \left[\sum_k R_{nk}(\lambda\mu) Q_i^\dagger + \sum_{jl} P_{nlj}(\lambda\mu) Q_j^\dagger Q_i^\dagger \right] \left| 0 \right\rangle , \quad (22)$$

where the index k stands for the number of the solution of the RPA equation for a given multipole $\lambda\mu$. The index i summarizes the triplet $k\lambda\mu$. The sum over $k = k'\lambda'\mu'$ and $l = k''\lambda''\mu''$ is restricted to values of $\lambda'\mu'$ and $\lambda''\mu''$ compatible with $\lambda\mu$. In practical situations, Soloviev *et al.*[5] introduced the first ten solutions of the RPA equation for each of the following multipoles $\lambda\mu = 20$, 22, 30, 31, 32, 33, 41, 43, and 44. (In some specific cases even more higher multipoles are introduced.) This choice allows them to take into account the coupling of the collective phonons to some of the noncollective ones. The energies E_n and the coefficients R and P of the trial wave functions are then obtained by a variational procedure.

As in the MPM, the Pauli principle is taken into account in the QPNM. In the evaluation of $\langle n \mid n \rangle$ one has to calculate terms like $\langle 0 Q_{l'} Q_{j'} Q_j^\dagger Q_l^\dagger 0 \rangle$ which can be written in the form

$$\langle 0 Q_{l'} Q_{j'} Q_j^\dagger Q_l^\dagger 0 \rangle = \delta_{jj'}\delta_{ll'} + \delta_{jl'}\delta_{j'l} + \mathcal{K}(j',l';j,l) . \quad (23)$$

As this stage, the authors of the QPNM make the approximation which consists to retain in (23) only the diagonal $j = j'$ and $l = l'$ contribution in $\mathcal{K}$. It has been shown[22] that this approximation can reasonably be made. Within this framework, the "two phonon" states are pushed to energies of the order of 3–4 MeV. In this energy region the density of levels is, of course, very high and the experimental identification of these specific states is quite impossible since, according to the predictions of the QPNM, they lose completely their collectivity. Therefore, the authors of the QPNM conclude that "two phonon" states do not exist in deformed nuclei.

In Sec. III, we have shown that the stability of the "two phonon" states is very sensitive to the maximum number $n_{\max}$ of the phonons $p+q$ introduced in the "basis" (4). In particular, it was found that for $n_{\max} = 3$ and a fixed value of χ_3, one observes an important over estimation of the energy of the "two phonon" states. As a consequence, it seems not surprising to us that, in the QPNM, these states are pushed to high energies. Indeed, the trial wave function (22) used by Soloviev *et al.* only takes into account one and two phonon contributions and neglects the effect of the components with 3, 4, and even more phonons which, for *collective* phonons, have been shown in Sec. III to play an important role.

TABLE VII. Comparison of the energies (in MeV) of the "two phonon" states obtained in the QPNM of Ref. 5 with those calculated in the MPM2 where $n_{\max} = 3$.

Nucleus	K^π	$\lambda_1\mu_1 k_1$	$\lambda_2\mu_2 k_2$	QPNM	MPM2
^{240}Pu	0^+	2 0 1	2 0 1	4.2	2.5
		3 0 1	3 0 1	3.3	2.8
	0^-	2 0 1	3 0 1	4.2	2.4
^{232}U	0^+	2 0 1	2 0 1	6.0	2.8
		3 0 1	3 0 1	3.7	2.5

In Table VII we compare the results of the QPNM and the MPM2 for the energies of the excited states 0^+ and 0^- for which the wave functions have their main component on two collective phonons. The values given in the column labeled QPNM are taken from Ref. 5, those indicated in the column labeled MPM2 are obtained with $n_{\max} = 3$. At first sight, this choice of $n_{\max}$ might appear surprising. However, if one wants to compare with the QPNM, where the basic phonons are of the RPA type and contain therefore, some effect of the part H_{40} of the Hamiltonian, one also needs to introduce the effect of this term in the MPM2 and it is only for $n_{\max} \geq 3$ that this term influences the one phonon collective state. It is worthwhile to note that with this value of $n_{\max}$, the fitted strength parameters χ_2 and χ_3 are only slightly different from those using $n_{\max} = 8$. As a consequence, it seems difficult to admit that the effect of the components with 3, 4, . . . , 8 phonons in a generalized form of (22) might be simulated by a simple renormalization of the strength parameters χ, as already assumed in Ref. 5.

Even if the energies obtained with the MPM2 and $n_{\max} = 3$ are somewhat smaller than those given by the QPNM, it is clear, in conclusion, that a collective "basis" with a small number of phonons over estimates the energies of the "two phonon" states compared to a "basis" where $n_{\max}$ is fixed on a criterium of stability, as in Sec. III.

V. CONCLUSIONS

We would like to summarize the different important points we learned concerning the vibrational states with $K = 0$ in the mass region $224 \leq A \leq 240$.

First, we confirm, as already expected from Chasman's work,[17] that in this region the "one phonon" $K = 0$ states can practically be described by taking into account only the pairing and the octupole degrees of freedom. This leaves, in fact, only one free parameter χ_3 in the problem.

Second, as for the quasiparticle phonon nuclear model of Soloviev *et al.*, the full treatment of the Pauli principle leads to dilated energy spectra. However, at variance with the mentioned model, we found that the "two phonon" states are predicted in an energy region of 1.5–2.0 MeV. Further, they keep some collective character in their electric transitions to the "one phonon" states. This property should help to find some experimental evidence for their existence. A systematic experimental search of

these states remains to be undertaken. We may add here that similar conclusions have recently been obtained[23] for the "two γ phonon" states in the rare earth region.

Third, we found that the $K^{\pi}=0^{+}$ mode, which is mainly of the pairing type in this region, is only weakly coupled to the octupole mode. This gives, *a posteriori*, a justification of our previous use[1] of the MPM1 version of our approach. It would be of interest to see if this coupling also remains small in the case where the $K^{\pi}=0^{+}$ is expected to be of collective quadrupole nature (e.g., in ^{152}Sm and its neighbors). The same question concerning the coupling with the γ phonon can also be asked. Unfortunately, in this last situation the MPM "basis" must be extended so as to include 4 phonons: the present two $K=0$ phonons, the γ phonon with $K^{\pi}=2^{+}$ and its time reversed, carrying $K^{\pi}=-2^{+}$. This is, at least for the moment, out of the possibilities of the available computers. Furthermore, we feel that this improvements of the MPM should take place after some others dealing e.g., with a proper treatment of the nonconservation of the number of particles and with the introduction of the coupling with noncollective (two quasiparticle) states.

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