Fragmentation of the Two-Octupole Phonon Multiplet in ^{208}Pb

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The properties of the lowest two-phonon states in ^{208}Pb built on the collective octupole vibration are studied within the quasiparticle phonon model. Calculations including up to three-phonon coupling yield a strong fragmentation of the 6^+ member of the multiplet, explaining the failure of recent heavy ion Coulomb excitation experiments used to observe this state. Some splitting of the 2^+ strength is predicted while the 0^+ and 4^+ members remain relatively pure. The present results also question the proposed signature for the two-octupole states of enhanced $E1$ decays to one-phonon states. [S0031-9007(98)08236-2]

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The concept of phonons is a keystone for the description of collective excitations in many-body systems. The best way to examine its validity in real physical systems is to study deviations from the harmonic picture for multiphonon excitations. In nuclear physics the most collective modes are surface vibrations with spin and parity, 2^+ and 3^-, and giant resonances. Studies of the properties of the low-lying two-phonon multiplet $[2_1^+ \times 2_1^+]_{0^+,2^+,4^+}$ in many spherical nuclei as well as the first results on double giant resonances prove that anharmonicity effects are typically small in nuclei.

Uniquely among stable nuclei, in ^{208}Pb the octupole vibration is lowest in energy. Its collective nature is well established by an enhancement of 34 of the $B(E3)$ transition strength to the ground state (g.s.) with respect to single-particle units [1]. It thus represents an ideal candidate to search for another set of low-lying two-phonon states, viz., two-octupole phonon (TOP) vibrations which form a multiplet $[3_1^- \times 3_1^-]_{0^+,2^+,4^+,6^+}$ of positive parity states.

Despite considerable efforts using a variety of reactions and experimental techniques [2–7] information on TOP states is at best dubious with the exception of the 0^+ level. Using inelastic neutron scattering, the excitation of a $J^\pi = 0^+$ state at $E_x = 5.241$ MeV was observed followed by a characteristic $E3$-$E3$ γ-decay sequence to the g.s. which proves its dominant two-phonon character [8]. A particularly disturbing result is the failure of several recent experiments aiming at a population of the 6^+ multiplet member by two-step heavy ion Coulomb excitation [9–11]. Despite experimental sensitivities, about an order of magnitude below the expected cross sections (which is well understood for the Coulomb excitation process), no clear signal could be observed.

It is the goal of this paper to provide an explanation for some of these surprising results with a calculation that includes all relevant degrees of freedom in the low-energy region. This study is carried out within the quasiparticle-phonon model (QPM) described in detail in [12]. The properties of the $[3_1^- \times 3_1^-]$ multiplet in ^{208}Pb have been studied in a variety of theoretical approaches [13–20], but have been mainly focused on the anharmonicity shift relative to the harmonic value of twice the excitation energy of the 3_1^- state (2.615 MeV). Here, the main subject is purity of the TOP states. Is it really preserved or is the TOP strength more strongly mixed with other excitations than usually thought? Furthermore, the $^{208}\mathrm{Pb}(n, n'\gamma)$ experiments described above also suggest possible candidates for the 2^+ and 4^+ multiplet members [21]. Their identification is based on the observation of enhanced $E1$ decays from the observed levels to the 3_1^- state. The validity of this argument is critically examined in this paper.

For numerical calculations, single-particle wave functions and energies are determined from a Woods-Saxon potential with the parameters of [22]. For levels near the Fermi surface, experimental values of single-particle energies are taken from the neighboring odd nuclei [23,24]. These may be found in Ref. [25]. The energy gap between particles and holes is also taken from experiment [26]. The strength of the residual interaction with isoscalar, $\kappa_{\mathrm{is}}(\lambda^\pi)$, and isovector, $\kappa_{\mathrm{iv}}(\lambda^\pi)$, parts has been adjusted in the following way. The ratio $\kappa_{\mathrm{iv}}(\lambda^\pi)/\kappa_{\mathrm{is}}(\lambda^\pi) = -1.2$ was fixed from previous QPM studies of spherical nuclei [27]. The values of $\kappa_{\mathrm{is}}(2^+)$ and $\kappa_{\mathrm{is}}(3^-)$ were adjusted to reproduce the experimental values [1] of $B(E2, \text{g.s.} \rightarrow 2_1^+)$ and $B(E3, \text{g.s.} \rightarrow 3^-)$, respectively. The resulting $\kappa_{\mathrm{is}}(2^+)$ and $\kappa_{\mathrm{is}}(3^-)$ agree within 10%. Furthermore, we set $\kappa_{\mathrm{is}}(\lambda^+) = \kappa_{\mathrm{is}}(2^+)$ and $\kappa_{\mathrm{is}}(\lambda^-) = \kappa_{\mathrm{is}}(3^-)$ since the radial form factor of the residual forces (a derivative of the average field) is λ independent.

The calculations were performed with wave functions of excited states, including one-, two-, and three-phonon configurations. The physical properties of phonons, their excitation energy, and internal fermion structure, were obtained by solving the random-phase approximation (RPA) equations. Please note that within the QPM the term phonon refers to any solution of the RPA and is not restricted to the surface vibrations only. Thus, the latter are described as "collective" phonons whenever necessary to avoid

confusion. Below, we use the following notations for one-phonon configurations. $\lambda^\pi n$ represents the nth RPA solution for the multipolarity λ^π. For example, 2^+1 stands for the lowest 2^+ phonon, etc. Then, two-phonon configurations are denoted as $[\lambda_1^{\pi_1} n_1 \times \lambda_2^{\pi_2} n_2]_{\lambda^\pi}$, where λ^π gives the total spin and parity to which the phonons $\lambda_1^{\pi_1} n_1$ and $\lambda_2^{\pi_2} n_2$ are coupled. Two- and three-phonon terms of the wave functions were constructed from natural-parity phonons with $\lambda^\pi = 0^+ - 6^+$.

All one-phonon configurations with excitation energies below 7 MeV were included in the wave functions of excited states. The upper limits for two- and three-phonon configurations were 9 and 12 MeV, respectively. We have neglected two- and three-phonon states made of noncollective phonons only. These couple very weakly to other configurations and their omission does not affect the final result for the states under consideration. The diagonalization of the model Hamiltonian with typical model spaces of about 500 for a given λ^π yields a set of excited states λ_ν^π, where the index ν denotes the ordering by increasing excitation energy.

In order to test the present approach, we first consider the properties of the lowest excited states for which experimental data are available. The agreement with experiment for excitation energies and g.s. transition strengths presented in Table I is very good. The calculations reproduce the energies within a few tens of keV and the $B(E\lambda)$ values within 10%–15%. The only exception is the $B(E6)$ result which is underestimated by roughly a factor of 2. However, it has already been demonstrated elsewhere [27] that the collectivity of the lowest phonons of high multipolarity is underestimated in calculations if the single-particle continuum is approximated by narrow quasibound states as in the present study.

In the last column of Table I the contribution of the lowest phonon $\lambda^\pi 1$ to the wave function of the λ_1^π state is shown. This provides a measure for the one-phonon purity of the lowest vibrations in ^{208}Pb. As expected from general principles, it is rather high for this doubly magic nucleus. There are two main reasons for this. Firstly, the energy gap between the lowest one- and two-phonon configurations is sufficiently large. Secondly, the matrix elements of the interaction between these configurations are small for closed-shell nuclei. The first argument is less strict for positive parity states in this specific nucleus since the energy difference between one-phonon, λ^+1, and two-phonon, $[3^-1 \times 3^-1]_{\lambda^+}$ and $[3^-1 \times 5^-1]_{\lambda^+}$, configurations is approximately 1 MeV only. For negative parity states, the lowest two-phonon configuration, $[2^+1 \times 3^-1]_{\lambda^-}$, is located much closer (and, as a result, interacts stronger) to the lowest three-phonon configuration, $[3^-1 \times 3^-1]_{l^+} \times 3^-1]_{\lambda^-}$. For this reason the lowest 3^- and 5^- states are practically pure one-phonon states. The 6_1^+ state has large admixtures due to the stronger matrix elements coupling between n- and $(n+1)$-phonon configurations.

TABLE I. Calculated excitation energies and transition probabilities for the lowest 2^+, 3^-, 4^+, 5^-, and 6^+ states in ^{208}Pb in comparison with experimental data [1].

	E_x (MeV)		$B(E\lambda, 0^+ \to \lambda_1^\pi)$ (e^2 b$^\lambda$)		
λ_ν^π	expt.	calc.	expt.	calc.	One-phonon contribution
3_1^-	2.62	2.60	0.61	0.60	98.7%
5_1^-	3.20	3.15	0.045	0.041	99.2%
2_1^+	4.09	4.11	0.32	0.27	91.5%
4_1^+	4.32	4.26	0.16	0.15	95.7%
6_1^+	4.42	4.34	0.067	0.035	79.8%

In the next step we investigate the lowest two-phonon states $[3^-1 \times 3^-1]_{0^+,2^+,4^+,6^+}$, allowing for their interaction with other two-phonon configurations as well as with one- and three-phonon states. Treating phonons as ideal bosons, the multiplet would be degenerate in energy, equal to twice the energy of the first 3^- phonon. However, when their internal fermion structure is taken into account the self-energy of two-phonon configurations is shifted due to "Pauli principle corrections" which are obtained by applying exact commutation relations between phonon operators. This energy shift upwards for the pure $[3^-1 \times 3^-1]_{\lambda^+}$ configuration in ^{208}Pb equals, in our calculation, 0.33, 0.19, 0.16, and 0.49 MeV for $\lambda = 0$, 2, 4, and 6, respectively.

The properties of the lowest 2^+ and 3^- states and some components of the two-phonon multiplets built on 2^+1 and 3^-1 phonons in ^{208}Pb have been calculated recently with Skyrme SGII forces in a basis including one particle-one hole and selected two particle-two hole states [28]. The Skyrme interaction provides approximately similar one-phonon purity of the 2_1^+ and 3_1^- states and about 2 times larger energy shifts for the pure one-phonon 2^+1 or 3^-1, as well as for the $\lambda = 0, 2$ two-phonon configurations.

The final results for the TOP multiplet, including energy shifts and fragmentation induced by the interaction with the full model space of one-, two-, and three-phonon configurations, are given in Table II. Only states which carry at least 1% of the $[3^-1 \times 3^-1]$ configuration (see column three) are listed. In the fourth column we present the leading component of the wave function with a different structure. In the last column we give the $B(E\lambda)$ values of the g.s. transition strength. Since the direct excitation of the two-phonon configurations from the ground state is typically orders of magnitude smaller as compared to the excitation of one-phonon configurations, the $B(E\lambda)$ values mainly reflect the presence of the latter in the wave function.

Our calculations indicate that the 0^+ and 4^+ members of the $[3^-1 \times 3^-1]$ quartet remain practically pure. For the 0^+ state this is easily understood because the lowest three-phonon configuration is more than 4 MeV higher and the first noncollective (i.e., particle-hole) one-phonon 0^+ configuration is located at very high energy. The situation is

TABLE II. Fragmentation of the two-phonon configurations $[3^-1 \times 3^-1]_{0^+,2^+,4^+,6^+}$ over low-lying states in ^{208}Pb. The third column gives the TOP contribution to the wave functions. The fourth column is the largest component of other one- or two-phonon configurations. The last column is the $B(E\lambda,\ 0^+ \rightarrow \lambda^\pi_\nu)$ value for this state. Only states with a TOP contribution larger than 1% are shown.

λ^π_ν	E_x (MeV)	TOP	Other	$B(E\lambda)$ (e^2 b$^\lambda$)
0^+_1	5.43	95.5%	<1%	...
2^+_1	4.11	6.3%	2^+1 (91.5%)	0.27
2^+_2	5.29	76.4%	2^+2 (9.3%)	0.55×10^{-2}
2^+_3	5.55	7.9%	2^+2 (89.0%)	0.47×10^{-2}
2^+_6	6.20	3.2%	2^+4 (66.0%)	0.61×10^{-1}
2^+_7	6.32	1.0%	$[5^-1 \times 5^-1]_{2^+}$ (70.4%)	0.26×10^{-1}
4^+_2	5.27	94.5%	<1%	0.17×10^{-2}
6^+_1	4.34	14.2%	6^+1 (79.8%)	0.35×10^{-1}
6^+_2	4.97	14.8%	6^+2 (64.6%)	0.13×10^{-3}
6^+_4	5.47	8.2%	6^+4 (58.1%)	0.39×10^{-2}
6^+_5	5.72	1.0%	$[3^-1 \times 5^-1]_{6^+}$ (94.3%)	0.12×10^{-3}
6^+_6	5.74	4.0%	6^+5 (74.6%)	0.35×10^{-2}
6^+_7	5.81	2.3%	6^+6 (85.4%)	0.28×10^{-3}
6^+_8	5.99	33.1%	6^+7 (14.9%)	0.48×10^{-2}
6^+_9	6.10	6.6%	6^+7 (79.5%)	0.63×10^{-2}
6^+_{10}	6.28	1.7%	$[5^-1 \times 5^-1]_{6^+}$ (90.3%)	0.55×10^{-4}
6^+_{11}	6.41	5.1%	6^+8 (70.5%)	0.10×10^{-2}
6^+_{12}	6.45	1.4%	$[3^-1 \times 5^-2]_{6^+}$ (82.0%)	0.54×10^{-4}
6^+_{13}	6.61	1.3%	6^+9 (95.5%)	0.55×10^{-5}

different for the 4^+ states, where the second and third one-phonon configurations have very close excitation energies of 5.49 and 5.73 MeV, respectively. Nevertheless, the interaction matrix elements between these one-phonon and two-phonon $[3^-1 \times 3^-1]_{4^+}$ configurations are very small and they hardly mix with each other. A similar situation is encountered for $\lambda^\pi = 2^+$ with the presence of the second and third 2^+ phonons in the vicinity of the TOP quartet, but the coupling matrix elements are larger. As a result, the TOP configuration is distributed over the three lowest 2^+ states. The main fraction of about 75% is still concentrated in a state at 5.29 MeV.

The picture is very different for the 6^+ states. Firstly, the Pauli corrections move the TOP state to higher excitation energies, where the density of other configurations is increased. Secondly, and even more important, the matrix elements of the interaction between one-, two-, and three-phonon configurations are larger than for $\lambda^\pi = 2^+$. As a result, the 6^+ component of the two-octupole multiplet mixes with practically all one-phonon and also with a few two-phonon configurations. Its strength is fragmented over an energy region of about 2 MeV, as can be seen from the second column of Table II. This finding suggests a theoretical explanation of the failure to observe the $[3^-_1 \times 3^-_1]_{6^+}$ state in heavy ion Coulomb excitation experiments [9–11]. One should note that the cross sections for a population of the level at 5.99 MeV with the largest two-phonon component in Table II are reduced by a factor of 3 with respect to the harmonic case (amplitude 33.1%). The high energy (750 keV above twice the energy of the 3^-_1 state) leads to an extra reduction factor 10 with respect to the harmonic picture due to the steep energy dependence of the Coulomb excitation probability.

The calculation gives an energy of the TOP 0^+ state about 200 keV higher than observed in the experiment [8], but the agreement is still reasonable. The model prediction of the concentration of this multiplet member in a single state seems also to be in accord with experiment. However, it should be noted that some additional fragmentation of the 0^+ strength might occur due to mixing with proton and neutron pairing vibrations [13,19,29] which are outside the present model. The main problem with an identification of the 2^+ and 4^+ components of the TOP multiplet is related to the fact that the cleanest signature, i.e., a collective $E3$ decay to the 3^-_1 state, is masked by non-collective $E1$ transitions to the same state [21,30]. Two possible TOP candidates at 5.216 and 5.286 MeV have been reported recently [21]. The spin of the first state was identified as 4^+ and the second one as $(2^+, 4^+)$. Their identification has been based on the large $B(E1)$ values of the decay to the collective 3^-_1 state. These two experimentally observed levels correspond very well to the 4^+_2 state at 5.27 MeV and the 2^+_2 state at 5.29 MeV, respectively, in Table II which carry the largest fractions of the $[3^-1 \times 3^-1]$ configuration. However, the proposed signature for TOP states of enhanced $E1$ transitions to the collective one-phonon states requires some additional studies.

Because of the absence of collective $E1$ transitions in the low-energy spectra of nuclei many different so-called boson forbidden transitions play an essential role [31]. One example is the decays of TOP multiplet members into the low-lying 3^- and 5^- vibrations which is allowed only because of the internal fermion structure of phonons. The corresponding $E1$ transition strengths are given in the first four rows of Table III. These values are quite large compared to the average $E1$ strengths expected between low-lying levels in this mass region, which has led to the suggestion of using them as a signature of TOP states. However, there are also other cases, such as $E1$ transitions between one-phonon states. Some examples are given in the lower part of Table III for the energy region, where the states with dominant TOP structure are expected. Obviously, $E1$ transition strengths comparable to those observed for the two-phonon decay are quite common and of the same order of magnitude as observed experimentally [21]. This puts the proposed identification of the 2^+ and 4^+ TOP states into question.

So far, the calculations presented in Table III have been performed for pure one- and two-phonon configurations

TABLE III. Examples of $E1$ decay probabilities in ^{208}Pb of low-lying positive parity one- and two-phonon configurations to the collective one-phonon 3^- and 5^- states in the energy range where the TOP strength is expected.

J_i	E_i (MeV)	J_f	E_f (MeV)	$B(E1)$ $10^{-3}\ e^2\,\mathrm{fm}^2$
$[3^-1 \times 3^-1]_{2^+}$	5.32	3^-1	2.67	0.16
$[3^-1 \times 3^-1]_{4^+}$	5.32	3^-1	2.67	0.31
$[3^-1 \times 3^-1]_{4^+}$	5.32	5^-1	3.19	0.96
$[3^-1 \times 3^-1]_{6^+}$	5.32	5^-1	3.19	2.75
2^+1	4.28	3^-1	2.67	4.54
2^+2	5.57	3^-1	2.67	0.28
2^+3	5.82	3^-1	2.67	0.38
2^+4	6.30	3^-1	2.67	6.84
4^+1	4.45	3^-1	2.67	5.52
4^+3	5.73	3^-1	2.67	0.89
4^+4	6.02	3^-1	2.67	0.40
4^+1	4.45	5^-1	3.19	2.81
4^+2	5.49	5^-1	3.19	1.35
4^+3	5.73	5^-1	3.19	0.47
4^+4	6.02	5^-1	3.19	1.17
6^+2	5.24	5^-1	3.19	0.15
6^+3	5.48	5^-1	3.19	0.12
6^+6	5.92	5^-1	3.19	0.67
6^+7	6.17	5^-1	3.19	0.44
6^+8	6.41	5^-1	3.19	0.33

only. Interference effects as well as $[\lambda^\pi n \times \mathrm{GDR}]$ components in the wave functions could significantly alter the results. Even weak admixtures of the latter type, which leave the results in Tables I and II completely unaffected, can change the $E1$ transition strengths significantly. The importance of the giant dipole resonance (GDR) admixture in the low-energy electric dipole response has recently been demonstrated [31,32]. Calculations of the $E1$ decay properties using realistic wave functions for the low-energy states are clearly needed. Work along these lines is in progress.

In conclusion, QPM calculations with wave function, including all relevant one-phonon and multiphonon configurations in the low-energy region, indicate that the 6^+ member of the two-phonon octupole multiplet in ^{208}Pb is strongly fragmented. This explains the failure of recent experiments designed to preferentially populate this state. At the same time, the 0^+ and 4^+ partners are mainly concentrated in a single excited state and the strength of the 2^+ component is shared between three states. We also point out that the proposed identification of TOP states by strong $E1$ transitions to the collective 3^-_1 and 5^-_1 levels is highly questionable. Based on these findings, there is no easy route in the longstanding experimental search for the TOP multiplet members beyond the 0^+ state. Rather, one will need a complete spectroscopy of ^{208}Pb in the energy region of interest which represents a considerable task even with state-of-the-art techniques.

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Note added.—An excellent experimental confirmation of our predictions concerning the 6^+ TOP member has been published [33] after submission of the original manuscript. It demonstrates that about 20% of its strength is concentrated in the 6^+_1 state. No other traces of it are found at higher energies with a detection limit of 15% at 5.2 MeV and 100% at 6.0 MeV.

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