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Theory of the Anharmonicity in the Vibrational Motion of Even-Even Spherical Nuclei*

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By extending a method developed recently for the electron gas problem to the case of even-even spherical nuclei, corrections to the random phase or harmonic description of the vibrational spectrum have been obtained. The calculation, which was based on the conventional pairing plus quadrupole force effective Hamiltonian, indicates a systematic sequence of higher approximations taking into account successively more complicated phonon-phonon and phonon-quasiparticle pair interactions. The approximation of this paper, which includes only the leading phonon-phonon interaction in its effect on the one and two phonon states, is in principle rich enough to allow all of the observed orderings of the two-phonon triplet. The case of Ni^{62} has been studied in detail, with all parameters except the strength of the quadrupole force taken directly from the work of Kisslinger and Sorensen and the latter constant readjusted so that the improved theory gives the energy of the first 2^+ state. A parameter-free accord to the ordering $(0^+, 2^+, 4^+)$ of the two-phonon state is obtained, though the energies are on the whole high. The main defects of the calculation are discussed.

I. INTRODUCTION

THE last few years have seen the accumulation of experimental data on the low-lying spectra of even-even nuclei in the vibrational regions.[1–4] It appears that, besides the well-determined lowest 2^+ state, some higher states have also been seen which could be assigned to the triplet 0^+, 2^+, 4^+ of the vibrational model of nuclei.[5–6] But these states are never degenerate and the ratios of their energies to the energy of the first 2^+ state deviate from the value 2 of the harmonic theory.

Two attempts to understand these results have recently been made. Kerman and Shakin[7] proposed a phenomenological theory based on the collective model of Bohr, adding to the harmonic Hamiltonian terms cubic in the quadrupole tensor. The model, which contains two parameters describing the anharmonicity, gives a reasonable fit to the vibrational spectrum of Ni^{62}, but according to the authors, will not reproduce the trends in the behavior of the 4^+ level. Against this model may be voiced the objection that its effective Hamiltonian would follow from a microscopic theory, such as the one of this paper, at best only in the adiabatic limit. Since this limit is mostly invalid in the vibrational region, the physical significance of the parameters of the theory is somewhat doubtful.

The second attempt is that of Beliaev and Zelevinsky[8] who developed a microscopic theory of vibrations of spherical nuclei using the conventional Hamiltonian with two fundamental parameters, namely, the strengths of the pairing and quadrupole forces. Their method, aside from questions of detail, alluded to at the appropriate place in the Appendix, corresponds to the adiabatic limit of the method of this paper. This questionable limit, however, enforces in their theory a definite ordering of levels not in general accord with experiment.

In this paper we begin the development of a microscopic theory of nuclear vibrations, using the density operator formalism for which the random phase approximation is the zeroth-order approximation. The

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formalism has been developed in a previous paper[9] where the electron gas problem was treated as an example. The generalization to the case of a finite nucleus is straightforward and is in outline the following:

Let a_α, $a_\alpha^\dagger$ be the annihilation and creation operators of a particle in the shell-model state α with energy ϵ_a. The density matrix equation for $\rho_{\alpha\beta}=a_\alpha^\dagger a_\beta$ is, for example, of the form

$$[\omega-(\epsilon_a-\epsilon_b)]\langle 1,2q|\rho_{\alpha\beta}|0\rangle = \sum \Gamma_{\alpha\beta}(\gamma\delta\lambda\mu)\langle 1,2q|a_\gamma^\dagger a_\delta^\dagger a_\lambda a_\mu|0\rangle, \quad (1.1)$$

where $|0\rangle$ is the ground state and $|1,2q\rangle$ is the first excited state of interest, the one-phonon state of angular momentum 2, z-component q; ω is the excitation energy. The Hartree-Fock factorization of the right-hand side leads to the usual random phase approximation (RPA). There remain, however, terms which cannot be decomposed in this way and of which we propose to take account by the following standard process:

$$\langle 1,2q|a_\gamma^\dagger\alpha_\delta^\dagger\alpha_\lambda\alpha_\mu|0\rangle = \sum_{n'}\langle 1,2q|a_\gamma^\dagger a_\mu|n'\rangle\langle n'|a_\delta^\dagger a_\lambda|0\rangle, \quad (1.2)$$

where $|n'\rangle$ is a complete set of states of the total Hamiltonian. We start from the RPA where the only nonvanishing matrix elements of $\rho_{\alpha\beta}$ are those between two states differing by one phonon, and which can all be reduced to the form $\langle 1|\rho_{\alpha\beta}|0\rangle$. To reach the first step beyond the RPA, we retain in (1.2) only those states $|n'\rangle$ for which one of the resulting matrix elements in the product is the collective one computed from the RPA. This means for the example at hand either that $|n'\rangle=|1,2q\rangle$ or else $|n'\rangle$ is a two-phonon incoherent state:

$$|n'\rangle=|2,JM\rangle \simeq(1/\sqrt{2})\sum_{qq'}\langle 22qq'|JM\rangle|1,2q\rangle\otimes|1,2q'\rangle; \quad (1.3)$$

(the first factor on the right is a Clebsch-Gordon coefficient) since, in the latter event,

$$\langle 1,2q|\rho_{\alpha\beta}|2,JM\rangle \cong\sqrt{2}\langle 22qM-q|JM\rangle\langle 0|\rho_{\alpha\beta}|1,M-q\rangle \quad (1.4)$$

is again of the form admitted.

There now occur, however, new matrix elements in which the phonon number differs by zero or by two (the crossover matrix element). When these are analyzed by the analog of (1.1) and by the same techniques, they are found to be coupled to the dominant amplitudes, to themselves, and to yet higher amplitudes. The approximation of this paper consists of retaining only the coupling back to the dominant amplitudes and of solving the resulting equations, in what amounts to a form of collective state perturbation theory.

In the above summary we have implicitly made another approximation to which we adhere in this paper, but which will be removed in future applications. The RPA determines in addition to the collective state a set of states of quasiparticle pairs, the whole forming a complete set for the problem at hand. These quasiparticle states can and should be retained in general as intermediate states in (1.2). In (temporarily) neglecting them, we can say that we are neglecting the phonon-quasiparticle coupling compared to the phonon-phonon interaction. That this is partly justifiable on an individual term basis is clear from (1.2) since the ratio is at least as small as that of the *amplitudes* for single particle collective $E2$ transitions and, perhaps as small as the square of this ratio.

There are, of course, a relatively large number of such terms, but here, one may be provisionally optimistic and anticipate the operation at this state of a random phase mechanism.

We have outlined above the procedure for obtaining a corrected theory of the first-phonon state. To obtain a theory of the two-phonon states we proceed analogously, this time starting with an equation for $\langle 2,JM|\rho_{\alpha\beta}|1,2q\rangle$. A major virtue of the present method is that it works with the transition amplitudes of *direct physical interest* in the problem in contrast to the higher random phase approximation.[10,11] It is also simpler, but whether it will prove as accurate remains undetermined.

In Sec. II, the form of the Hamiltonian is discussed, and the transformation from shell-model particles to quasiparticles reviewed. The corrections to the first 2^+ state and the splitting of the triplet $(0^+,2^+,4^+)$ are calculated as described above in Secs. III and IV, respectively. It is shown qualitatively that the triplet can have all the experimentally observed orderings, namely,

$$(0^+, 2^+, 4^+),$$
$$(2^+, 0^+, 4^+),$$
$$(0^+, 4^+, 2^+).$$

Section V deals with the computation of $E2$ transition probabilities, and finally numerical results and discussion for Ni^{62} are given in the last Sec. VI.

II. HAMILTONIAN

Following Kisslinger and Sorensen,[12] we suppose that the low-energy states of spherical nuclei are eigenstates of the Hamiltonian which, in terms of the creation and annihilation operators $a_\alpha^\dagger$, a_α of a particle in the shell-

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[12] L. S. Kisslinger and R. A. Sorenson, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **32**, No. 9 (1960).

model state α, is written as:

$$H=\sum_\alpha(\epsilon_a{}^0-\lambda)a_\alpha{}^\dagger a_\alpha-\tfrac{1}{2}G\sum_{\alpha\beta}(s_\alpha a_\alpha{}^\dagger a_{-\alpha}{}^\dagger)(s_\beta a_{-\beta}a_\beta) -\tfrac{1}{2}\mathrm{X}\sum_{\alpha\beta\gamma\delta}V_2(\alpha\beta,\gamma\delta)a_\alpha{}^\dagger a_\beta{}^\dagger a_\delta a_\gamma, \quad (2.1)$$
$$\equiv H_0+H_p+H_2,$$

where $\epsilon_a{}^0$ is the single-particle energy in the state α. The Hamiltonian thus contains two parameters, namely, G, the strength of the pairing force, and X, that of the long-range quadrupole force. Furthermore, λ is the chemical potential and $s_\alpha=(-1)^{j_a-m_\alpha}$. The quadrupole force, on the other hand, may be written as[13]

$$V_2(\alpha\beta,\gamma\delta)=\sum_q F(acdb)s_\gamma\langle j_a j_c m_\alpha-m_\gamma|2q\rangle \times s_\beta\langle j_a j_b m_\delta-m_\beta|2q\rangle, \quad (2.2)$$

where, for a separable central interaction, which we here assume,

$$F(acdb)=g(ac)g(db) \quad (2.3)$$

$$g(ac)=-\theta(ac)g(ca), \quad \theta(ac)=(-1)^{j_a+j_c}. \quad (2.4)$$

We neglect the tensor part of the force, e.g., the spin-dependent force.[14]

With the Hamiltonian (2.1), Kisslinger and Sorensen have succeeded in fitting almost all the first 2^+ states of spherical nuclei with one magic number, and in giving qualitatively correct results for the reduced transition probabilities.[15]

Now, in order to remove the degeneracy of the shell model states of (2.1) and determine the chemical potential, we make the Valatin-Bogoliubov transformation

$$d_\alpha{}^\dagger=u_a a_\alpha{}^\dagger-s_\alpha v_a a_{-\alpha}, \qquad u_a{}^2+v_a{}^2=1, \quad (2.5)$$

defining quasiparticle creation and annihilation operators, $d_\alpha{}^\dagger$, d_α which satisfy the usual anticommutation relations of fermions. The transformed Hamiltonian may be written in the form

$$H=U+H_{11}+H_{20}+H_{\mathrm{qp}}+H_2, \quad (2.6)$$

where the first three terms have the usual meanings[16] and H_{qp} contains products of four quasiparticle operators coming entirely from the pairing force. In the following, we neglect H_{qp}, which, being of short-range nature, should not appreciably affect the collective motion coming from H_2, though it can modify the quasiparticle spectrum.

The condition that $H_{20}=0$ defines u and v. We find

$$u_a{}^2=\tfrac{1}{2}\{1+[(\epsilon_a-\lambda)/E_a]\}, \qquad v_a{}^2=\tfrac{1}{2}\{1-[(\epsilon_a-\lambda)/E_a]\}, \quad (2.7)$$

where

$$\epsilon_a=\epsilon_a{}^0-2Gv_a{}^2+\frac{\mathrm{X}}{2}\sum_b(5/2j_a+1)F(abab), \quad (2.8)$$
$$E_a=[(\epsilon_a-\lambda)^2+\Delta^2]^{1/2},$$

and the gap Δ is given by the "gap equation"

$$\frac{G}{2}\sum_a\frac{\Omega_a}{E_a}=1, \quad (2.9)$$

with $\Omega_a=\frac{1}{2}(2j_a+1)$. $u_a{}^2$ and $v_a{}^2$ are then the probabilities of nonoccupancy and occupancy, respectively, of the state α and $-\alpha$ by a pair of particles. From this, the chemical potential is given by the implicit equation

$$2\sum_a\Omega_a v_a{}^2=N, \quad (2.10)$$

where N is the total number of particles in the shells under study. In the case where we assume that there is no interference between protons and neutrons in giving the collective motion, and we are interested only in one kind of particles, say in neutrons, N will then usually be the number of neutrons outside closed shells.

The long-range quadrupole part, on the other hand, may be written as

$$H_2=-\tfrac{1}{2}\mathrm{X}\sum_q Q_q Q_q{}^\dagger, \quad (2.11)$$

where the "quadrupole moment" Q_q, in terms of quasiparticle operators, is

$$Q_q=\sum_{ac}g(ac)[\xi(ac)A_q{}^{(+)}(ac)+\eta(ac)B_q{}^{(+)}(ac)], \quad (2.12)$$

where

$$\xi(ac)=\tfrac{1}{2}(u_a v_c+u_c v_a)=\xi(ca), \qquad \eta(ac)=\tfrac{1}{2}(u_a u_c-v_a v_c)=\eta(ca), \quad (2.13)$$

and

$$A_q{}^{(\pm)}(ac)=A_q{}^\dagger(ac)\pm(-1)^q A_{-q}(ac)=-\theta(ac)A_q{}^{(\pm)}(ca),$$
$$B_q{}^{(\pm)}(ac)=B_q{}^\dagger(ac)\pm(-1)^q B_{-q}(ac)=\mp\theta(ac)B_q{}^{(\pm)}(ca),$$
$$A_q{}^\dagger(ac)=\sum_{m_\alpha m_\gamma}\langle j_a j_c m_\alpha m_\gamma|2q\rangle d_\alpha{}^\dagger d_\gamma{}^\dagger=-\theta(ac)A_q{}^\dagger(ca), \quad (2.14)$$
$$B_q{}^\dagger(ac)=\sum_{m_\alpha m_\gamma}s_\gamma\langle j_a j_c m_\alpha-m_\gamma|2q\rangle d_\alpha{}^\dagger d_\gamma=-\theta(ac)(-1)^q B_{-q}(ca).$$

[13] We use the phase convention of A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, New Jersey, 1957).
[14] L. S. Kisslinger, Nucl. Phys. **35**, 114 (1962).
[15] See also T. Tamura and T. Udagawa, Prog. Theoret. Phys. (Kyoto) **26**, 947 (1961).
[16] R. Arvieu and M. Véneroni, Compt. Rend. **250**, 995 (1960). See also S. T. Belaiev, Kgl. Danske Videnskab. Selskab, Mat. Fys. Medd. **31**, No. 11 (1959).

It can be shown that the operators (2.14) satisfy the following commutation rules:

$$
\begin{aligned}
&[A_{-q'}{}^{(+)}(a'b'),A_q{}^{(+)}(ab)]=-[(1+p_{ab})(1+p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle B_{q-q'}{}^{(-)}(bb')],\\
&[A_{-q'}{}^{(-)}(a'b'),A_q{}^{(-)}(ab)]=[(1+p_{ab})(1+p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle B_{q-q'}{}^{(-)}(bb')],\\
&[A_{-q'}{}^{(-)}(a'b'),A_q{}^{(+)}(ab)]=-2\delta(q,q')(-1)^q(1+p_{ab})\delta(aa')\delta(bb')\\
&\qquad\qquad+[(1+p_{ab})(1+p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle B_{q-q'}{}^{(+)}(bb')], \qquad (2.15)\\
&[B_{-q'}{}^{(+)}(a'b'),A_q{}^{(\pm)}(ab)]=[(1+p_{ab})(1+p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle A_{q-q'}{}^{(\mp)}(bb')],\\
&[B_{-q'}{}^{(-)}(a'b'),A_q{}^{(\pm)}(ab)]=-[(1+p_{ab})(1-p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle A_{q-q'}{}^{(\pm)}(bb')],\\
&[B_{-q'}{}^{(\pm)}(a'b'),B_q{}^{(+)}(ab)]=-[(1+p_{ab})(1\pm p_{a'b'})\delta(aa')(-1)^{q'}z(abb')\theta(ab')\langle 22q'q-q'\,|\,2q\rangle B_{q-q'}{}^{(\mp)}(bb')],
\end{aligned}
$$

where $z(abb')=5\theta(ab)W(2j_a2j_b;\,j_{b'}2)$, with W a Racah coefficient, and the permutation operator p_{ab} is defined as

$$p_{ab}f(ab)=-\theta(ab)f(ba). \qquad (2.16)$$

We see from the 3rd commutation relation that, in first approximation, all operators commute except the $A_q{}^{(\pm)}$ which satisfy nearly the usual boson commutation relation. This approximation, as will be seen in the next section, is the random phase approximation (RPA).

III. CORRECTION TO THE FIRST 2^+ STATE

A. Random Phase Approximation

The Hamiltonian with which we study the collective vibrational motion is that for quasiparticles interacting through quadrupole forces

$$H_c=\textstyle\sum_\alpha E_a d_\alpha{}^\dagger d_\alpha-\frac{1}{2}\chi\sum_q Q_qQ_q{}^\dagger. \qquad (3.1)$$

Using the commutation relations (2.15), we obtain the following equations:

$$
\begin{aligned}
[H_c,A_q{}^{(+)}(ab)]=&E(ab)A_q{}^{(-)}(ab)+\textstyle\sum_{a'b'cd,q'}\chi g(a'b')g(cd)\langle 22q'q-q'\,|\,2q\rangle\\
&\times\{\xi(cd)\xi(a'b')\{P^+(ab,a'b')[A_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(-)}(bb')+B_{q-q'}{}^{(-)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&-\xi(cd)\eta(a'b')\{P^+(ab,a'b')[A_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(-)}(bb')+A_{q-q'}{}^{(-)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&+\eta(cd)\xi(a'b')\{P^+(ab,a'b')[B_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(-)}(bb')+B_{q-q'}{}^{(-)}(bb')B_{q'}{}^{(+)}(cd)]\}\\
&-\eta(cd)\eta(a'b')\{P^+(ab,a'b')[B_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(-)}(bb')+A_{q-q'}{}^{(-)}(bb')B_{q'}{}^{(+)}(cd)]\}), \qquad (3.2)\\
[H_c,A_q{}^{(-)}(ab)]=&E(ab)A_q{}^{(+)}(ab)-4\chi\xi(ab)g(ab)Q_q+\textstyle\sum_{a'b'cdq}\chi g(a'b')g(cd)\langle 22q'q-q'\,|\,2q\rangle\\
&\times\{\xi(cd)\xi(a'b')\{P^+(ab,a'b')[A_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(+)}(bb')+B_{q-q'}{}^{(+)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&-\xi(cd)\eta(a'b')\{P^+(ab,a'b')[A_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(+)}(bb')+A_{q-q'}{}^{(+)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&+\eta(cd)\xi(a'b')\{P^+(ab,a'b')[B_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(+)}(bb')+B_{q-q'}{}^{(+)}(bb')B_{q'}{}^{(+)}(cd)]\}\\
&-\eta(cd)\eta(a'b')\{P^\pm(ab,a'b')[B_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(+)}(bb')+A_{q-q'}{}^{(+)}(bb')B_{q'}{}^{(+)}(cd)]\}), \qquad (3.3)\\
[H_c,B_q{}^{(\pm)}(ab)]=&\epsilon(ab)B_q{}^{(\mp)}(ab)\pm\textstyle\sum_{c'b'cdq'}\chi g(a'b')g(cd)\langle 22q'q-q'\,|\,2q\rangle\\
&\times\{\xi(cd)\xi(a'b')\{P^\pm(ab,a'b')[A_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(\mp)}(bb')+A_{q-q'}{}^{(\mp)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&+\xi(cd)\eta(a'b')\{P^+(ab,a'b')[A_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(\mp)}(bb')+B_{q-q'}{}^{(\mp)}(bb')A_{q'}{}^{(+)}(cd)]\}\\
&+\eta(cd)\xi(a'b')\{P^\pm(ab,a'b')[B_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(\mp)}(bb')+A_{q-q'}{}^{(\mp)}(bb')B_{q'}{}^{(+)}(cd)]\}\\
&+\eta(cd)\eta(a'b')\{P^\pm(ab,a'b')[B_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(\mp)}(bb')+B_{q-q'}{}^{(\mp)}(bb')B_{q'}{}^{(+)}(cd)]\}), \qquad (3.4)
\end{aligned}
$$

where

$$E(ab)=E_a+E_b,$$
$$\epsilon(ab)=E_a-E_b,$$

and

$$P^\pm(ab,a'b')=(1\pm p_{ab})\delta(aa')z(abb')\theta(ab'). \qquad (3.5)$$

In a first approximation, we consider only the operators $A_q{}^{(\pm)}$ which satisfy the equations

$$
\begin{aligned}
&[H_c,A_q{}^{(+)}(ab)]=\omega_0A_q{}^{(+)}(ab)=E(ab)A_q{}^{(-)}(ab),\\
&[H_c,A_q{}^{(-)}(ab)]=\omega_0A_q{}^{(-)}(ab)=E(ab)A_q{}^{(+)}(ab)-4\chi\xi(ab)g(ab)\textstyle\sum_{cd}\xi(cd)g(cd)A_q{}^{(+)}(cd),
\end{aligned} \qquad (3.6)
$$

the solution of which is given by the equation

$$1=4\chi\textstyle\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_0{}^2]^{-1}. \qquad (3.7)$$

If the quadrupole force is strong, the phonon frequency ω_0 is small, $\omega_0 \ll E_{\min}(ab)$, (adiabatic limit) and is given by

$$\omega_0^2=[1-4X\sum_{ab}\xi^2(ab)g^2(ab)E^{-1}(ab)][4X\sum_{ab}\xi^2(ab)g^2(ab)E^{-3}(ab)]^{-1}. \tag{3.8}$$

Let $C_q^\dagger$ be the creation operator of a phonon, of the form

$$C_q^\dagger=\sum_{ab}[\lambda_+(ab)A_q^{(+)}(ab)+\lambda_-(ab)A_q^{(-)}(ab)],\quad \lambda_\pm(ab)=-\theta(ab)\lambda_\pm(ba). \tag{3.9}$$

Then, the ground state and the one-phonon state (the first 2^+ state) are defined by:

$$C_q|0\rangle=0,\quad C_q^\dagger|0\rangle=|1,2q\rangle. \tag{3.10}$$

From the condition that C_q, $C_q^\dagger$ satisfy the boson commutation rule, namely $[C_{q'},C_q^\dagger]=\delta_{qq'}$, we find

$$8\sum_{ab}\lambda_+(ab)\lambda_-(ab)=1. \tag{3.11}$$

Using (2.15) and (3.10), we have

$$\langle 1,2q|A_q^{(\pm)}(ab)|0\rangle=4\lambda_\mp(ab), \tag{3.12}$$

and, from (3.6)

$$\langle 1,2q|A_q^{(+)}(ab)|0\rangle=[E(ab)/\omega_0]\langle 1,2q|A_q^{(-)}(ab)|0\rangle=\rho 4XE(ab)\xi(ab)g(ab)[E^2(ab)-\omega_0^2]^{-1}, \tag{3.13}$$

where

$$\rho=\{8X^2\omega_0\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_0^2]^{-2}\}^{-1/2}. \tag{3.14}$$

Thus, in the random phase approximation, the problem is completely solved, giving the energy and wave function of the first 2^+ state. We remark that, in (3.12), we have chosen the matrix elements to be independent of q. This choice is arbitrary, but is made once for all, i.e., all other matrix elements must be defined correspondingly. We have, for example,

$$\langle 0|A_{-q}^{(\pm)}(ab)|1,2q\rangle=\pm(-1)^q\langle 1,2q|A_q^{(\pm)}(ab)|0\rangle, \tag{3.15}$$

which now depend on q by a phase factor.

B. First-Order Correction

With the RPA as zero approximation, we want to take account of the remaining terms in Eqs. (3.2) and (3.3), using the linearization procedure mentioned in the introduction, namely the spectral decomposition process. In the first-order correction, we neglect all terms which require more than one step in the iteration process, for example terms quadratic in $B^{(\pm)}$, because in the RPA, the matrix elements $\langle 1,2q|B_q^{(\pm)}(ab)|0\rangle$ are zero, and thus both of them must be iterated.

Taking the matrix element of (3.2) and (3.3) between the ground state and the one-phonon state we may write them in the form:

$$\begin{aligned}
&\omega\langle 1,2q|A_q^{(+)}(ab)|0\rangle=E(ab)\langle 1,2q|A_q^{(-)}(ab)|0\rangle+M_q^{(+)}(ab),\\
&\omega\langle 1,2q|A_q^{(-)}(ab)|0\rangle=E(ab)\langle 1,2q|A_q^{(+)}(ab)|0\rangle+M_q^{(-)}(ab)-4X\xi(ab)g(ab)\sum_{cd}\xi(cd)g(cd)\langle 1,2q|A_q^{(+)}(cd)|0\rangle\\
&\qquad\qquad -4X\xi(ab)g(ab)\sum_{cd}\eta(cd)g(cd)\langle 1,2q|B_q^{(+)}(cd)|0\rangle.
\end{aligned} \tag{3.16}$$

With the definition

$$\mathfrak{A}_q^{(+)}=\sum_{ab}\xi(ab)g(ab)\langle 1,2q|A_q^{(+)}(ab)|0\rangle, \tag{3.17}$$

the system of Eqs. (3.16) may be reduced to one equation defining ω:

$$\begin{aligned}
&\{1-4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega^2]^{-1}\}\mathfrak{A}_q^{(+)}\\
&\quad=\sum_{ab}E(ab)[\omega^2-E^2(ab)]^{-1}\xi(ab)g(ab)M_q^{(-)}(ab)+\sum_{ab}\omega[\omega^2-E^2(ab)]^{-1}\xi(ab)g(ab)M_q^{(+)}(ab)\\
&\qquad -4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[\omega^2-E^2(ab)]^{-1}\sum_{cd}\eta(cd)g(cd)\langle 1,2q|B_q^{(+)}(cd)|0\rangle.
\end{aligned} \tag{3.18}$$

The right-hand side, which is the correction to the RPA, must be small to assure the validity of the first approximation. We may thus replace in it the true phonon frequency ω by ω_0, and rewrite (3.18) as

$$\{1-4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega^2]^{-1}\}\mathfrak{A}_q^{(+)}\equiv\mathfrak{M}^{(+)}+\mathfrak{M}^{(-)}+\mathfrak{N}, \tag{3.19}$$

where, using (3.2) and (3.3),

$$\begin{aligned}
\mathfrak{M}^{(-)}=&\sum_{abb'cdq'}2E(ab)[\omega^2-E^2(ab)]^{-1}\theta(ab')z(abb')\xi(ab)g(ab)Xg(ab')g(cd)\langle 22q'q-q'|2q\rangle\\
&\times\{\xi(cd)\xi(ab')\langle 1,2q|[A_{q'}^{(+)}(cd)B_{q-q'}^{(+)}(bb')+B_{q-q'}^{(+)}(bb')A_{q'}^{(+)}(cd)]|0\rangle\\
&-\xi(cd)\eta(ab')\langle 1,2q|[A_{q'}^{(+)}(cd)A_{q-q'}^{(+)}(bb')+A_{q-q'}^{(+)}(bb')A_{q'}^{(+)}(cd)]|0\rangle\\
&-\eta(cd)\eta(ab')\langle 1,2q|[B_{q'}^{(+)}(cd)A_{q-q'}^{(+)}(bb')+A_{q-q'}^{(+)}(bb')B_{q'}^{(+)}(cd)]|0\rangle\},
\end{aligned} \tag{3.20}$$

$$\mathfrak{M}^{(+)}=\sum_{abb'cd,q'} 2\omega[\omega^2-E^2(ab)]^{-1}\theta(ab')z(abb')\xi(ab)g(ab)\mathrm{X}g(ab')g(cd)\langle 22q'q-q'|2q\rangle$$
$$\times\{\xi(cd)\xi(ab')\langle 1,2q|[A_{q'}{}^{(+)}(cd)B_{q-q'}{}^{(-)}(bb')+B_{q-q'}{}^{(-)}(bb')A_{q'}{}^{(+)}(cd)]|0\rangle$$
$$-\xi(cd)\eta(ab')\langle 1,2q|[A_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(-)}(bb')+A_{q-q'}{}^{(-)}(bb')A_{q'}{}^{(+)}(cd)]|0\rangle$$
$$-\eta(cd)\eta(ab')\langle 1,2q|[B_{q'}{}^{(+)}(cd)A_{q-q'}{}^{(-)}(bb')+A_{q-q'}{}^{(-)}(bb')B_{q'}{}^{(+)}(cd)]|0\rangle\}\,, \quad (3.21)$$

$$\mathfrak{N}=\sum_{ab}\eta(ab)g(ab)\langle 1,2q|B_q{}^{(+)}(ab)|0\rangle\,. \quad (3.22)$$

In the spectral decomposition, two intermediate states contribute to the first order, namely, the one-phonon state (3.10) and the two-phonon state with $J=2$:

$$|2,2M\rangle=(1/\sqrt{2})\sum_{q_1q_2}\langle 22q_1q_2|2M\rangle|1,2q_1\rangle\otimes|1,2q_2\rangle\,. \quad (3.23)$$

For the one-phonon intermediate state, four new matrix elements between one-phonon states need to be calculated. It can be shown from (3.2) and (3.4), however, that in our approximation

$$\langle 1,2q|B_{q'}{}^{(-)}(ab)|1,2q-q'\rangle=0\,,\quad \langle 1,2q|A_{q'}{}^{(-)}(ab)|1,2q-q'\rangle=0\,, \quad (3.24)$$

so that two of these do not contribute.

We now calculate the remaining two amplitudes. From (3.4), for example,

$$\epsilon(ab)\langle 1,2q|B_{q'}{}^{(+)}(ab)|1,2q-q'\rangle$$
$$=\sum_{a'b'cdq''}\mathrm{X}g(a'b')g(cd)\langle 22q''q'-q''|2q'\rangle\xi(cd)\xi(ab')\{(1-p_{ab})\delta(aa')z(abb')\theta(ab')$$
$$\times\langle 1,2q|[A_{q''}{}^{(+)}(cd)A_{q'-q''}{}^{(+)}(bb')+A_{q'-q''}{}^{(+)}(bb')A_{q''}{}^{(+)}(cd)]|1,2q-q'\rangle\}\,. \quad (3.25)$$

The decomposition process may be applied to the right-hand side, involving zero- and two-phonon intermediate states. Using (3.13), and after straightforward calculations, we find

$$\langle 1,2q|B_{q'}{}^{(+)}(ab)|1,2q-q'\rangle$$
$$=16\rho\mathrm{X}^2\sum_{b'}z(abb')\theta(ab')\xi(ab')\xi(bb')\langle 22q'q-q'|2q\rangle g(ab')g(bb')[\omega^2+E(ab')E(bb')]$$
$$\times\{[E^2(ab')-\omega^2][E^2(bb')-\omega^2]\}^{-1}\mathfrak{A}_q{}^{(+)}\equiv Y_1(ab)\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}\,. \quad (3.26)$$

On the other hand, Eq. (3.3) gives:

$$E(ab)\langle 1,2q|A_{q'}{}^{(+)}(ab)|1,2q-q'\rangle$$
$$=4\mathrm{X}\xi(ab)g(ab)\sum_{cd}\xi(cd)g(cd)\langle 1,2q|A_{q'}{}^{(+)}(cd)|1,2q-q'\rangle+4\mathrm{X}\xi(ab)g(ab)\sum_{cd}\eta(cd)g(cd)Y_1(cd)\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}$$
$$+2E(ab)X_1{}^{(+)}(ab)\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}\,, \quad (3.27)$$

where

$$X_1{}^{(\pm)}(ab)=8E^{-1}(ab)\sum_{a'b'}\mathrm{X}\eta(a'b')g(a'b')[(1+p_{ab})\delta(aa')z(abb')\theta(ab')\lambda_{\mp}(bb')]\,. \quad (3.28)$$

Equation (3.27) can be solved immediately, and we have

$$\sum_{ab}\xi(ab)g(ab)\langle 1,2q|A_{q'}{}^{(+)}(ab)|1,2q-q'\rangle$$
$$=[1-4\mathrm{X}\sum_{cd}\xi^2(cd)g^2(cd)E^{-1}(cd)]^{-1}\{4\mathrm{X}\sum_{ab}\xi^2(ab)g^2(ab)E^{-1}(ab)\sum_{a'b'}\eta(a'b')g(a'b')Y_1(a'b')$$
$$+2\sum_{ab}\xi(ab)g(ab)X_1{}^{(+)}(ab)\}\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}\equiv x_1\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)} \quad (3.29)$$

$$\langle 1,2q|A_{q'}{}^{(+)}(ab)|1,2q-q'\rangle\equiv y_1(ab)\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}$$
$$=\{4\xi(ab)\mathrm{X}g(ab)E^{-1}(ab)[x_1+\sum_{cd}\eta(cd)g(cd)Y_1(cd)]+2X_1{}^{(+)}(ab)\}\langle 22q'q-q'|2q\rangle\mathfrak{A}_q{}^{(+)}\,. \quad (3.30)$$

Collecting results, the one-phonon intermediate state contribution to (3.20) and (3.21) is

$$\mathfrak{M}_1{}^{(-)}=-\sum_{abb',cd}8E(ab)[E^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\xi(ab)g(ab)\mathrm{X}g(ab')g(cd)$$
$$\times\{\xi(cd)\xi(ab')\lambda_-(cd)Y_1(bb')-\eta(cd)\eta(ab')\lambda_-(bb')Y_1(cd)$$
$$-\xi(cd)\eta(ab')[\lambda_-(bb')y_1(cd)+\lambda_-(cd)y_1(bb')]\}\mathfrak{A}_q{}^{(+)}\equiv m_1{}^{(-)}\mathfrak{A}_q{}^{(+)}\,, \quad (3.31)$$

$$\mathfrak{M}_1{}^{(+)}=\sum_{abb'cd}8\omega[E^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\xi(ab)g(ab)\mathrm{X}g(ab')g(cd)\eta(ab')\lambda_+(bb')$$
$$\times[\xi(cd)y_1(bb')+\eta(cd)Y_1(bb')]\mathfrak{A}_q{}^{(+)}\equiv m_1{}^{(+)}\mathfrak{A}_q{}^{(+)}\,. \quad (3.32)$$

Next, we calculate the two-phonon contribution. The new matrix elements of $A^{(\pm)}$ and $B^{(\pm)}$ are those between the ground state and the two-phonon state with spin $J=2$.

From Eq. (3.4), we have

$$\langle 2,2q|B_q{}^{(\pm)}(ab)|0\rangle=\pm\{Y_2{}^{(\mp)}(ab)-[\epsilon(ab)/\Omega_2]Y_2{}^{(\pm)}(ab)\}\sqrt{2}\mathfrak{A}_q{}^{(+)}\,, \quad (3.33)$$

where Ω_2 is the excitation energy of the second 2^+ state, and

$$Y_2^{(\pm)}(ab)=8X\Omega_2[\Omega_2^2-\epsilon^2(ab)]^{-1}\sum_{a'b'}\xi(a'b')g(a'b')[(1\mp p_{ab})\delta(aa')z(abb')\theta(ab')\lambda_{\mp}(bb')]. \tag{3.34}$$

The intermediate state which has contributed here is clearly the one-phonon state.

In the same way, we find from (3.2) and (3.3):

$$\begin{aligned}\sum_{ab}\xi(ab)g(ab)\langle 2,2q|A_q^{(+)}(ab)|0\rangle&\equiv x_2\sqrt{2}\,\mathfrak{A}_q^{(+)}\\ &=\{1-4X\textstyle\sum_{cd}\xi^2(cd)g^2(cd)E(cd)[E^2(cd)-\Omega_2^2]^{-1}\}^{-1}\{4X\textstyle\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\Omega_2^2]^{-1}\\ &\quad\times\textstyle\sum_{a'b'}\eta(a'b')g(a'b')\{Y_2^{(-)}(a'b')-[\epsilon(a'b')/\Omega_2]Y_2^{(+)}(a'b')\}+\sum_{ab}\xi(ab)g(ab)E^2(ab)X_1^{(+)}(ab)[E^2(ab)-\Omega_2^2]^{-1}\\ &\quad+\textstyle\sum_{ab}\xi(ab)g(ab)\Omega_2E(ab)X_1^{(-)}(ab)[E^2(ab)-\Omega_2^2]^{-1}\}\sqrt{2}\,\mathfrak{A}_q^{(+)}.\end{aligned} \tag{3.35}$$

We then find that

$$\begin{aligned}\langle 2,2q|A_q^{(+)}(ab)|0\rangle&=E(ab)[E^2(ab)-\Omega_2^2]^{-1}[4X\xi(ab)g(ab)(x_2+\textstyle\sum_{cd}\eta(cd)g(cd)\{Y_2^{(-)}(cd)-[\epsilon(cd)/\Omega_2]Y_2^{(+)}(cd)\})\\ &\quad+\Omega_2X_1^{(-)}(ab)+E(ab)X_1^{(+)}(ab)]\sqrt{2}\,\mathfrak{A}_q^{(+)}\equiv y_2(ab)\sqrt{2}\,\mathfrak{A}_q^{(+)},\end{aligned} \tag{3.36}$$

and

$$\langle 2,2q|A_q^{(-)}(ab)|0\rangle=\{[\Omega_2/E(ab)]y_2(ab)+X_1^{(-)}(ab)\}\sqrt{2}\,\mathfrak{A}_q^{(+)}, \tag{3.37}$$

and that the two-phonon intermediate state contributes to (3.20) the following quantities:

$$\begin{aligned}\mathfrak{M}_2^{(-)}\equiv 2m_2^{(-)}\mathfrak{A}_q^{(+)}&=-\textstyle\sum_{abb'cd}16E(ab)[E^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\xi(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times(\xi(cd)\xi(ab')\lambda_-(cd)\{Y_2^{(-)}(bb')-[\epsilon(bb')/\Omega_2]Y_2^{(+)}(bb')\}\\ &\quad-\xi(cd)\eta(ab')[\lambda_-(cd)y_2(bb')+\lambda_-(bb')y_2(cd)]\\ &\quad-\eta(cd)\eta(ab')\lambda_-(bb')\{Y_2^{(-)}(cd)-[\epsilon(cd)/\Omega_2]Y_2^{(+)}(cd)\})\mathfrak{A}_q^{(+)},\end{aligned} \tag{3.38}$$

$$\begin{aligned}\mathfrak{M}_2^{(+)}\equiv 2m_2^{(+)}\mathfrak{A}_q^{(+)}&=\textstyle\sum_{abb'cd}16\omega[E^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\xi(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times[\xi(cd)\xi(ab')\lambda_-(cd)\{Y_2^{(+)}(bb')-[\epsilon(bb')/\Omega_2]Y_2^{(-)}(bb')\}\\ &\quad+\xi(cd)\eta(ab')(\lambda_-(cd)\{[\Omega_2/E(bb')]y_2(bb')+X_1^{(-)}(bb')\}-\lambda_+(bb')y_2(cd))\\ &\quad-\eta(cd)\eta(ab')\lambda_+(bb')\{Y_2^{(-)}(cd)-[\epsilon(cd)/\Omega_2]Y_2^{(+)}(cd)\}]\mathfrak{A}_q^{(+)}.\end{aligned} \tag{3.39}$$

Using the above results for different matrix elements appearing in the decomposition process, the expression (3.22) for $\mathfrak{N}$ can be calculated easily. We have, from (3.4)

$$[\omega^2-\epsilon^2(ab)]\langle 1,2q|B_q^{(+)}(ab)|0\rangle=\epsilon(ab)n^{(-)}(ab)+\omega n^{(+)}(ab), \tag{3.40}$$

where $n^{(\pm)}(ab)$ are matrix elements of the nonlinear terms in the equations for $B^{(\pm)}(ab)$ between the ground state and the one-phonon state. Then

$$\mathfrak{N}=-\textstyle\sum_{ab}\eta(ab)g(ab)[E^2(ab)-\omega^2]^{-1}[\epsilon(ab)n^{(-)}(ab)+\omega n^{(+)}(ab)]\equiv\mathfrak{N}^{(-)}+\mathfrak{N}^{(+)}, \tag{3.41}$$

where, after some algebra, and again distinguishing contributions from the one- and two-phonon intermediate states, we find

$$\begin{aligned}\mathfrak{N}_1^{(-)}\equiv n_1^{(-)}\mathfrak{A}_q^{(+)}&=\textstyle\sum_{abb'cd}8\epsilon(ab)[\epsilon^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\eta(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times\{\xi(cd)\xi(ab')[\lambda_-(bb')y_1(cd)+\lambda_-(cd)y_1(bb')]+\xi(cd)\eta(ab')\lambda_-(cd)Y_1(bb')\\ &\quad+\eta(cd)\xi(ab')\lambda_-(bb')Y_1(cd)\}\mathfrak{A}_q^{(+)},\end{aligned} \tag{3.42}$$

$$\begin{aligned}\mathfrak{N}_2^{(-)}\equiv 2n_2^{(-)}\mathfrak{A}_q^{(+)}&=\textstyle\sum_{abb'cd}16\epsilon(ab)[\epsilon^2(a_d)-\omega^2]^{-1}z(abb')\theta(ab')\eta(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times(\xi(cd)\xi(ab')[\lambda_-(cd)y_2(bb')+\lambda_-(bb')y_2(cd)]\\ &\quad+\xi(cd)\eta(ab')\lambda_-(cd)\{Y_2^{(-)}(bb')-[\epsilon(bb')/\Omega_2]Y_2^{(+)}(bb')\}\\ &\quad+\eta(cd)\eta(ab')\lambda_-(bb')\{Y_2^{(-)}(cd)-[\epsilon(cd)/\Omega_2]Y_2^{(+)}(cd)\})\mathfrak{A}_q^{(+)},\end{aligned} \tag{3.43}$$

$$\begin{aligned}\mathfrak{N}_1^{(+)}\equiv n_1^{(+)}\mathfrak{A}_q^{(+)}&=-\textstyle\sum_{abb'cd}8\omega[\epsilon^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\eta(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times\xi(ab')\lambda_+(bb')[\xi(cd)y_1(cd)+\eta(cd)Y_1(cd)]\}\mathfrak{A}_q^{(+)},\end{aligned} \tag{3.44}$$

$$\begin{aligned}\mathfrak{N}_2^{(+)}\equiv 2n_2^{(+)}\mathfrak{A}_q^{(+)}&=\textstyle\sum_{abb'cd}16\omega[\epsilon^2(ab)-\omega^2]^{-1}z(abb')\theta(ab')\eta(ab)g(ab)Xg(ab')g(cd)\\ &\quad\times[\xi(cd)\xi(ab')(\lambda_+(bb')y_2(cd)-\lambda_-(cd)\{[\Omega_2/E(bb')]y_2(bb')+X_1^{(-)}(bb')\})\\ &\quad+\xi(cd)\eta(ab')\lambda_-(cd)\{Y_2^{(+)}(bb')-[\epsilon(bb')/\Omega_2]Y_2^{(-)}(bb')\}\\ &\quad+\eta(cd)\xi(ab')\lambda_+(bb')\{Y_2^{(-)}(cd)-[\epsilon(cd)/\Omega_2]Y_2^{(+)}(cd)\}]\mathfrak{A}_q^{(+)}.\end{aligned} \tag{3.45}$$

We see that, in the above expressions for $\mathfrak{M}^{(\pm)}$ and $\mathfrak{N}$, if we replace $\lambda_\pm$ by their values obtained from the RPA, expression (3.13), they are then all linear in $\mathcal{C}_q^{(+)}$. Equation (3.19) can thus be written in the form

$$1+\alpha=4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega^2]^{-1}, \tag{3.46}$$

$$\alpha=-(m_1^{(+)}+m_1^{(-)}+n_1^{(+)}+n_1^{(-)})-2(m_2^{(+)}+m_2^{(-)}+n_2^{(+)}+n_2^{(-)}), \tag{3.47}$$

where, in order to assure the validity of our approximation, α should be $\ll 1$. If it turns out that this is not the case, we have to take into account higher order corrections.

With

$$\omega=\omega_0+\omega', \tag{3.48}$$

Eq. (3.46) can be solved approximately to yield

$$\alpha=8X\omega_0\omega'\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_0^2]^{-2}. \tag{3.49}$$

We remark that, to be consistent, α should be considered as a function of the true phonon frequency ω instead of that given by the RPA, and this has been done in the actual numerical evaluation to be discussed in Sec. VI.

IV. SPLITTING OF THE TRIPLET 0^+, 2^+, 4^+

This section is devoted to an explicit calculation to first order of the splitting of the triplet which is degenerate in the RPA.

Let $|1, 2M-q\rangle$ and $|2,JM\rangle$ be, respectively, the true one-phonon state and two-phonon state with spin J ($J=0$, 2, 4), and let $\omega_J=E_J'-E_2$ be the energy difference. We define a matrix element of $A_q^{(+)}(ab)$ between these states according to

$$C_{JM}^{(\pm)}(ab)=\sum_q\langle 22qM-q|JM\rangle\langle 2,JM|A_q^{(\pm)}(ab)|1,M-q\rangle. \tag{4.1}$$

From Eqs. (3.2), (3.3), we then have

$$\{1-4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_J^2]^{-1}\}\mathcal{C}_{JM}^{(+)}=\mathfrak{M}_{JM}^{(-)}+\mathfrak{M}_{JM}^{(+)}+\mathfrak{N}_{JM}, \tag{4.2}$$

where

$$\mathcal{C}_{JM}^{(+)}=\sum_{ab}\xi(ab)g(ab)C_{JM}^{(+)}(ab), \tag{4.3}$$

and where $\mathfrak{M}_{JM}^{(\pm)}$ and $\mathfrak{N}_{JM}$ are defined in the same way as $\mathfrak{M}^{(\pm)}$ and $\mathfrak{N}$ in Sec. III with slight modifications coming from the definition of $C_{JM}^{(\pm)}$ in (4.1). For example, $\mathfrak{N}_{JM}$ is given by

$$\mathfrak{N}_{JM}=\sum_{abq}\eta(ab)g(ab)\langle 22qM-q|JM\rangle\langle 2,JM|B_q^{(+)}(ab)|1,2M-q\rangle. \tag{4.4}$$

The explicit calculations of the last section can be repeated exactly. As intermediate states, besides the two and three phonon states which are the analogs of the one- and two-phonon states of the last section, we have to take also into account the contributions from zero- and one-phonon intermediate states. A three-phonon state with spin k is of the form

$$\begin{aligned}&|3,k\mu\rangle_J=[1+2a(J,k)]^{-1/2}\sum_{qM}\langle J2Mq|k\mu\rangle|2,JM\rangle\otimes|1,2q\rangle,\\&a(J,k)=(-1)^k(2J+1)W(22k2,JJ),\end{aligned} \tag{4.5}$$

where J is the spin of the two-phonon state. We record here the results of lengthy but straightforward calculations.

$$\begin{aligned}&\mathfrak{M}_{JM,0}^{(\pm)}=\delta(J,2)m_2^{(\pm)}\mathcal{C}_{JM}^{(+)},\\&\mathfrak{M}_{JM,1}^{(\pm)}=W_Jm_1^{(\pm)}\mathcal{C}_{JM}^{(+)},\\&\mathfrak{M}_{JM,2}^{(\pm)}=\zeta_Jm_1^{(\pm)}\mathcal{C}_{JM}^{(+)},\\&\mathfrak{M}_{JM,3}^{(\pm)}=\rho_Jm_2^{(\pm)}\mathcal{C}_{JM}^{(+)},\end{aligned} \tag{4.6}$$

where

$$\begin{aligned}&W_J=5W(22J2,22),\\&\zeta_J=10\sum_{k=0,2,4}(2k+1)W^2(22J2,2k),\\&\rho_J=\sum_{k=0,2,4}\sum_{k'=0,2,3,4,6}5(2k+1)(2k'+1)W(22Jk',22)W(22k'2,k2)\\&\qquad\times[(2k+1)(2J+1)]^{-1/2}[1+2a(kk')]^{1/2}[1+2a(J,k')]^{1/2}.\end{aligned} \tag{4.7}$$

The corresponding expressions are valid for $\mathfrak{N}_{JM}$, for example,

$$\mathfrak{N}_{JM,0}^{(\pm)}=\delta(J,2)n_2^{(\pm)}\mathcal{C}_{JM}^{(+)}. \tag{4.8}$$

Thus, $\mathfrak{M}_{JM}{}^{(\pm)}$ and $\mathfrak{N}_{JM}$ are again linear in $\mathfrak{C}_{JM}{}^{(+)}$ and Eq. (4.2) can now be put in the form

$$1+\alpha_J=4X\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_J{}^2]^{-1}, \tag{4.9}$$

which, to first order in α_J, can be written

$$\alpha_J=8X\omega_0\omega_J'\sum_{ab}\xi^2(ab)g^2(ab)E(ab)[E^2(ab)-\omega_0{}^2]^{-2}, \tag{4.10}$$

where $\omega_J'=\omega_J-\omega_0$. The excitation energy of the two-phonon state J is

$$E_J'=\omega+\omega_J. \tag{4.11}$$

We remark finally that from the expressions ($m_i=m_i{}^{(+)}+m_i{}^{(-)}$)

$$\begin{aligned}\alpha&=-(m_1+n_1)-2(m_2+n_2),\\ \alpha_J&=-(W_J+\zeta_J)(m_1+n_1)-(\delta(J,2)+\rho_J)(m_2+n_2),\end{aligned} \tag{4.12}$$

depending on the relative values of (m_1+n_1) and (m_2+n_2), and with numerical values of W_J, ζ_J, ρ_J given by (4.7), α_J and α can have all possible relative order and signs giving the sequences (1.5). The detailed argument for this assertion is given in the Appendix.

V. ELECTROMAGNETIC TRANSITION PROBABILITIES

In order to calculate the transition probabilities between different collective states, we need from the beginning consider both neutrons and protons simultaneously. Though we have not mentioned this explicitly, our procedure, once the isotopic spin is understood in the indices $\alpha, \beta\cdots$, applies to this general case. However, we consider only the case of one closed shell nuclei (protons or neutrons) for which the collective oscillations are the effects of outside nucleons, with effective charge e. The need to introduce effective charge comes from the fact that, due to the interactions between the outside nucleons and the core, the latter is polarized.[17]

The quadrupole operator which induces electromagnetic transitions is

$$Q_q=5^{-1/2}\sum_{\alpha\beta}\langle a\|er^2Y_2\|b\rangle s_\beta\langle j_a j_b m_\alpha -m_\beta|2q\rangle a_\alpha{}^\dagger a_\beta, \tag{5.1}$$

which becomes, after carrying out the quasiparticle transformation:

$$Q_q=5^{-1/2}\sum_{ab}\langle a\|er^2Y_2\|b\rangle\{\xi(ab)A_q{}^{(+)}(ab)+\eta(ab)B_q{}^{(+)}(ab)\}. \tag{5.2}$$

The reduced transition probability between two states $|n,k\mu\rangle$ and $|n',k'\mu'\rangle$ is given by:

$$B(E_2, k_n\rightarrow k_{n'})=\sum_{\mu\mu'}|\langle n',k'\mu'|Q_q|n,k\mu\rangle|^2. \tag{5.3}$$

We now show how the considerations of the previous sections yield the matrix elements required in (5.3) to the desired accuracy. We take the example of the matrix element $\langle 1,2q|A_q{}^{(+)}(ab)|0\rangle$. The procedure is easily seen to apply to all other matrix elements.

Let us go back to Eq. (3.16) and write it in the form

$$\begin{aligned}\langle 1,2q|A_q{}^{(+)}(ab)|0\rangle=&-[E^2(ab)-\omega^2]^{-1}\{\omega M_q{}^{(+)}(ab)+E(ab)M_q{}^{(-)}(ab)+E(ab)N_q(ab)\\ &-4X\xi(ab)g(ab)E(ab)\sum_{cd}\xi(cd)g(cd)\langle 1,2q|A_q{}^{(+)}(cd)|0\rangle\}\equiv-\mu(ab)+D(ab)\mathfrak{A}_q{}^{(+)},\end{aligned} \tag{5.4}$$

where

$$D(ab)=4X\xi(ab)g(ab)E(ab)[E^2(ab)-\omega^2]^{-1}, \tag{5.5}$$

and

$$\mathfrak{A}_q{}^{(+)}=\sum_{ab}\xi(ab)g(ab)\langle 1,2q|A_q{}^{(+)}(ab)|0\rangle. \tag{5.6}$$

Since the final equation (3.19) is linear in $\mathfrak{A}_q{}^{(+)}$, we can again use Eq. (3.11) for normalization. We obtain in this way to first order, the equation

$$\omega\mathfrak{A}_q{}^{(+)2}\sum_{ab}E^{-1}(ab)D^2(ab)=2+\rho\sum_{ab}E^{-1}(ab)\{2\omega\mu(ab)+M_q{}^{(+)}(ab)\}D(ab), \tag{5.7}$$

or

$$\begin{aligned}\mathfrak{A}_q{}^{(+)}&=\mathfrak{D}^{-1/2}\{1-\rho^{-1}\mathfrak{N}_q+\rho X\sum_{ab}\xi(ab)g(ab)[E^2(ab)-\omega^2]^{-2}[(E^2(ab)+\omega^2)M_q{}^{(+)}+2\omega E(ab)M_q{}^{(-)}(ab)]\},\\ \mathfrak{D}&=\tfrac{1}{2}\omega\sum_{ab}E^{-1}(ab)D^2(ab).\end{aligned} \tag{5.8}$$

[17] K. Alder, A. Bohr, T. Huus, B. Mottelson, and A. Winther, Rev. Mod. Phys. **28**, 432 (1956).

TABLE I. Comparison of theoretical and experimental values for the one- and two-phonon states of Ni^{62}. The experimental results are taken from P. H. Stelson and F. K. McGowan [Bull. Am. Phys. Soc. 4, 232 (1959)]. (The energies are in MeV.)

	E_2	E_0'	E_2'	E_4'	$\frac{B(E_2, 2'^+ \to 2^+)}{B(E_2, 2^+ \to 0)}$	$\frac{B(E_2, 2'^+ \to 2^+)}{B(E_2, 2'^+ \to 0)}$	$\frac{B(E_2, 2^+ \to 0)}{e^2\,10^{-48}\ \mathrm{cm}^4}$
Theory X=1.66	1.172	2.278	2.332	2.402	1.97	42	0.089
RPA X=1.83	1.172	2.344	2.344	2.344	2.00	∞	0.070
Exp.	1.172	2.048	2.302	2.336			0.083

Thus, according to (5.3), the reduced transition probability $2^+ \to 0^+$ is, with ν defined by $\langle a\|r^2Y_2\|b\rangle = \nu g(ab)$,

$$B(E_2, 2^+ \to 0^+) = \mathfrak{D}^{-1}(1+\beta)^2\nu^2,$$

where

$$\beta = \rho X \sum_{ab} \xi(ab)g(ab)[E^2(ab)-\omega^2]^{-1}\{[E^2(ab)+\omega^2]M_q^{(+)}(ab)+2\omega E(ab)M_q^{(-)}(ab)\}. \tag{5.9}$$

In the same way, we have

$$B(E_2, J \to 2^+) = 2\mathfrak{D}^{-1}(1+\beta_J)^2\nu^2, \tag{5.10}$$

where β_J is given by (5.9) with the $M_q^{(\pm)}(ab)$ replaced by the corresponding $M_{JM}^{(\pm)}(ab)$.

Finally, we have to calculate $B(2^{+\prime} \to 0^+)$. This is given by (3.34), (3.35) and (3.37), namely

$$B(E_2, 2^{+\prime} \to 0^+) = 2\nu^2[\textstyle\sum_{ab} g(ab)(\xi(ab)y_2(ab)+\eta(ab)\{Y_2^{(-)}(ab)-[\epsilon(ab)/\Omega_2]Y_2^{(+)}(ab)\})]^2. \tag{5.11}$$

VI. NUMERICAL RESULTS FOR NI^{62} AND DISCUSSION

It is clear from Secs. III and IV that in order to calculate the corrections to the RPA, we need to know the shell model single-particle energies. In fact, the quantities α and α_J depend strongly on these energies, and also on the pairing force G which is used to define the gap Δ and the chemical potential λ.

In order to have a semiquantitative idea about the first-order correction, we have tentatively considered the case of Ni^{62} which has a closed shell for protons,[18] and treat the anharmonic effect through the outside neutrons with the shell model results given by Kisslinger and Sorensen.[12] This means that we adopt the value of the pairing force G given by these authors, and keep the quadrupole force as a parameter which will be defined such that, together with the correction, the first 2^+ state is fitted with the experimental value.

We have solved Eqs. (3.4) and (4.9) on the IBM-1620 computer of the University of Pennsylvania. The results are given in Table I, together with experimental values when known.

The value of X used to fit the first 2^+ level is X = 1.660 which corresponds to $\omega_0 = 1.36$ MeV.

We see that though the ordering of the two-phonon triplet is correctly reproduced, deviations from their absolute position are still appreciable especially for the 0^+ two-phonon state. We can advance several reasons for this discrepancy assuming the adequacy of the effective Hamiltonian:

(a) First, as remarked previously, we ought to use "correct" values for single-particle energies. In the work of Kisslinger and Sorensen, these have been chosen merely to fit the first 2^+ state in their approximation which is equivalent to the RPA.

(b) Next, we should note that it is improbable that the collective oscillations are due singly to the 6 outside neutrons and we ought to take into account the oscillations of the core. In order to do this, we have to modify slightly the schemes of Secs. III and IV to take care of the interactions between neutrons and protons.[15]

(c) Another kind of correction which can also be important, at least for medium-weight nuclei, and which we have neglected in the above calculations comes from the fact that, in the spectral decomposition, besides the collective intermediate states, we should also include "pair states" as mentioned in the introduction.

To take the above delinquencies into account requires a considerable increase in numerical and/or algebraic effort, the fruit of which will be reported in a subsequent publication.

[18] There are, of course, nuclei where the data is more complete, for example, those analyzed by Yoshizawa, Ref. 2, but these involve both neutrons and protons in unfilled shell and a larger number of single-particle levels. Our choice of Ni^{62} as a first example has thus been determined by numerical expediency.

APPENDIX

In this Appendix, we consider a simplified model of a nucleus consisting of one single shell with energy $(E/2)$. We shall prove in this particular case an assertion in the text, namely, that the first-order correction to the RPA could give rise to all observed orderings of the two-phonon triplet. The quantities m, n defined in the text

can be expressed in terms of u, v, z, g, which are now single valued. We find

$$
\begin{aligned}
&n_1^{(-)}=n_2^{(-)}=0,\\
&m_1^{(+)}=(u^2-v^2)^2g^2z^2\frac{\mathrm{X}}{E}\frac{\omega}{E}\left[\frac{E^2}{\omega^2}\left(3+\frac{\omega^2}{E^2}\right)+\left(1-\frac{\omega^2}{E^2}\right)^{-1}\left(1+\frac{\omega^2}{E^2}\right)\right],\\
&m_1^{(-)}=(u^2-v^2)^2g^2z^2\frac{\mathrm{X}}{E}\frac{2E^3}{\omega^3}\left(3+\frac{\omega^2}{E^2}\right)+\left[\tfrac{1}{4}(u^2-v^2)^2-u^2v^2\right]g^2z^2\frac{\mathrm{X}}{E}\frac{4E}{\omega}\left(1-\frac{\omega^2}{E^2}\right)^{-1}\left(1+\frac{\omega^2}{E^2}\right),\\
&n_1^{(+)}=(u^2-v^2)^2g^2z^2\frac{\mathrm{X}}{E}\frac{E}{\omega}\left[\left(1+\frac{\omega^2}{E^2}\right)+\frac{E^2}{\omega^2}\left(1-\frac{\omega^2}{E^2}\right)\left(3+\frac{\omega^2}{E^2}\right)\right],\\
&m_2^{(+)}=-(u^2-v^2)^2g^2z^2\frac{\mathrm{X}}{E}\frac{E}{\omega},\\
&m_2^{(-)}=-\frac{\mathrm{X}}{E}\frac{2E}{\omega}g^2z^2\left\{\left[u^2v^2-\tfrac{1}{4}(u^2-v^2)^2\right]+\tfrac{1}{2}(u^2-v^2)^2\frac{E^2}{\omega^2}\left(1+\frac{\omega^2}{E^2}\right)\right\},\\
&n_2^{(+)}=-(u^2-v^2)^2g^2z^2\frac{\mathrm{X}}{E}\frac{E^3}{\omega^3}\left(1-\frac{\omega^2}{E^2}\right).
\end{aligned}
\tag{A1}
$$

Furthermore, using numerical values for W_J, ξ_J, and ρ_J, we find

$$
\begin{aligned}
\alpha&=-(m_1+n_1)-2(m_2+n_2),\\
\alpha_0&=-3(m_1+n_1)-2(m_2+n_2),\\
\alpha_2&=-(4/7)(m_1+n_1)-(16/7)(m_2+n_2),\\
\alpha_4&=-(11/7)(m_1+n_1)-(58/49)(m_2+n_2).
\end{aligned}
\tag{A2}
$$

We see that, except for very large values of (m_2+n_2), the ordering of the triplet is mainly fixed by (m_1+n_1).

Let us consider first the case $(\omega/E)\ll 1$ (adiabatic limit). From (A1), we have:

$$
\begin{aligned}
m_1+n_1&\simeq 9(u^2-v^2)^2g^2z^2(\mathrm{X}/E)(E^3/\omega^3),\\
m_2+n_2&\simeq -\tfrac{3}{2}(u^2-v^2)^2g^2z^2(\mathrm{X}/E)(E^3/\omega^3).
\end{aligned}
\tag{A3}
$$

We see from (A2) that

$$
\alpha_0<\alpha_4<\alpha_0<\alpha_2, \tag{A4}
$$

which corresponds to the ordering

$$
\begin{aligned}
&E_0'<E_4'<E_2',\\
&(E_{0,4}'/E_2)<2\quad(E_2'/E_2)>2.
\end{aligned}
\tag{A5}
$$

This agrees with the results in Ref. 8. Experimental data in fact show that we are, in realistic cases, far from the adiabatic limit, and, in general, $(\omega/E)\simeq 0.5$. We distinguish two cases:

(a) For nearly closed and/or empty shells where $(u^2-v^2)^2>u^2v^2$, it can be seen from (A1) that (A5) is still valid;

(b) however, at the middle of the shell where $(u^2-v^2)\simeq 0$ and $u^2v^2\sim\frac{1}{4}$, we have

$$
\begin{aligned}
m_1+n_1&\simeq -(uvgz)^2\frac{\mathrm{X}}{E}\frac{4E}{\omega}\left(1-\frac{\omega^2}{E^2}\right)^{-1}\left(1+\frac{\omega^2}{E^2}\right),\\
m_2+n_2&\simeq -(uvgz)^2\frac{\mathrm{X}}{E}\frac{2E}{\omega}.
\end{aligned}
\tag{A6}
$$

From (A3) we see that

$$
\alpha_2<\alpha<\alpha_4<\alpha_0,
$$

which corresponds to

$$
\begin{aligned}
&E_2'<E_4'<E_0',\\
&(E_2'/E_2)<2\quad(E_{0,4}'/E_2)>2.
\end{aligned}
\tag{A7}
$$

This result has not been reached by Beliaev and Zelevinsky[8] who found that (E_2'/E_2) is always larger than 2. In realistic cases, however, the condition $(u^2-v^2)\ll uv$ is never adequately satisfied and this explains why the ordering $E_4'<E_0'$ has never been observed. Needless to say, between the two limits given by (A5) and (A7), we can have, for example,

$$
\begin{aligned}
&E_2'<E_0'<E_4',\\
&E_0'<E_2'<E_4'.
\end{aligned}
\tag{A8}
$$

An example of this has been found already in the text for the case of Ni^{62}.