Variational approach to anharmonic collective motion

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We derive large-amplitude collective equations of motion from the variational principle for the time-dependent Schrödinger equation. These equations reduce to the well-known diabatic formulas for vibrational frequencies in the small-amplitude limit. The finite-amplitude expression allows departures from the harmonic behavior of giant resonances to be simply estimated. The relative shift of the second phonon falls with nuclear mass $A$ as $A^{-2/3}$ in the three modes we consider: monopole, dipole, and quadrupole. Numerically, the effect is very small in heavy nuclei, as was found with other approaches.

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I. INTRODUCTION

There has been recent interest in the harmonicity of collective motion, with experimental data now available on double-phonon excitations of the giant dipole resonance [1–3]. Time-dependent mean-field theory provides a useful tool to study this topic, and a number of calculations have been reported [4–15]. Since the calculations in the full mean-field theory are rather opaque, it may be of some interest to find simple approximations that contain the same physics. The resulting equations of motion are quite intuitive when expressed in Hamiltonian form. With the nonlinear equation of motion we can calculate the anharmonicity in the giant dipole resonance, relevant to the measurements of [1,2].

A very successful general procedure to find approximations is to employ to the variational principle for the Schrödinger equation [16]

$$
\delta \int dt \langle \Psi | i \partial_t - H | \Psi \rangle = 0. \tag{1}
$$

This is of the form of the Lagrangian variational principle, with the integrand playing the role of the Lagrangian. The variational principle has been attributed to Dirac; it gives a natural way to derive the time-dependent Hartree-Fock approximation [17] and has been widely employed in that connection. It also produces useful equations of motion with more restricted assumptions about the wave function. Nörenberg and co-workers [18,19] put collective motion into the dynamics by adding variational fields corresponding to possible velocity potentials and the corresponding displacement fields. Our treatment is a special case of theirs in which only two coordinates are kept, the minimum number that can give Hamiltonian dynamics.

It is well known that collective motion can be generated by a local velocity field $Q(\vec{r})$ acting on a ground-state wave function [20–22]. To get any kind of Hamiltonian dynamics, an additional degree of freedom corresponding to displacements is required. There is a natural choice for the displacement field, which has been extensively investigated by Bohigas et al. [23]. These authors studied small-amplitude motion, using sum rules to simplify the discussion. We use the same fields and shall follow their notation, but we are not restricted to small amplitudes. The result will be formulas for anharmonic effects that involve only integrals over ground-state densities. The energy shift of the second phonon excitation $\Delta^{(2)}E$ is calculated from the semiclassical requantization of the Hamiltonian equations of motion. The semiclassical requantization recently has been favorably compared with the boson expansion approach in the context of a solvable model [24]. We find that this quantity is of order $\omega_0 / A^{4/3}$, where $\omega_0$ is the harmonic frequency of the phonon and $A$ is the atomic mass number. This strong $A$ dependence makes the shift very small in practice.

II. ANHARMONIC COLLECTIVE DYNAMICS

As mentioned above, the starting point is the variational principle, Eq. (1) above. If $|\Psi\rangle$ is varied in the space of Slater determinants, the result is time-dependent Hartree-Fock theory. Interesting simplified models are constructed by restricting $|\Psi\rangle$ further. In Refs. [25,26] $|\Psi\rangle$ is taken to be an antisymmetrized product of Gaussian single-particle wave functions. In this paper we shall assume that the motion is completely collective in the sense that it can be generated by a single-particle velocity field $Q(\vec{r})$.

$$
|\Psi(t=0,\vec{r})\rangle = e^{i\phi} |\Psi_0\rangle. \tag{2}
$$

Here $|\Psi_0\rangle$ is the ground-state wave function. The operator $Q$ of course acts on all the single-particle coordinates; we shall write operators of the form $\Sigma \hat{M}(\vec{r})$ as $\hat{M}(\vec{r})$. Integrating the time-dependent Schrödinger equation starting from the initial condition of Eq. (2) gives the series

$$
|\Psi(\vec{r})\rangle = (1 + iQ + t[H,Q] + \cdots)|\Psi_0\rangle. \tag{3}
$$

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Our ansatz will be implemented by taking the functional form for $|\Psi\rangle$ from a unitary generalization of Eq. (3). We multiply the fields $Q$ and $[H,Q]$ by time-dependent coefficients that define our dynamic variables. The operators are exponentiated to make the transformation of the wave function unitary. Thus we consider a trial wave function of the form

$$|\Psi_{\alpha\beta}\rangle = e^{i\lambda(t)Q} |\Psi_\beta\rangle = e^{i\lambda(t)Q} e^{B(t)\sigma_k Q} |\Psi_\beta\rangle.$$  

(4)

We have put in a factor of the nucleon mass $m_N$ for later convenience. This trial wave function is a special case of Eq. (2.7) of Ref. [19] with a single field $Q$. The two unitary transformations defined here were employed in Ref. [23] in treating small-amplitude collective motion. The commutator $[H,Q]$ occurs very frequently and we shall loosely follow the notation of Ref. [23] with the abbreviation

$$Q_1 = m_N[H,Q].$$  

(5)

When Eq. (4) is inserted into Eq. (1), the following Lagrangian is obtained for the coordinates $\alpha$ and $\beta$:

$$\langle \Psi_{\alpha\beta}|i\partial_t - H|\Psi_{\alpha\beta}\rangle = -\dot{\alpha} \langle \Psi_\beta|Q|\Psi_\beta\rangle - \langle \Psi_\beta|H|\Psi_\beta\rangle - \frac{\alpha^2}{2m_N} \langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle.$$  

(6)

The derivation is in Appendix A, along with the equations of motion that follow from the Lagrange equations. We will find the phonon frequencies by requantizing the equations of motion, but this requires a Hamiltonian formulation. Thus we seek a transformation of variables $(\alpha,\beta) \rightarrow (x,p)$ together with a Hamiltonian $\mathcal{H}(x,p)$ such that the equations of motion can be expressed in the form

$$\dot{p} = -\partial_x \mathcal{H},$$  

$$\dot{x} = \partial_p \mathcal{H}.$$  

(7)

Reference [17] describes a procedure for obtaining a canonical pair of variables when there are two degrees of freedom. However, the choice of variables in [17] is inconvenient in that it does not produce a quadratic Hamiltonian in the momentum. From Eq. (6) it is easy to see that the choice $x = \langle \Psi_\beta|Q|\Psi_\beta\rangle$, $p = \alpha$ gives a canonical pair $(x,p)$ with a simple kinetic-energy term in the Hamiltonian. However, the Hamiltonian is difficult to express explicitly in terms of $x$. Therefore, we use instead the canonical pair $(\beta,p_\beta)$ with

$$p_\beta = \alpha \langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle,$$

which allows us to keep $\beta$ as the coordinate variable.\(^1\) The Hamiltonian in this representation may be written as

$$\mathcal{H}(\beta,p_\beta) = \frac{p_\beta^2}{2m(\beta)} + U(\beta).$$  

(8)

with

$$m(\beta) = m_N \langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle,$$

$$U(\beta) = \langle \Psi_\beta|H|\Psi_\beta\rangle.$$  

(9)

The reader may verify that the Hamiltonian equations (7) defined this way are equivalent to Eqs. (A11) and (A12).

To make use of the equations of motion, we need to evaluate $\beta$-dependent matrix elements appearing in Eq. (9). This can be done in two different ways, depending on whether we apply the operator $e^{BQ_1}$ to the left or to the right. The first way is to apply explicitly the operator on the wave function. We shall only consider nonrelativistic Hamiltonians with local potentials, so the operator $Q_1 = m_N[H,Q]$ is a first-order derivative:

$$Q_1 = -\frac{\nabla^2 Q}{2} - \nabla Q \cdot \nabla.$$  

(10)

The exponentiated operator $e^{BQ_1}$ acting on a function of a single coordinate variable, such as $x$, may be expressed in closed form as

$$e^{BQ_1}f(x,y,z) = \sqrt{\frac{\partial_x Q(x)}{\partial_x Q(x)}} f(x',y,z).$$  

(11)

The displaced coordinate $x'$ is obtained by integrating from $x$ a distance $x-x'$ that satisfies

$$\beta = \int_{x'}^x ds \partial_s Q(s).$$  

(12)

The derivation of this formula may be found in Ref. [27].

The other way to calculate matrix elements is to apply the unitary transformation to the operator being evaluated. Thus we use the identity

$$\langle \Psi_\beta|M|\Psi_\beta\rangle = \langle \Psi_\beta|(e^{-BQ_1}Me^{BQ_1})|\Psi_\beta\rangle.$$  

(13)

This will turn out to be very convenient for matrix elements of scaling displacements.

The special case where $Q$ is a quadratic function of the coordinates gives a particularly simple form for the transformed coordinates [23], namely, they are scaled by a factor. For example, for the field $Q = x^2/2$, the transformation is

$$e^{BQ_1} = e^{-BQ_1} = e^{-\beta x,y,z}$$  

(14)

and the wave function $\Psi_\beta$ is given by

$$\Psi_\beta(x,y,z) = e^{-\beta/2} \Psi_0(e^{-\beta}x,y,z).$$  

(15)

III. A MODEL HAMILTONIAN

We wish to apply the equations of motion to a variety of giant resonances and will need a detailed model for $H$ in order to construct $U(\beta) = \langle \Psi_\beta|H|\Psi_\beta\rangle$. A good balance between simplicity and realism is provided by the Skyrme-like form for the Hamiltonian density,

$$h = \rho_0[\tau + v_\sigma n^2 + v_tr^{3/2}],$$  

(16)

where $\tau$ is the kinetic-energy density and $n$ the density, both in units of nuclear matter density $\rho_0$. The coefficients $v_\sigma$ and
$v_b$ are determined to reproduce nuclear saturation density $\rho_0=0.16$ fm$^{-3}$ (with the corresponding Fermi energy $e_F\approx 36$ MeV) and the nuclear matter binding energy $B\approx 16$ MeV per nucleon. If we express $n$ in units of the saturation density the parameters may be expressed as

$$v_a = -4B - \frac{6}{5} e_F \approx -107 \text{ MeV},$$

$$v_b = 3B + \frac{3}{5} e_F \approx 70 \text{ MeV}.$$

The power dependence of the third term in Eq. (16) is not obvious. From many-body theory, one expects the energy of a dilute Fermi gas to be a series in powers of $k_F$ or $n^{1/3}$. Equation (16) thus represents the first three terms of that series. The parametrization $n^{2/3}$ for the third term also predicts a compressibility not far from that required by the empirical single-particle Hamiltonians. It should be mentioned that Eq. (16) lacks momentum-dependent interactions, which are certainly present in the empirical single-particle Hamiltonian.

For treating the giant dipole resonance, we also need to know the isospin dependence of $H$. The kinetic energy has an obvious isospin dependence arising from the separate Fermi energies of neutrons and protons. We shall add isospin-dependent potential-energy terms with the same density dependence as in Eq. (16) and require the isospin dependence of the semiempirical mass formula to be reproduced. The binding energy per particle $\epsilon(n,n_\pi)=\hbar n/\rho_0$ in the Fermi gas approximation is expressed as follows, with proton and neutron densities written $n_p=n/2+n_\pi$ and $n_n=n/2-n_\pi$:

$$\epsilon(n,n_\pi) = \frac{3}{5} e_F\left[ \frac{n^{2/3}}{2} \left( \frac{1+2n_\pi}{n} \right)^{5/3} + \left( 1 - \frac{2n_\pi}{n} \right)^{5/3} \right] + v_an + v_bn^{3/2} + v_{nn}n^2/n. \tag{18}$$

Expanding this in powers of $n_\pi$, we have

$$\epsilon(n,n_\pi) \approx \epsilon(n,0) + \frac{4}{3} e_F n^{2/3} + v_{nn} n^{1/2}. \tag{19}$$

The semiempirical mass formula has isospin dependent terms,

$$B(A,Z) = B(A,A/2) + b_{\text{sym}} \left( \frac{N-Z}{A} \right)^2 + b_c Z^2 \frac{A}{A^{1/3}} + \cdots, \tag{20}$$

with $b_{\text{sym}}=25$ MeV. Assuming that neutrons and protons occupy the same volume, $n_\pi/n=(Z-N)/2A$ and we may relate the coefficient in Eq. (18) to $b_{\text{sym}}$ as

$$\frac{4e_F}{3} + v_\pi = 4b_{\text{sym}}. \tag{21}$$

Putting in the Fermi energy $e_F\approx 36$ MeV, we find numerically

$$v_\pi \approx 50 \text{ MeV}. \tag{22}$$

**IV. FINITE-AMPLITUDE EXCITATIONS**

We now treat the excitation of the giant monopole, dipole, and quadrupole modes of vibration. For each multipole, we will define a collective field $Q$ and then evaluate the harmonic frequency and the nonlinear corrections. The functions that are required for this are the expectation of the Hamiltonian in the $\beta$-deformed state, which we expand as

$$U(\beta) = \frac{k}{2} \beta^2 + \frac{k_3}{3} \beta^3 + \frac{k_4}{4} \beta^4 + \cdots. \tag{23}$$

We have defined the energy scale so that $U(0)=0$. The linear term in the expansion vanishes because of the stability of the ground state. We also need to expand the inertia to second order in $\beta$. We write this as

$$m(\beta) = m_0(\Psi_\beta|Q, Q_1|\Psi_\beta) = m(1 + m_1\beta + m_2\beta^2 + \cdots). \tag{24}$$

The key formulas are the equation for the frequency in the harmonic limit

$$\omega_0 = \sqrt{\frac{k}{m}}$$

and the formula for the energy shift of the second phonon. It is convenient to express this in terms of an energy parameter $E_{\text{anh}}$ as

$$\Delta(1)^2 E = E_2 - 2E_1 + E_0 = \frac{2\omega_0^2}{E_{\text{anh}}} \tag{25}$$

The derivation of the expression for $E_{\text{anh}}$ in terms of the nonlinear coefficients $k_3, k_4, m_1$ and $m_2$ is given in Appendix B. The result is

$$E_{\text{anh}} = \frac{5}{12} k_3 - \frac{3}{8} k_4 - \frac{3}{4} k_4 m_1 + \frac{m_1^2}{16k} + \frac{m_2}{4k}. \tag{26}$$

All the $k$’s in this equation scale with mass number as $k_j\sim A$, while the $m_j$’s are independent of $A$ in the droplet limit. We thus see that the anharmonicity energy scale $E_{\text{anh}}$ varies with mass number as

$$E_{\text{anh}} \sim A.$$

We now consider the various multipoles in turn, starting with the isoscalar monopole and quadrupole modes.

**A. Monopole**

The monopole field $Q$ for a uniform sphere with a sharp edge would be proportional to the $J_0$ spherical Bessel function, but in practice the nuclear surface cannot be ignored even for large nuclei. The compressibility is less in the surface and this has the consequence that the velocity potential is more like the simple scaling form [28]

$$Q = r^2/2. \tag{27}$$

We shall construct the nonlinear dynamics with this field. It is most convenient to apply the transformation to the Hamiltonian in this case. The inertia is
TABLE I. Coefficients of the collective inertia expansion (24).

<table>
<thead>
<tr>
<th>Mode</th>
<th>$m/A$</th>
<th>$m_1$</th>
<th>$m_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>monopole</td>
<td>$m_N(r^2)$</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>quadrupole</td>
<td>$2m_N(r^2)$</td>
<td>2</td>
<td>2</td>
</tr>
<tr>
<td>dipole</td>
<td>$32m_N/63$</td>
<td>0</td>
<td>$-0.58/R^2$</td>
</tr>
</tbody>
</table>

$$m(\beta) = m_N(\Psi_\beta[H(\Psi_\beta)]^{1/2}) = e^{2\beta}m_N(r^2)$$

$$= e^{2\beta}m_N A(r^2), \quad (28)$$

where $\langle r^2 \rangle$ denotes the mean-square radius of the ground state. The expectation value of the Hamiltonian is

$$\langle \Psi_\beta | H | \Psi_\beta \rangle = \rho_0 \left[ e^{-2\beta} \int d^3r \tau_0(\vec{r}) + e^{-3\beta}v_a \int d^3r n_0^2(\vec{r}) + e^{-4\beta}v_b \int d^3r n_0^7(\vec{r}) \right]. \quad (29)$$

where $\tau_0(\vec{r})$ denotes the kinetic-energy density of the ground state $|\Psi_0\rangle$ and $n_0(\vec{r})$ the particle density. This formula is derived using the relations (13) and (15), which yield for the monopole field

$$e^{-\beta Q_1} \tau_0(\vec{r}) e^{\beta Q_1} = e^{-3\beta} \tau_0(e^{-\beta} \vec{r})$$

and

$$e^{-\beta Q_1} n_0(\vec{r}) e^{\beta Q_1} = e^{-3\beta} n_0(e^{-\beta} \vec{r}).$$

We next expand Eq. (29) in the power series in $\beta$. The linear term vanishes because of the saturation condition, (17). The quadratic term, giving the effective restoring force, is

$$k = \sigma\beta^2 \langle \Psi_\beta | H | \Psi_\beta \rangle |_{\beta = 0} = \rho_0 \left[ 4 \int d^3r \tau_0(\vec{r}) + 9v_a \int d^3r n_0^2(\vec{r}) + 16v_b \int d^3r n_0^7(\vec{r}) \right]. \quad (30)$$

For a spherical drop with radius $R$, this is equal to $A$ times the nuclear matter compressibility $K$ if one can make the large-$A$ approximation $n_0(\vec{r}) = \theta(R-\vec{r})$, which yields $\rho_0 \int d^3r n_0^2(\vec{r}) = \rho_0 \int d^3r n_0^7(\vec{r}) = A$ and $\langle \Psi_0 | -\nabla^2/2m_N | \Psi_0 \rangle = \rho_0 \int d^3r \tau_0(\vec{r}) = 3e_F A/5$. Surface effects of course spoil this approximation, and the fact of the matter is that they have an exaggerated importance because the $v_a$ and $v_b$ are both large, but with opposite sign. However, for our purposes it is an unnecessary refinement to improve on the nuclear matter approximation. The harmonic approximation for the frequency is then the well-known collective formula

$$\omega^2 = \frac{K}{m_N(r^2)}. \quad (31)$$

The nonlinear coefficients in the expansion of $U$ and $m(\beta)$ are given numerically in Table I. Combining these according to Eq. (26), we obtain for the anharmonicity parameter

$$E_{\text{anh}} = 40 A \text{ MeV}.$$
C. Giant dipole resonance

The field $Q$ for the giant dipole resonance is not as simple as the other cases. In light nuclei, the energetics of the giant dipole state suggests that the $Q$ is close to being the simple operator $\tau_z$, but this form gives the wrong $A$ dependence to describe the dipole energies in heavy nuclei. The Steinwedel-Jensen model takes an opposite extreme, positing that the displacement field vanishes at the nuclear surface. This can be generated by a velocity field such as

$$Q = \tau_z (1 - r^2/3R^2),$$

where $R$ is the nuclear radius. We will adopt this form to investigate the nonlinearity, although the model predicts too high a frequency for the dipole. The displacement field then has the form

$$Q_1 = \frac{x^2 + y^2 + 3z^2 - 3R^2}{3R^2} \partial_z + \frac{5z}{3R^2} + \frac{2z}{3R^2} (x \partial_x + y \partial_y).$$

The coordinate $\beta$ associated with this $Q_1$ has the dimensions of length. Unlike in the monopole and quadrupole cases, we were unable to find an analytic form for the needed expectation values. This is due to the mixing of the Cartesian coordinates in $Q_1$. Instead, we use the expansion (A4) explicitly to evaluate the inertia and the kinetic-energy term of the Hamiltonian. For the inertia, we must expand to second order as

$$\langle \Psi_\beta | [Q_1, Q_1] | \Psi_\beta \rangle = \langle \Psi_\beta | [Q_1, Q_1] | \Psi_\beta \rangle - \beta \langle \Psi_\beta | [Q_1, [Q_1, Q_1]] | \Psi_\beta \rangle + \frac{\beta^2}{2} \langle \Psi_\beta | [Q_1, [Q_1, [Q_1, Q_1]]] | \Psi_\beta \rangle.$$

We shall evaluate this by approximating the density as that of a uniform drop. The result is

$$m = A m_N (32 \frac{32}{63} - 2944 \frac{\beta^2}{R^2} + \frac{2944 \beta^2}{5103} - \cdots).$$

Notice that $\beta$ appears in the nonlinear terms in the dimensionless ratio $\beta/R$.

The kinetic-energy operator is treated by applying Eq. (A4) to each of the two gradients that it contains. The algebra is very tedious and we have used a computer program for the manipulations. We quote here as an example the $x$ gradients expanded to second order in $\beta$,

$$\partial_x = \partial_x + \frac{2 \beta}{3R^2} (z \partial_z + x \partial_x) + \frac{\beta^2}{9R^2} [5x + (3R^2 + x^2 - y^2 - z^2) \partial_x + 2xy \partial_y + 6xz \partial_z].$$

We actually need the gradient evaluated to fourth order, but it does not seem useful to display the full expression. We evaluate the kinetic energy in the Fermi gas approximation by applying the transformed gradient to a plane-wave state, taking the modulus squared, and integrating over a spherical Fermi surface. For each term in $\beta$, we keep only the highest power of $R$. This is

$$\langle \Psi_\beta \mid - \frac{\nabla^2}{2m} \mid \Psi_\beta \rangle = \frac{3}{5} e_F A + \frac{44}{45} e_F A \frac{\beta^2}{R^2} + \frac{168}{212} \frac{853}{625} e_F A \frac{\beta^4}{R^4} + \cdots.$$

It still remains to evaluate the potential energy. We found this easier to do by applying the transformation to the wave function and explicitly constructing the transformed single-particle density to the needed order in $\beta$. We will again approximate the ground state as a uniform sphere, which means that gradients of the wave function are ignored in evaluating the effect of the operator. Of course, the wave-function gradients cannot be ignored in the surface region, but we have constructed the operator $Q$ to have no surface contributions. We only need the wave function to third order in $\beta$ to evaluate the potential energy to fourth order. The wave function is

$$\langle \Psi_\beta \rangle = (1 + \beta Q_1 + \frac{\beta^2}{2} Q_1 + \frac{\beta^3}{6} Q_1 + \cdots) \langle \Psi_0 \rangle.$$

Calculating the density from this, we see that both the isovector and isoscalar densities are affected by the transformation. The densities are given to order $\beta^2$ as

$$n_{\tau\beta} (\tilde{r}) = - \frac{5\beta z}{3R^2} + \cdots.$$

Inserting the densities in the potential-energy function, we find the expression for the potential energy to order $\beta^4$,

$$\langle \Psi_\beta | v | \Psi_\beta \rangle = \langle \Psi_0 | v | \Psi_0 \rangle + \frac{5}{9} v \tau A \frac{\beta^2}{R^2} - \frac{260}{1701} v \tau A \frac{\beta^4}{R^4} + \cdots.$$

Notice that the coefficients of $\beta^n$ are all of the form of a constant times $A/R^n$. This implies that $E_{\text{anh}}$ will depend on nuclear size as $A$. There is a cancellation between the kinetic and potential contributions to $k_4$, so the resulting anharmonicity is very small,

$$|E_{\text{anh}}| > 100 A \text{ MeV},$$

giving once more a negligible energy shift for the double excitation.

V. CONCLUSION

Our conclusion, that anharmonic effects are extremely small in giant vibrations, is in agreement with earlier studies. However, the precise $A$ dependence had not been made clear. For example, in Ref. [8], the authors stated that $\Delta^{(2)} E$ should scale with $A$ as $\Delta^{(2)} E \sim A^{-2/3}$. See also [29]. This implies that $E_{\text{anh}}$ would be independent of $A$, disagreeing with our linear $A$ dependence.

One aspect of the original experimental findings on the double dipole phonon caused considerable discussion. This was the apparent increased strength of the second phonon as compared to the harmonic limit. In our treatment, the energy-
weighted sum rule is respected for all initial states and so it is not possible to alter the strength of the second phonon if its energy does not change from the harmonic limit. It is likely that the apparent increase in strength from these experiments was due to the presence of other multipoles that contribute incoherently to the cross sections.

Our model could be improved in a number of ways. The actual field for the dipole has a considerable amount of surface displacement, even for heavy nuclei, and a more realistic field could be employed. The Hamiltonian should include momentum-dependent interactions to be more realistic. However, there is no reason to think that these improvements would change the picture in a qualitative way and it hardly seems worthwhile to calculate the very small effect more accurately.

More interesting and challenging is the development of a nonlinear collective description of the low collective modes, in particular the octupole vibration. Part of the doubly excited octupole has been identified recently in 208Pb [30], but the only theory up to now is the rather opaque second random-phase approximation. To use our treatment, one would have to find a nearly local operator that would generate the low octupole, i.e., a field for which the associated sum rule would be nearly exhausted by the low mode.

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APPENDIX A: DERIVATION OF EQUATION OF MOTION

We shall evaluate the variational principle, (1) using, as a trial wave function (4),

\[ |\Psi_{\alpha\beta}\rangle = \exp(i\alpha Q)\exp(\beta m_N[H,Q])|\Psi_0\rangle = \exp(i\alpha Q)|\Psi_\beta\rangle. \]

The variational principle will be used to determine the coefficients \( \alpha \) and \( \beta \); \( Q \) is some local function of position and \( |\Psi_0\rangle \) is the ground state. The variation with respect to \( \alpha \) and \( \beta \) in Eq. (1) yields the usual Lagrangian equations of motion

\[ \frac{d}{dt} \frac{\partial L}{\partial \dot{\alpha}} - \frac{\partial L}{\partial \alpha} = 0, \quad (A1) \]

\[ \frac{d}{dt} \frac{\partial L}{\partial \dot{\beta}} - \frac{\partial L}{\partial \beta} = 0, \quad (A2) \]

where \( L \) is the integrand in Eq. (1),

\[ L = \langle \Psi_{\alpha\beta}|i\partial_t|\Psi_{\alpha\beta}\rangle - \langle \Psi_{\alpha\beta}|H|\Psi_{\alpha\beta}\rangle. \quad (A3) \]

From this point it is simply algebra to reduce Eqs. (A1)–(A3) to a more transparent form. We shall use an identity for expanding the operator product \( e^{-\Delta B}e^{\Delta A} \) in commutators,

\[ e^{-\Delta B}e^{\Delta A} = B + [B,A] + \frac{1}{2}[[B,A],A] \]

\[ + \frac{1}{3!}[[[B,A],A],A] + \cdots. \quad (A4) \]

We also need two corollary identities

\[ \partial_\alpha(e^{-\alpha\Delta B}e^{\alpha A}) = e^{-\alpha\Delta}[B,A]e^{\alpha A} \quad (A5) \]

and

\[ e^{-\Delta A}e^{\Delta A} = A. \quad (A6) \]

We begin by examining the second term in Eq. (A3), \( \langle \Psi_{\alpha\beta}|H|\Psi_{\alpha\beta}\rangle \). The \( \alpha \) dependence is made more explicit by expanding \( e^{-i\alpha Q}H e^{i\alpha Q} \) in commutators. If \( Q \) is local (and \( H \) is quadratic in the momenta), the third- and higher-order commutators vanish. From Eqs. (A4) and (5) the expansion is

\[ \langle \Psi_{\alpha\beta}|H|\Psi_{\alpha\beta}\rangle = \langle \Psi_{\beta}|H|\Psi_\beta\rangle + i\frac{\alpha}{m_N}\langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle \]

\[ + \frac{\alpha^2}{2m_N}\langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle. \quad (A7) \]

Next we argue that the middle term in this sum vanishes. First observe that by Eq. (A6) \( \langle \Psi_{\beta}|Q_1|\Psi_\beta\rangle = \langle \Psi_0|Q_1|\Psi_0\rangle \) does not depend on \( \beta \). The fact that \( |\Psi_0\rangle \) is a stationary state of the Hamiltonian implies

\[ 0 = \partial_\alpha\langle \Psi_{\alpha,\beta=0}|H|\Psi_{\alpha,\beta=0}\rangle|_{\alpha=0} = \frac{i}{m_N}\langle \Psi_0|Q_1|\Psi_0\rangle \]

\[ = \frac{i}{m_N}\langle \Psi_\beta|Q_1|\Psi_\beta\rangle. \quad (A8) \]

The time derivative in Eq. (A3) contains two terms

\[ \langle \Psi_{\alpha\beta}|i\partial_t|\Psi_{\alpha\beta}\rangle = -\partial_\alpha\langle \Psi_{\beta}|Q|\Psi_\beta\rangle + i\beta\langle \Psi_\beta|Q_1|\Psi_\beta\rangle. \]

The second term vanishes by Eq. (A8). The complete expression for the Lagrangian then becomes

\[ \mathcal{L} = -\partial_\alpha\langle \Psi_{\beta}|Q|\Psi_\beta\rangle - \langle \Psi_\beta|H|\Psi_\beta\rangle \]

\[ - \frac{\alpha^2}{2m_N}\langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle. \quad (A9) \]

The dependence on \( \alpha \) is now entirely explicit. Inserting \( \mathcal{L} \) into Eq. (A1) yields

\[ -\partial_\alpha\langle \Psi_\beta|Q|\Psi_\beta\rangle + \frac{\alpha}{m_N}\langle \Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle = 0. \quad (A10) \]

This is simplified with the help of Eq. (A5) to

\[ \beta = \frac{\alpha}{m_N}. \quad (A11) \]

We next carry out the derivatives in the second Lagrangian equation, (A2) to obtain
\[
\hat{\alpha}\beta\langle\Psi_\beta|Q|\Psi_\beta\rangle + \partial_\beta\langle\Psi_\beta|H|\Psi_\beta\rangle \\
+ \frac{\alpha^2}{2m_N}\partial_\beta\langle\Psi_\beta|[Q,Q_1]|\Psi_\beta\rangle = 0.
\]  
(A12)

This is as far as we can simplify it without approximation beyond the basic ansatz (4). The harmonic limit is obtained by using the expansion (A4) and by keeping in Eq. (A12) only linear terms in \(\alpha\) and \(\beta\). By the stationarity of the ground state \(\partial_\beta\langle\Psi_\beta|H|\Psi_\beta\rangle = 0\) and the equation of motion reduces to

\[
\hat{\alpha}\langle\Psi_0|[Q,Q_1]\rangle|\Psi_0\rangle + \beta\langle\Psi_0|[H,Q_1]|\Psi_0\rangle = 0.
\]  
(A13)

The frequency of oscillation [23] is then given by inserting Eq. (A11) into Eq. (A13) as

\[
\omega^2 = \frac{\langle\Psi_0|[H,Q_1]|\Psi_0\rangle}{m_N\langle\Psi_0|[Q,Q_1]|\Psi_0\rangle} = \frac{M_1}{M_1},
\]  
(A14)

where \(M_n\) is the \(n\)th energy moment of the transition strength,

\[
M_n = \sum_i \langle 0|Q_i|^2|E_i - E_0\rangle^n.
\]

**APPENDIX B: ANHARMONICITY**

In this appendix we derive the frequency shift of multiple phonon excitations due to the anharmonicity of the equation of motion (A12). Our derivation proceeds through the Bohr-Sommerfeld quantization of the classical orbits. This requires the phase integral

\[
\phi = \int p\, dx
\]

using a Hamiltonian representation of the equations of motion. The condition that an eigenstate be at energy \(E\) is that classical phase accumulated over the orbit be an integral multiple of \(2\pi\). Taking the form (8) for the Hamiltonian, the energy \(E_n\) of the \(n\)th state satisfies

\[
\phi(E_n) = \int_{\beta_1}^{\beta_2} \sqrt{2m(\beta)[E_n - U(\beta)]} d\beta = n\pi,
\]

\(n = 0,1,2,\ldots\). 

(B1)

where \(\beta_{1,2}\) are the classical turning points given by \(U(\beta_{1,2}) = E_n\).2

We now assume that the anharmonicity is weak, so that all quantities can be expanded in power series in \(\beta\), which we wrote as Eqs. (23) and (24). To evaluate the integral Eq. (B1) to a given order in \(\beta\), we change variable to make the energy difference under the square root a simple quadratic function. Defining a variable \(z = \sqrt{U(\beta)/E}\), we write the integral as

\[
\phi(E) = \sqrt{2E} \int_{-1}^{1} \frac{1}{\sqrt{1 - z^2}} \frac{\sqrt{m(\beta)}}{dz} dz.
\]  
(B2)

The second square root and derivative are expanded in powers of \(\beta\). The latter is obtained via the series for \(\beta(z)\),

\[
\beta = \sqrt{\frac{2E}{k}} \left[ z - \frac{2^{1/2}k_3}{3k^{3/2}} E^{1/2}z^2 + \left( \frac{5 k_3^2}{9 k^3} - \frac{k_4}{2k^2} \right) E^{3}z + \cdots \right],
\]  
(B3)

with the derivative

\[
d\beta = \sqrt{\frac{2E}{k}} \left[ 1 - 2^{3/2} \frac{k_3}{3k^{3/2}} E^{1/2}z \\
+ \left( \frac{5 k_3^2}{3 k^3} - \frac{3k_4}{2k^2} \right) E^{3}z + \cdots \right] dz.
\]

The integrals are then elementary to evaluate, giving, for \(\phi\),

\[
\phi = \pi \frac{E}{\omega_0} + \pi \left( \frac{5 k_3}{12 k^3} - \frac{3k_4}{8 k^2} - \frac{k_1 m_4}{4k^2} - \frac{m_1^2}{16k} + \frac{m_2^2}{4k} \right) \frac{E}{\omega_0} + \cdots
\]

(B4)

where \(\omega_0 = \sqrt{k/m}\) is the small-amplitude harmonic frequency. The anharmonicity in this limit is controlled by the combination of nonlinear coefficients in parentheses. It has the dimensions of inverse energy and we shall abbreviate it as

\[
E_{\text{anh}}^{-1} = \frac{5 k_3}{12 k^3} - \frac{3k_4}{8 k^2} - \frac{k_1 m_4}{4k^2} - \frac{m_1^2}{16k} + \frac{m_2^2}{4k}.
\]  
(B5)

To find the energy shifts, we insert Eq. (B4) into Eq. (B1) and invert the resulting power series that expresses \(n\) in terms of \(E\). The result is

\[
E_n = n\omega_0 - n^2 \frac{\omega_0^2}{E_{\text{anh}}} + \cdots.
\]  
(B6)

The shift of the double phonon with respect to the single-phonon excitation is of direct experimental interest. To leading order, this is evaluated from Eq. (B6) as

\[
\Delta^{(2)}E = E_2 - 2E_1 + E_0 = -2 \frac{\omega_0^2}{E_{\text{anh}}}.
\]  
(B7)

Equation (B6) is used in Sec. IV.

Another way to derive the frequency shift would be to find the classical frequencies at energies \(\omega_0\) and \(2\omega_0\). Those energies correspond to wave packets made of states \(n = 0,1\) and \(n = 1,2\), respectively. Thus the classical frequencies correspond to the energy differences \(E_1 - E_0\) and \(E_2 - E_1\), respectively, and the shift would be calculated as

\[
\Delta^{(2)}E = \omega(\omega_1) - \omega(\omega_0).
\]  
(B8)

Classical orbital perturbation theory may be used to express these frequencies in terms of the anharmonicities \(m_1\), \(m_2\), \(k_3\), and \(k_4\). We have verified that the two methods give the same result for the \(k_4\) dependence.