

Rotation of a self-bound many-body system

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We discuss the rotational excitations of highly quantum many-body systems (for example, polyatomic molecules or microdroplets of helium). For a general system, the states $F\Phi$, where $F = \frac{1}{2} \sum_i \sum_j (x_{ij} + iy_{ij})^{L_f} f(r_{ij})$ and Φ is a rotationally invariant ground or vibrational state, are shown to be eigenfunctions of \mathbf{L}^2 and L_z , with eigenvalues $L(L+1)\hbar^2$ and $L\hbar$ (for even L). These wavefunctions preserve the translational invariance and the permutation and inversion symmetries of the N -particle state Φ . For harmonic pair interactions, the $f=1$ wavefunctions are shown to be exact eigenstates of the N -body hamiltonian. For large N , the states $F\Phi$ ($f=1$) represent surface oscillations of the type first proposed by Bohr. An inequality for the rotational excitation energy is obtained variationally; it depends on two, three, and four-particle correlations. Other translationally invariant angular momentum eigenfunctions are also discussed.

1. INTRODUCTION

The rotational states of polyatomic molecules are normally discussed in terms of rigid-body rotation; refinements to allow for non-rigidity are then built on to the rigid-rotation theoretical framework [1, 2]. Such a treatment is suitable for most molecules, but not for the loosely bound, highly quantum molecules of the isotopes of helium. This can be seen clearly by letting the number of atoms in the system get large: in the case of helium we end up with a droplet of a quantum fluid, which certainly cannot be regarded as rigid in any approximation. In this paper we obtain certain exact many-body orbital angular momentum eigenfunctions by examining the three-body case. These rotational states are then shown to be the quantum analogues of the surface oscillations of a liquid drop, and thus correspond precisely to what one would expect for the low-lying excitations of a helium microdroplet.

We shall discuss the rotational excitations of a non-relativistic system of N particles (bosons or fermions), with hamiltonian

$$H = -\frac{\hbar^2}{2m} \sum_{i=1}^N \nabla_i^2 + V(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N). \quad (1)$$

The potential energy V is assumed to be symmetric with respect to permutations of the particle coordinates \mathbf{r}_i , and also to be translationally and rotationally invariant. We further assume (in this paper) that V is independent of spin coordinates, so that orbital angular momentum is a good quantum number. Throughout this paper we ignore the spin coordinates, and angular momentum

is understood to mean *orbital* angular momentum. The total angular momentum operator is

$$\mathbf{L} = -i\hbar \sum_{i=1}^N \mathbf{r}_i \times \nabla_i. \quad (2)$$

On making the transformation $\mathbf{r}_i \rightarrow \mathbf{r}_i + \mathbf{a}$, we see that, as in classical mechanics, the angular momentum of the system is independent of the choice of origin if and only if we work in the rest frame of the system. We therefore restrict our considerations to states Ψ of zero total momentum \mathbf{P} :

$$\mathbf{P}\Psi = \left(-i\hbar \sum_{i=1}^N \nabla_i \right) \Psi = 0. \quad (3)$$

In classical mechanics the total momentum can be set equal to zero, and at the same time the centre of mass coordinate

$$\mathbf{R} = N^{-1} \sum_{i=1}^N \mathbf{r}_i \quad (4)$$

can be fixed at the origin. In quantum mechanics this is not possible: \mathbf{R} and \mathbf{P} are conjugate variables, with

$$[R_x, P_x] = i\hbar, \text{ etc.}, \quad (5)$$

so that when the total momentum is zero the centre of mass coordinate is completely indeterminate by the uncertainty principle. The true rotational excitations of an isolated nucleus or molecule are therefore translationally invariant states.

It is convenient to at first ignore this fact in constructing many-particle wavefunctions from single-particle wavefunctions (as is done in the shell model) and then to construct translationally invariant states by the substitution $\mathbf{r}_i \rightarrow \boldsymbol{\rho}_i$, where

$$\boldsymbol{\rho}_i = \mathbf{r}_i - \mathbf{R}. \quad (6)$$

The replacement of the \mathbf{r}_i coordinates by the coordinates relative to the centre of mass gives exact eigenstates in the special case of harmonically interacting fermions forming closed shells, as was shown by Bethe and Rose [3]. However, in the general case there are severe difficulties, arising from the fact that the $\boldsymbol{\rho}_i$ are not independent coordinates:

$$\sum_{i=1}^N \boldsymbol{\rho}_i = 0. \quad (7)$$

If one treats the $\boldsymbol{\rho}_i$ as independent coordinates, one has $3N$ variables instead of the $3N-3$ variables needed to specify an N -particle state in its rest frame. This leads to 'spurious states' which do not correspond to any state of internal motion, as pointed out by Elliott and Skyrme [4]. The problem of eliminating the spurious states has been discussed by several authors [5-7]: it has been shown to have a solution only in the special case of harmonic interactions.

A different approach [8–10] is to transform to normal coordinates ξ_λ , defined by

$$\xi_\lambda = \left(\frac{\lambda}{\lambda+1} \right)^{1/2} \left[\mathbf{r}_{\lambda+1} - \lambda^{-1} \sum_{i=1}^{\lambda} \mathbf{r}_i \right] \quad (\lambda=1, 2, \dots, N-1), \quad (8)$$

$$\xi_N = N^{-1/2} \sum_{i=1}^N \mathbf{r}_i. \quad (9)$$

This transformation achieves the desired result of separating the centre of mass motion from the internal motion: the ξ_λ for $\lambda < N$ are relative coordinates, independent of the choice of origin, ξ_N is proportional to \mathbf{R} , and

$$\sum_{i=1}^N \nabla_i^2 = \sum_{\lambda=1}^N \nabla_\lambda^2, \quad (10)$$

$$\mathbf{L} = -i\hbar \sum_{\lambda=1}^N \xi_\lambda \times \nabla_\lambda. \quad (11)$$

The N -particle problem has been transformed to that of N quasiparticles, one of which represents the motion of the whole body, and the other relative motion within the body. Further, since

$$\frac{1}{2} \sum_{j=1}^N \sum_{i=1}^N \mathbf{r}_{ij}^2 = N \sum_{\lambda=1}^{N-1} \xi_\lambda^2, \quad (12)$$

the quasiparticles are non-interacting when the particles interact with harmonic pair forces. The quasiparticles are however no longer bosons or fermions, but have a very complicated permutation symmetry.

In this paper we use the method of normal coordinates to obtain a set of angular momentum eigenstates for the three-body case, and then generalize to the N -body system. The N -body angular momentum eigenstates have the necessary translational invariance, and the correct permutation symmetry. The difficulties mentioned in the previous two paragraphs are thus entirely avoided. On the other hand, the approach is intuitive rather than systematic, and we cannot hope to obtain all the rotational states in this way.

2. MANY-BODY EIGENSTATES OF \mathbf{L}^2 AND L_z

We consider the three-body problem first, since in this case all the rotational states may formally be written down. The normal coordinates are

$$\left. \begin{aligned} \xi_1 &= \frac{1}{\sqrt{2}} \mathbf{r}_{21}, \\ \xi_2 &= \frac{1}{\sqrt{6}} (\mathbf{r}_{31} + \mathbf{r}_{32}), \\ \xi_3 &= \frac{1}{\sqrt{3}} (\mathbf{r}_1 + \mathbf{r}_2 + \mathbf{r}_3). \end{aligned} \right\} \quad (13)$$

The angular momentum operator in the rest frame is

$$\mathbf{L} = -i\hbar \left(\boldsymbol{\xi}_1 \times \frac{\partial}{\partial \boldsymbol{\xi}_1} + \boldsymbol{\xi}_2 \times \frac{\partial}{\partial \boldsymbol{\xi}_2} \right). \quad (14)$$

Let $S(1, 2, 3)$ be any rotationally and translationally invariant state. Then

$$\psi(1, 2, 3) = \sum_{m_1} \sum_{m_2} (LM | l_1 m_1 l_2 m_2) Y_{l_1 m_1}(\boldsymbol{\xi}_1) Y_{l_2 m_2}(\boldsymbol{\xi}_2) S_{L l_1 l_2} \quad (15)$$

is a translationally invariant eigenstate of \mathbf{L}^2 and L_z with eigenvalues $L(L+1)\hbar^2$ and $M\hbar$. (The Y_{lm} are spherical harmonics and $(LM | l_1 m_1 l_2 m_2)$ are Clebsch-Gordan coefficients.) The problem is now to symmetrize these states. One set of states with the same permutation symmetry as S may be obtained easily by setting $l_2=0$. The non-zero terms in (15) have $l_1=L$. A symmetrized sum formed from (15) is proportional to

$$Y_{LM}(\mathbf{r}_{12}) + Y_{LM}(\mathbf{r}_{13}) + Y_{LM}(\mathbf{r}_{23})$$

with even L (because $Y_{LM}(-\mathbf{r}) = (-)^L Y_{LM}(\mathbf{r})$). This expression leads us to consider many-body wavefunctions proportional to

$$\frac{1}{2} \sum_{j=1}^N \sum_{i=1}^N Y_{LL}(\mathbf{r}_{ij}) \sim \frac{1}{2} \sum_{j=1}^N \sum_{i=1}^N \left(\frac{x_{ij} + iy_{ij}}{r_{ij}} \right)^L.$$

(It is sufficient to consider the state with eigenvalue of L_z equal to $L\hbar$, since the whole set of $2L+1$ states with different magnetic quantum numbers may then be obtained by means of the ladder operator $L_x - iL_y$).

We can generalize again, and consider many-body wavefunctions of the form $\Psi = F\Phi$, where

$$F = \frac{1}{2} \sum_{j=1}^N \sum_{i=1}^N (x_{ij} + iy_{ij})^L f(r_{ij}) \quad (16)$$

and Φ is any translationally and rotationally invariant state with either Bose or Fermi permutation symmetry. L must be even, otherwise F is zero. We shall now verify that the states $F\Phi$ are eigenfunctions of \mathbf{L}^2 and L_z with eigenvalues $L(L+1)\hbar^2$ and $L\hbar$, for arbitrary f . For $f = \text{constant}$ it will be shown that the wavefunctions are also energy eigenstates of a system with harmonic pair interactions. We have

$$\left. \begin{aligned} \frac{\partial F}{\partial x_k} &= \sum_j (x_{kj} + iy_{kj})^L x_{kj} r_{kj}^{-1} f'(r_{kj}) + L \sum_j (x_{kj} + iy_{kj})^{L-1} f(r_{kj}), \\ \frac{\partial F}{\partial y_k} &= \sum_j (x_{kj} + iy_{kj})^L y_{kj} r_{kj}^{-1} f'(r_{kj}) + iL \sum_j (x_{kj} + iy_{kj})^{L-1} f(r_{kj}), \\ \frac{\partial F}{\partial z_k} &= \sum_j (x_{kj} + iy_{kj})^L z_{kj} r_{kj}^{-1} f'(r_{kj}). \end{aligned} \right\} \quad (17)$$

Therefore

$$\begin{aligned} L_z F &= -i\hbar \sum_k \left(x_k \frac{\partial}{\partial y_k} - y_k \frac{\partial}{\partial x_k} \right) F \\ &= L\hbar \sum_k \sum_j (x_k + iy_k)(x_{kj} + iy_{kj})^{L-1} f(r_{kj}) \\ &\quad + i\hbar \sum_k \sum_j (x_k y_j - x_j y_k)(x_{kj} + iy_{kj})^L r_{kj}^{-1} f'(r_{kj}). \end{aligned}$$

The second term is zero ; to the first term we apply the identity

$$\sum_k \sum_j a_k A_{kj} = \sum_{k < j} (a_k - a_j) A_{kj} \quad (A_{kj} + A_{jk} = 0) \quad (18)$$

to obtain

$$L_z F = L\hbar F. \quad (19)$$

In a similar way we get

$$\begin{aligned} L_x F &= -i\hbar \sum_k \left(y_k \frac{\partial}{\partial z_k} - z_k \frac{\partial}{\partial y_k} \right) F \\ &= -L\hbar \sum_k \sum_j z_k (x_{kj} + iy_{kj})^{L-1} f(r_{kj}), \end{aligned} \quad (20)$$

$$\begin{aligned} L_y F &= -i\hbar \sum_k \left(z_k \frac{\partial}{\partial x_k} - x_k \frac{\partial}{\partial z_k} \right) F \\ &= -iL\hbar \sum_k \sum_j z_k (x_{kj} + iy_{kj})^{L-1} f(r_{kj}). \end{aligned} \quad (21)$$

Thus $(L_x + iL_y)F = 0$ (this verifies that we have the uppermost state) ; using the fact that

$$\begin{aligned} (L_x - iL_y)(L_x + iL_y) &= L_x^2 + L_y^2 + i[L_x, L_y] \\ &= L_x^2 + L_y^2 - \hbar L_z \end{aligned}$$

we find

$$(L_x^2 + L_y^2)F = \hbar L_z F = L\hbar^2 F. \quad (22)$$

Therefore

$$\mathbf{L}^2 F = L(L+1)\hbar^2 F. \quad (23)$$

The wavefunctions $\Psi = F\Phi$ have thus been shown to be angular momentum eigenstates, with $f(r)$ arbitrary, apart from the obvious requirements that F and its first and second derivatives should exist.

3. HARMONIC INTERACTIONS

We shall show that when Φ is the ground state of a system of N bosons with harmonic interactions, i.e.

$$V(\mathbf{r}_1, \mathbf{r}_2, \dots, \mathbf{r}_N) = \sum_{i < j} \sum v \left[1 + \frac{r_{ij}^2}{a^2} \right], \quad (24)$$

the state $\Psi = F\Phi$ with $f=1$ is an exact eigenstate of H as well as of L_z and \mathbf{L}^2 . For hamiltonians of type (1), with $H\Phi = E_0\Phi$, we have the identity

$$(H - E_0)\Psi = -\frac{\hbar^2}{2m} \Phi^{-1} \sum_k \nabla_k \cdot (\Phi^2 \nabla_k F). \quad (25)$$

When $f=1$ we see from (17) that $\nabla_k^2 F=0$, so that

$$(H-E_0)\Psi = -\frac{\hbar^2}{m} \sum_k \nabla_k \Phi \cdot \nabla_k F. \quad (26)$$

For harmonic pair interactions the ground state is well known [8–11]:

$$E_0 = \frac{1}{2}N(N-1)v + \frac{3}{2}(N-1) \left(\frac{\hbar^2 N v}{m a^2} \right)^{1/2}, \quad (27)$$

$$\Phi = \exp \left\{ - \left(\frac{m v}{N \hbar^2 a^2} \right)^{1/2} \sum_{i < j} \sum r_{ij}^2 \right\}. \quad (28)$$

Thus

$$\begin{aligned} \sum_k \nabla_k \Phi \cdot \nabla_k F &= -2L \left(\frac{m v}{N \hbar^2 a^2} \right)^{1/2} \Phi \sum_k \sum_j \sum_l (x_{kj} + i y_{kj})(x_{kl} + i y_{kl})^{L-1} \\ &= -2L \left(\frac{N m v}{\hbar^2 a^2} \right)^{1/2} \Phi \sum_k \sum_l (x_k + i y_k)(x_{kl} + i y_{kl})^{L-1} \\ &= -2L \left(\frac{N m v}{\hbar^2 a^2} \right)^{1/2} F \Phi, \end{aligned} \quad (29)$$

using the identity (18). Combining (29) with (26) we see that $\Psi = F\Phi$ is an eigenstate of H , with energy

$$E_L = E_0 + 2L \left(\frac{\hbar^2 N v}{m a^2} \right)^{1/2}. \quad (30)$$

Comparison with the two-body system [12] leads us to expect that these states have the lowest energy for given angular momentum L . The fact that the rotational excitation energy increases as L is not a general property of these angular momentum eigenstates, but it is interesting to note that there are many nuclei which approximate this behaviour [13].

4. ROTATIONAL ENERGIES IN THE GENERAL CASE

In § 2 it was shown that the functions $F\Phi$ have angular momentum L . They are therefore orthogonal to any state with $L=0$, and are thus suitable trial functions in a variational calculation. In general, the best f for given L and Φ may be determined by many-body variational methods which have recently been developed [14]. Here we shall make the simplest possible choice and take $f=1$. It will appear that the resulting rotational states are surface oscillations of the system.

By the variational principle, with $\Psi = F\Phi$,

$$E_L - E_0 \leq \frac{\int d1 \dots dN \Psi^* (H - E_0) \Psi}{\int d1 \dots dN |\Psi|^2} = \frac{N \hbar^2 \int d1 \dots dN |\nabla_1 F|^2 \Phi^2}{2m \int d1 \dots dN |F|^2 \Phi^2}. \quad (31)$$

We have used (25) and the fact the Φ has either Bose or Fermi permutation symmetry. We set $f=1$ and consider the two-body case first. Then $\Phi(\mathbf{r}_1, \mathbf{r}_2) = \Phi(r_{12})$, and

$$E_L - E_0 \leq \frac{2L^2 \hbar^2 \int d\mathbf{r} (x^2 + y^2)^{L-1} \Phi^2(r)}{m \int d\mathbf{r} (x^2 + y^2)^L \Phi^2(r)} = L(L + \frac{1}{2}) \frac{2\hbar^2}{m\bar{r}^2}, \quad (32)$$

where

$$\bar{r}^2 = \frac{\int_0^\infty dr r^{2L+2} \Phi^2(r)}{\int_0^\infty dr r^{2L} \Phi^2(r)}. \quad (33)$$

The general case of arbitrary N and L is much more complicated. We consider $L=2$ only, and use the notation

$$\langle A \rangle = \int d1 \dots dN A \Phi^2 / \int d1 \dots dN \Phi^2 \quad (34)$$

We have

$$|\nabla_1 F|^2 = 8 \sum_j \sum_k (x_{1j} x_{1k} + y_{1j} y_{1k}), \quad (35)$$

so that

$$\begin{aligned} \langle |\nabla_1 F|^2 \rangle &= 8(N-1) \langle x_{12}^2 + y_{12}^2 \rangle + 8(N-1)(N-2) \langle x_{12} x_{13} + y_{12} y_{13} \rangle \\ &= 4N(N-1) \langle x_{12}^2 + y_{12}^2 \rangle, \end{aligned} \quad (36)$$

where we have made use of the identity

$$2x_{12} x_{13} = x_{12}^2 + x_{13}^2 - x_{23}^2. \quad (37)$$

Similarly,

$$|F|^2 = \frac{1}{4} \sum_i \sum_j \sum_k \sum_l \{ (x_{ij} x_{kl} + y_{ij} y_{kl})^2 - (x_{ij} y_{kl} - y_{ij} x_{kl})^2 \}, \quad (38)$$

so that

$$\begin{aligned} \langle |F|^2 \rangle &= \frac{1}{4} N(N-1) \langle \sum_k \sum_l \{ (x_{12} x_{kl} + y_{12} y_{kl})^2 - (x_{12} y_{kl} - y_{12} x_{kl})^2 \} \rangle \\ &= \frac{1}{2} N(N-1) \langle (x_{12}^2 + y_{12}^2)^2 \rangle \\ &\quad + \frac{1}{2} N(N-1)(N-2) \langle (x_{12} x_{13} + y_{12} y_{13})^2 - (x_{12} y_{13} - y_{12} x_{13})^2 \rangle \\ &\quad + \frac{1}{2} N(N-1)(N-2)(N-3) \langle (x_{12} x_{34} + y_{12} y_{34})^2 \\ &\quad - (x_{12} y_{34} - y_{12} x_{34})^2 \rangle. \end{aligned} \quad (39)$$

To define an effective moment of inertia for a given rotational state we compare the excitation energy with that of a rigid body, and write

$$E_L - E_0 = \frac{L(L+1)\hbar^2}{2I_L}. \quad (40)$$

Comparison with the variational equation (31) then gives a lower bound for the effective moment of inertia I_L :

$$I_L \geq \frac{L(L+1)m \langle |F|^2 \rangle}{N \langle |\nabla_1 F|^2 \rangle}. \quad (41)$$

For general N , $\langle |F|^2 \rangle$ as given by (39) contains terms depending on two, three and four-particle correlations. The four-particle term is dominant for large N ; neglecting terms of order N^{-1} we find

$$I_2 \geq \frac{3}{4} Nm \frac{\langle (x_{12}^2 - y_{12}^2)(x_{34}^2 - y_{34}^2) + 4x_{12}y_{12}x_{34}y_{34} \rangle}{\langle x_{12}^2 + y_{12}^2 \rangle}. \quad (42)$$

Bohr [15] has calculated the energy of an $L=2$ surface oscillation of a classical droplet. He was able to separate the rotational and vibrational parts of the kinetic energy; his definition of the moment of inertia is in terms of the rotational energy. He finds

$$\mathcal{I}_2 = Nm \frac{\int d\mathbf{r} \rho(\mathbf{r})(x^2 - y^2)^2}{\int d\mathbf{r} \rho(\mathbf{r})(x^2 + y^2)}, \quad (43)$$

where $\rho(\mathbf{r})$ is the mass density distribution function and the x and y coordinates are measured relative to the centre of mass. Comparison of (43) with (42) leads us to conclude that, for large N , the rotational states approximated by

$$\Psi = \frac{1}{2} \sum_i \sum_j (x_{ij} + iy_{ij})^L \Phi$$

represent surface oscillations of the system. (The second term in (42) corresponds to the vibrational energy which can be subtracted out in the classical case.) The fact that the excitation energy depends on four-particle correlations has a simple physical interpretation: we need at least two-particle states to construct translationally invariant excitations, and the simplest way to obtain a quadrupole oscillation is to excite two dipole oscillations simultaneously.

The two classical analogues of rotational excitations, namely surface oscillations and rigid-body rotations, have classical moments of inertia given respectively by (43) and

$$I = \int d\mathbf{r} \rho(\mathbf{r})(x^2 + y^2). \quad (44)$$

Nuclei appear to fall between these two extremes (of fluid and solid motion): for the so-called 'non-spherical' nuclei the moment of inertia lies between the values estimated from the classical expressions (43) and (44), being somewhat closer to the rigid-body value [16, 17]. In the molecular case, we expect large helium molecules to be microdroplets, i.e. to have surface oscillations of the liquid drop type, and the heavier rare-gas molecules to have rigid-body rotations. Our microscopic theory gives a value for the surface oscillation type of rotation which depends on four-particle correlations, and is therefore difficult to estimate. Nevertheless, it is clear from comparison of (42) and (43) that there may be substantial differences between the rotational energies calculated semi-classically and quantum-mechanically.

5. OTHER ROTATIONAL STATES

We have shown that the states $\Psi = F\Phi$ with

$$F = \frac{1}{2} \sum_i \sum_j (x_{ij} + iy_{ij})^L f(r_{ij}) \quad (16)$$

are angular momentum eigenstates for rotationally invariant states Φ . For each $L=0$ vibrational state Φ , including the ground state, one can in principle

obtain the best function $f(r)$ for given L . These states have even L and therefore have the same parity as Φ . One can construct states from products as well as sums, and from combinations of products and sums. The simplest products have the form

$$F = \prod_{i < j} (x_{ij} + iy_{ij})^K. \quad (45)$$

These are angular momentum eigenstates, with

$$L = \frac{1}{2}KN(N-1). \quad (46)$$

The exchange symmetry of these functions is $(-)^K$, and the inversion symmetry is $+$ for even K and $(-)^{(1/2)N(N-1)}$ for odd K . Their angular momentum is too high to be of interest, except for small N . Our main purpose in writing them down was to show that other many-body wavefunctions with the correct symmetries exist. The three and four-body problems have been studied extensively, and catalogues of angular momentum eigenfunctions have been given [18, 19]. The N -body problem has been studied group-theoretically [20], but the only translationally invariant wave functions with correct permutation symmetry known to the author have been given in this paper.

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