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1982 J. Phys. A: Math. Gen. 15 2785

(http://iopscience.iop.org/0305-4470/15/9/028)

J. Phys. A: Math. Gen. **15** (1982) 2785–2799. Printed in Great Britain

Collective rotations of asymmetrically deformed many-body systems

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Received 30 December 1981

Abstract. The angular momentum projection technique is worked out for systems with an intrinsic asymmetric deformation. These include asymmetric top molecules and triaxial nuclei. A parametrisation of the rotation group is introduced which allows an approximate but analytical evaluation of angular momentum projected matrix elements. Via symmetric orthonormalisation a quantal definition of the moments of inertia is obtained.

1. Introduction

Many-body systems such as nuclei and molecules display energy level patterns that strongly resemble the spectra of rigid rotors. It is common to explain this feature as being due to the collective rotation of the constituents (the nucleons in the nucleus, the electrons and nuclei in the molecule). Thus a quantitative explanation is obtained by identifying the system with a rigid rotor having internal degrees of freedom to account for the non-rotational aspects of the spectra. Models constructed along these lines are the Bohr–Mottelson (1975) approach for nuclei and the (crude) Born–Oppenheimer (1927) approximation for molecules. The main criticism against these models is that in their primitive form, in which they are usually presented in textbooks, they are not derived from the full microscopic Hamiltonian of the many-body system. This situation can be remedied in two ways: by transforming the Hamiltonian to a rotating frame of reference or by using angular momentum projection techniques. Both these alternatives make sure that rotational invariance is properly accounted for; however, they show some marked differences both from a theoretical and a practical point of view (Lathouwers 1980a, b). In using either one of these methods one has to distinguish between spherically symmetric, axially symmetric and asymmetric cases according to whether the spectra are reminiscent of those of the corresponding rotor types. Whereas the frame transformation method has been worked out for all three situations, the general projection theory is not available for asymmetrically deformed systems. The presentation of such a scheme is the purpose of this paper.

Approximate angular momentum projection for axially symmetric nuclei was developed by Peierls and Yoccoz (1957) within the framework of the generator coordinate (GC) method (Griffin and Wheeler 1957). This approach was carried over to the GC theory of diatomic molecules (Lathouwers 1978, 1980a, b, Lathouwers and

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0305-4470/82/092785+15$02.00

Van Leuven 1982). In both cases the starting point is a strongly deformed intrinsic state possessing axial symmetry. In order to generalise the above schemes to asymmetric situations, we will drop all restrictions on the choice of the intrinsic state except those of strong deformation in all directions and reality. Mathematically this can be expressed as

$$\langle\chi|\mathcal{R}(\omega, \boldsymbol{u})|\chi\rangle = \langle\chi|e^{-i\omega \boldsymbol{J}\cdot\boldsymbol{u}}|\chi\rangle = \text{a sharply peaked function of } \omega \tag{1}$$

where $\mathcal{R}(\omega, \boldsymbol{u})$ is the angle-axis form of a rotation operator, i.e. $\boldsymbol{J}$ is the total angular momentum of the system, ω the rotation angle and $\boldsymbol{u}$ a unit vector. Equation (1) states that the intrinsic function $\chi(x)$ is so deformed that any rotation, specified by a rotation angle ω and a unit vector $\boldsymbol{u}$, will transform it into a function having a small overlap with the original. In other words, the scalar product of $\chi(x)$ and $\mathcal{R}(\omega, \boldsymbol{u})\chi(x)$ is a rapidly decreasing function of ω. It is common to supplement (1) with a similar assumption for matrix elements obtained by inserting the Hamiltonian. Therefore as a second consequence of strong deformation we will assume that

$$\langle\chi|H\mathcal{R}(\omega, \boldsymbol{u})|\chi\rangle / \langle\chi|\mathcal{R}(\omega, \boldsymbol{u})|\chi\rangle = \text{a slowly varying function of } \omega. \tag{2}$$

Together with the convention that $\chi(x)$ is a real function, (1) and (2) constitute the basic assumptions of this paper. We will show that this includes all asymmetric top molecules if the molecular intrinsic functions are constructed as products of electronic states and strongly localised nuclear wavepackets. Triaxial nuclei in the mass region $A \cong 135$ and $A \cong 190$ also fall into this category. In this case the intrinsic state may be chosen as the result of an unrestricted Hartree–Fock or Hartree–Fock–Bogoliubov calculation.

We proceed as follows. Section 2 gives a summary of the angular momentum projection procedure and yields the basic matrix equation for the rotational energies. It is observed that the representation of the rotation operators in terms of Euler angles is not suited for the approximate evaluation of the projected overlap and Hamiltonian matrices. Section 3 therefore presents an alternative parametrisation in which infinitesimal rotations are easily characterised while the representations of the rotation group still keep a simple form. Equipped with this new parametrisation, the Gaussian overlap and quadratic approximations are established in § 4. Here an important tensor, the angular momentum fluctuation tensor, is introduced. In its most general form it reads

$$F_{kl} = \langle\chi|\tfrac{1}{2}(J_kJ_l + J_lJ_k)|\chi\rangle - \langle\chi|J_k|\chi\rangle\langle\chi|J_l|\chi\rangle \tag{3}$$

where the J_k are the components of the angular momentum vector. The integration over the rotation group is considerably simplified if the intrinsic state is oriented such as to diagonalise F_{kl}. The evaluation of the projected overlap and Hamiltonian matrices to lowest order is done in § 5. These results do not yet allow a clear-cut physical interpretation, due to the resulting non-diagonal metric. For this purpose we make use of symmetric orthonormalisation which reduces the generalised eigenvalue problem to a single matrix diagonalisation. The Hamiltonian matrix after orthonormalisation then has the desired form, i.e. depends upon the total angular momentum and azimuthal quantum numbers in exactly the same way as the matrix of a rigid rotor Hamiltonian in a basis of Wigner D functions. A quantal definition of the three moments of inertia for the asymmetrically deformed system is then possible. These

numbers turn out to be the eigenvalues of a symmetric inertia tensor which, in general, is not diagonal. The spectrum at the lowest level of approximation is that of an effective rigid rotor.

2. Angular momentum projections

Deformed states, such as those described in the introduction, are not necessarily eigenstates of the pair of operators J^2 and J_z. This is a consequence of the fact that they result from variational principles including functions out of a subspace of the system's full Hilbert space. The $\chi(x)$ therefore define specific directions in space. However, it is clear that all rotated states $\mathcal{R}(\omega, \boldsymbol{u})\chi(x)$ have the same energy expectation value. This degeneracy can be removed by considering the trial states

$$\Psi(x) = \int \mathrm{d}\Omega\, F(\Omega)\mathcal{R}(\Omega)\chi(x), \tag{4}$$

$$\mathcal{R}(\Omega) = \mathrm{e}^{-\mathrm{i}\varphi J_z}\,\mathrm{e}^{-\mathrm{i}\theta J_y}\,\mathrm{e}^{-\mathrm{i}\gamma J_z}, \qquad \int \mathrm{d}\Omega \equiv \int_0^{2\pi} \mathrm{d}\varphi \int_0^{\pi} \sin\theta\, \mathrm{d}\theta \int_0^{2\pi} \mathrm{d}\gamma, \tag{5}$$

where we have introduced the common expression (see e.g. Messiah 1960) for the rotation operators in terms of three Euler angles $\Omega \equiv (\varphi, \theta, \gamma)$. The variationally optimal functions $F(\Omega)$ satisfy the integral equation

$$\int [H(\Omega, \Omega') - E\Delta(\Omega, \Omega')]F(\Omega')\,\mathrm{d}\Omega' = 0, \tag{6}$$

$$H(\Omega, \Omega') = \langle\chi|\mathcal{R}^+(\Omega)H\mathcal{R}(\Omega')|\chi\rangle, \qquad \Delta(\Omega, \Omega') = \langle\chi|\mathcal{R}^+(\Omega)\mathcal{R}(\Omega')|\chi\rangle. \tag{7}$$

This is a special case of the Wheeler equation in the generator coordinate theory (Griffin and Wheeler 1957) if one considers the Euler angles as GC. When the parameters of a continuous group, here the rotation group, are used as GC, group representation theory allows a partial solution of the Wheeler integral equation. Indeed, if we expand the $F(\Omega)$ in terms of Wigner D functions

$$F(\Omega) = \sum_{JMK} c^J_{MK} D^{J*}_{MK}(\Omega), \tag{8}$$

$$D^J_{MK}(\Omega) \equiv D^J_{MK}(\varphi, \theta, \gamma) = \mathrm{e}^{-\mathrm{i}M\varphi}\, d^J_{MK}(\theta)\mathrm{e}^{-\mathrm{i}K\gamma}, \tag{9}$$

and make use of the well known properties of these so-called 'rotation matrices', the Wheeler equation (6) is reduced to a set of matrix equations, one for each total angular momentum value J:

$$\sum_L (H^J_{KL} - E^J \Delta^J_{KL}) c^J_L = 0, \tag{10}$$

$$H^J_{KL} = \frac{2J+1}{8\pi^2} \int \mathrm{d}\Omega\, D^{J*}_{KL}(\Omega)\langle\chi|H\mathcal{R}(\Omega)|\chi\rangle, \tag{11}$$

$$\Delta^J_{KL} = \frac{2J+1}{8\pi^2} \int \mathrm{d}\Omega\, D^{J*}_{KL}(\Omega)\langle\chi|\mathcal{R}(\Omega)|\chi\rangle. \tag{12}$$

The appearance of the factor $(2J+1)/8\pi^2$ is explained below. Each of these equations

yields a set of energy levels E_τ^J with $\tau = 1, 2, \ldots, 2J+1$. These can be interpreted as rotational states since they are generated by diagonalising the Hamiltonian in a basis set $\{\mathscr{R}(\Omega)\chi(x)\}$ of rotated intrinsic states.

The procedure just described within the framework of the GC method is equivalent to what is known as the angular momentum projection technique. Angular momentum projectors are integral operators of the form

$$P_{MK}^J = \frac{2J+1}{8\pi^2} \int \mathrm{d}\Omega\, D_{MK}^{J*}(\Omega)\mathscr{R}(\Omega). \tag{13}$$

They are continuous analogues of the projection operator for finite groups, i.e. the sums over the group of the group elements with coefficients given by the irreducible representations. The factor $2J+1$ refers to the dimension of the irreducible representations while $8\pi^2$ equals 'the number of group elements'. The action of P_{MK}^J on an arbitrary state $\chi(x)$ produces an eigenfunction of J^2 and J_z with quantum numbers J and M respectively. The connection between the angular momentum projectors and the GC type approach becomes clear if one substitutes the expanded form (8) of $F(\Omega)$ into the integral (6) to obtain (observe that the eigenvectors of the secular equations (10) are independent of M)

$$\Psi(x) = \sum_{JM} \Psi_{JM}(x), \tag{14}$$

$$\Psi_{JM}(x) = \sum_K c_K^J \int \mathrm{d}\Omega\, D_{MK}^{J*}(\Omega)\mathscr{R}(\Omega)\chi(x) = \frac{8\pi^2}{2J+1} \sum_K c_K^J P_{MK}^J \chi(x). \tag{15}$$

This shows that the $\Psi(x)$ are sums of J^2 and J_z eigenfunctions $\Psi_{JM}(x)$, each of which is a variationally determined superposition of angular momentum projected states $P_{MK}^J\chi(x)$. Reversing the arguments, one can say that the original intrinsic function, being strongly deformed, has a small uncertainty in orientation and hence, according to Heisenberg's principle, contains a large spread of angular momentum values. The state $\chi(x)$ can therefore be considered as a wavepacket of angular momentum eigenfunctions whose components are sorted out by the projection technique. The consistency of (10)–(12) with (14)–(15) is easily checked using the properties

$$(P_{MK}^J)^+ = P_{KM}^J, \qquad [H, P_{MK}^J] = 0, \qquad P_{KM}^J P_{ML}^J = P_{KL}^J, \tag{16}$$

which follow directly from the definition (13). For details on the use of the projection technique in axially symmetric cases, where considerable simplifications arise, we refer to Kelemen and Dreizler (1976). Here we will deal only with totally asymmetric situations. Small deviations from axial symmetry have been discussed by Villars and Rogerson (1971).

The main goal of this paper is to investigate the structure of the spectra resulting from secular equations of the type (10) if the starting point is a strongly deformed intrinsic state. We therefore need to know how the projected matrix elements (11) and (12) depend upon the quantum numbers J, K and L. In view of the consequences of strong deformation, equations (1) and (2), we expect the contributions from infinitesimal rotations to be most important in the calculation of Δ_{KL}^J and H_{KL}^J. Thus we have to use a parametrisation of the rotation group which allows us easily to identify small rotations. In addition, to perform the integrations in (11) and (12)

analytically the associated irreducible representations must be known explicitly and preferably be of a simple form. In the next section we will introduce a parametrisation of the rotation group that meets this dual requirement.

3. Parametrisations of the rotation group

We have already used two representations of rotation operators. The first one involved a single rotation angle ω and a rotation axis $\boldsymbol{u}$

$$\mathcal{R}(\omega, \boldsymbol{u}) = \mathrm{e}^{-\mathrm{i}\omega \boldsymbol{J}\cdot\boldsymbol{u}}. \tag{17}$$

It is known as the angle-axis form of rotation operators. Observe that $\|\boldsymbol{u}\| = 1$ and that the correspondence is not unique since $\mathcal{R}(\omega, \boldsymbol{u}) = \mathcal{R}(-\omega, -\boldsymbol{u})$. The second one is commonly used in the projection technique and employs the Euler parametrisation (5). The connection between the respective parameters is well known (Casimir 1931)†

$$\begin{aligned} u_x \sin\tfrac{1}{2}\omega &= \sin\delta \sin\tfrac{1}{2}\theta, & u_y \sin\tfrac{1}{2}\omega &= \cos\delta \sin\tfrac{1}{2}\theta, \\ u_z \sin\tfrac{1}{2}\omega &= \sin\varepsilon \cos\tfrac{1}{2}\theta, & \cos\tfrac{1}{2}\omega &= \cos\varepsilon \cos\tfrac{1}{2}\theta, \end{aligned} \tag{18}$$

where $\varepsilon = (\varphi + \gamma)/2$ and $\delta = (\varphi - \gamma)/2$ are the sum and difference angles. For our purpose, i.e., the approximate but analytical evaluation of the projected matrix elements H^J_{KL} and Δ^J_{KL}, we need a parametrisation which on the one hand allows a location of infinitesimal rotations and on the other hand yields a simple functional form of the group representations. The first form (17) has the first property, since infinitesimal rotations clearly correspond to infinitesimal rotation angles ω. However, the expression of Wigner D functions in terms of ω and the spherical angles of $\boldsymbol{u}$ is complicated (Kerman and Onishi 1977). Alternatively, the Wigner D-functions emerge in a natural way from the Euler representation, hence the second requirement is then fulfilled. However, it is easy to convince oneself that the infinitesimal angles φ, θ and γ are not the only ones which generate infinitesimal rotations. Therefore neither of the two forms considered so far is suitable for our needs and a third one is called for.

Fortunately it is relatively easy to discover which parametrisation has the two qualities mentioned above. Indeed, from the set of equations (18) it follows that infinitesimal angles ω necessarily correspond to infinitesimal values of θ and ε. The difference angle δ, however, is arbitrary in this respect. From the Euler form one can then derive the rotation operator, expressed in terms of θ, ε and δ, in the following way:

$$\begin{aligned} \mathrm{e}^{-\mathrm{i}\varphi J_z}\mathrm{e}^{-\mathrm{i}\theta J_y}\mathrm{e}^{-\mathrm{i}\gamma J_z} &= \mathrm{e}^{-\mathrm{i}\varepsilon J_z}\mathrm{e}^{-\mathrm{i}\delta J_z}\mathrm{e}^{-\mathrm{i}\theta J_y}\mathrm{e}^{-\mathrm{i}\delta J_z}\mathrm{e}^{-\mathrm{i}\varepsilon J_z} \\ &= \mathrm{e}^{-\mathrm{i}\varepsilon J_z}\,\mathcal{R}(\delta, \boldsymbol{e}_z)\,\mathrm{e}^{-\mathrm{i}\theta J_y}\,\mathcal{R}^{+}(\delta, \boldsymbol{e}_z)\,\mathrm{e}^{-\mathrm{i}\varepsilon J_z} \\ &= \mathrm{e}^{-\mathrm{i}\varepsilon J_z}\exp\left[-\mathrm{i}\theta(J_y\cos\delta - J_x\sin\delta)\right]\mathrm{e}^{-\mathrm{i}\varepsilon J_z}. \end{aligned} \tag{19}$$

For the group representations, a simple substitution of φ and γ in terms of ε and δ immediately gives the expression for the rotation matrices,

$$\mathrm{e}^{-\mathrm{i}K\varphi}\, d^J_{KL}(\theta)\,\mathrm{e}^{-\mathrm{i}L\varphi} = \mathrm{e}^{-\mathrm{i}(K+L)\varepsilon}\, d^J_{KL}(\theta)\,\mathrm{e}^{-\mathrm{i}(K-L)\delta}. \tag{20}$$

† It should be remarked that Casimir's (1931) definition of the Euler angles is different from that of Messiah (1960). One has $\varphi_C = \varphi_M + \pi/2$, $\theta_C = \theta_M$ and $\gamma_C = \gamma_M - \pi/2$.

Although the rotation operator derived in (19) is slightly more complicated than the Euler form, and considerably more complex than the angle-axis form, it satisfies our present needs. Indeed, we can identify infinitesimal rotations with infinitesimal values of the two angles ε and θ (not possible in Euler form), while the Wigner D functions are as simple as the common expressions (complications arise at this point with the angle-axis form).

The situation becomes even more clear if we consider the transformation from the Euler parametrisation to the new one in the φ, γ or ε, δ plane. The situation is illustrated in figure 1.

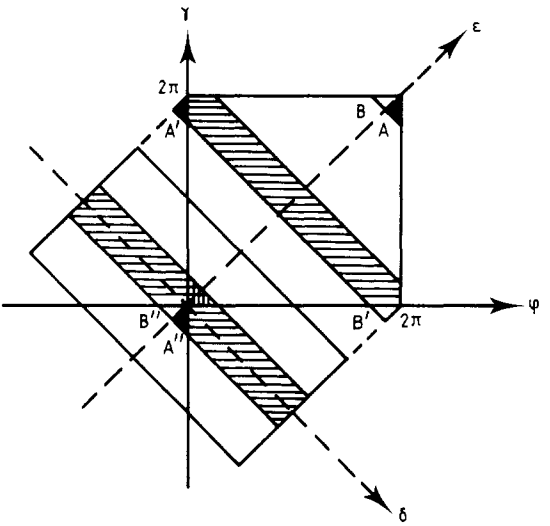

Figure 1. Locations of infinitesimal rotations in the (φ, γ) and (ϵ, δ) planes.

The integration domain originally consists of the square $0 \leq \varphi \leq 2\pi$, $0 \leq \gamma \leq 2\pi$. Within this domain the infinitesimal rotations are located in the lower left and upper right corners and along the diagonal from the upper left to the lower right corner. Thus, the regions of interest do not form a connected subset of the square which complicates the integration. However, since the integrand is a periodic function of φ and γ with period 2π, it is allowed to translate portions of the original square both vertically and horizontally over a distance 2π. A little jig-saw puzzle on figure 1 then shows that the integration over the square is equivalent to that over the rectangle $-\pi/2 \leq \varepsilon \leq +\pi/2$, $-\pi \leq \delta \leq +\pi$, i.e.

$$\int_0^{2\pi} \mathrm{d}\varphi \int_0^{2\pi} \mathrm{d}\gamma = 2 \int_{-\pi/2}^{+\pi/2} \mathrm{d}\varepsilon \int_{-\pi}^{+\pi} \mathrm{d}\delta \tag{21}$$

where the factor 2 arises from the Jacobian of the transformation.

Because of the advantages of the (ε, θ, δ) parametrisation, we will from now on denote rotation operators by

$$\mathscr{R}(\varepsilon, \theta, \delta) = \mathrm{e}^{-\mathrm{i}\varepsilon J_z} \exp\left[-\mathrm{i}\theta(J_y \cos\delta - J_x \sin\delta)\right] \mathrm{e}^{-\mathrm{i}\varepsilon J_z}. \tag{22}$$

Confusion with the previous notation for the Euler form will be avoided by always specifying the arguments. The use of the new parametrisation should also not be

confused with the way Euler angles actually parametrise the intrinsic states. The use of Euler angles as GC is based on defining a GC intrinsic state as $\chi(x|\Omega) \equiv \mathcal{R}(\Omega)\chi(x)$, whereas the $(\varepsilon, \theta, \delta)$ parametrisation is introduced at the level of the secular equations (10), resulting from a partial solution of the Wheeler equation (6).

4. Gaussian overlap and quadratic approximations

The purpose of this section is to provide simple, yet general, approximations for the expectation values appearing in the angular momentum projected matrix elements (11) and (12). First (19), (20) and (21) may be used to rewrite the definition of Δ^J_{KL} and H^J_{KL} as follows:

$$\Delta^J_{KL} = \frac{2J+1}{4\pi^2} \int_{-\pi/2}^{+\pi/2} d\varepsilon \int_0^{\pi} \sin\theta \, d\theta \int_{-\pi}^{+\pi} d\delta \, \Delta(\varepsilon, \theta, \delta) \, e^{i(K+L)\varepsilon} \, d^J_{KL}(\theta) \, e^{i(K-L)\delta}, \tag{23}$$

$$H^J_{KL} = \frac{2J+1}{4\pi^2} \int_{-\pi/2}^{+\pi/2} d\varepsilon \int_0^{\pi} \sin\theta \, d\theta \int_{-\pi}^{+\pi} d\delta \, H(\varepsilon, \theta, \delta) \, e^{i(K+L)\varepsilon} \, d^J_{KL}(\theta) \, e^{i(K-L)\delta}. \tag{24}$$

The functions $\Delta(\varepsilon, \theta, \delta)$ and $H(\varepsilon, \theta, \delta)$ are given in terms of the rotation operators in the form (22) by

$$\Delta(\varepsilon, \theta, \delta) = \langle\chi|\mathcal{R}(\varepsilon, \theta, \delta)|\chi\rangle, \qquad H(\varepsilon, \theta, \delta) = \langle\chi|H\mathcal{R}(\varepsilon, \theta, \delta)|\chi\rangle. \tag{25}$$

For strongly deformed states $\chi(x)$ these functions behave like generator coordinate kernels, corresponding to intrinsic states with a pronounced dependence on the GC. Hence, the same techniques will apply (Griffin and Wheeler 1957).

The Gaussian overlap approximation (GOA) is a celebrated procedure in GC methods. It assumes that the overlap between neighbouring intrinsic states is essentially a Gaussian in the difference of the GC. A similar situation arises here since we expect the overlap function $\Delta(\varepsilon, \theta, \delta)$ to be small for all but the infinitesimal rotations. Hence we assume the form

$$\Delta(\varepsilon, \theta, \delta) \cong \exp\left[-a(\delta)\varepsilon^2 - 2b(\delta)\varepsilon\theta - c(\delta)\theta^2\right] \tag{26}$$

consistent with the fact that infinitesimal rotations correspond to small angles θ and ε. Since the difference angle δ is arbitrary the damping coefficients in the GOA (26) are in general δ dependent. Direct identifications of the quadratic terms in the GOA with those obtained expanding the exponentials in the rotation operator yield the expressions

$$\begin{aligned} &a = 2\langle J_z^2\rangle, \qquad b(\delta) = \langle J_y J_z\rangle \cos\delta - \langle J_x J_z\rangle \sin\delta, \\ &c(\delta) = \tfrac{1}{2}(\langle J_x^2\rangle \sin^2\delta + \langle J_y^2\rangle \cos^2\delta - \langle J_x J_y\rangle \sin 2\delta), \end{aligned} \tag{27}$$

where we have adopted the notation $\langle J\rangle \equiv \langle\chi|J|\chi\rangle$ for expectation values of operators with respect to the intrinsic state. Notice that the scale factor a corresponding to rotations around the z axis is independent of δ. If $\chi(x)$ is real, as was assumed from the start, no linear terms appear due to time reversibility which implies that $\langle J_k\rangle = 0$.

One sees from (27) that the rate at which the overlap function goes to zero is measured by the components of the angular momentum fluctuation tensor, which in the time reversible case reduces to

$$F_{kl} = \langle J_k J_l\rangle \equiv \langle\chi|J_k J_l|\chi\rangle. \tag{28}$$

The numerical values of the F_{kl} depend upon the orientation of the intrinsic function. Indeed, if one computes these numbers for the rotated state $\mathcal{R}(\varepsilon_0, \theta_0, \delta_0)\chi(x)$, one obtains

$$F^0_{kl} = U^+(\varepsilon_0, \theta_0, \delta_0)F_{kl}\, U(\varepsilon_0, \theta_0, \delta_0) \tag{29}$$

where $U(\varepsilon_0, \theta_0, \delta_0)$ is the unitary matrix specifying the effect of $\mathcal{R}(\varepsilon_0, \theta_0, \delta_0)$ in coordinate space. This implies that the orientation of $\chi(x)$ is optional and may be chosen in such a way as to simplify the theory. It will turn out to be computationally favourable to orient $\chi(x)$ such that the angular momentum fluctuation tensor is diagonal. With this convention the GOA reduces to

$$\Delta(\varepsilon, \theta, \delta) = \exp(-a\varepsilon^2)\exp[-c(\delta)\theta^2], \tag{30}$$

$$c(\delta) = \tfrac{1}{4}[\langle J_x^2\rangle + \langle J_y^2\rangle - \cos 2\delta(\langle J_x^2\rangle - \langle J_y^2\rangle)], \tag{31}$$

which is a factorised expression in the angles, ε and θ, referring to infinitesimal rotations.

The actual values of a and $c(\delta)$ for molecular and nuclear many-body systems are determined by the expectation values $\langle J_k^2\rangle$. For strongly deformed nuclei Villars (1966) indicates that 10^2 is a reasonable value. In the molecular case the strong localisation of nuclei in the intrinsic state implies values of the order 10^4 for the $\langle J_k^2\rangle$ (Lathouwers and Van Leuven 1982). Thus in both cases the GOA will provide a natural cut-off radius for the θ and ε integrals.

Complementary to the GOA is the so-called quadratic approximation (QA). It is based on the assumption that the behaviour of the Hamiltonian kernel in a GC procedure is very similar to that of the overlap kernel. In that case their ratio will be a slowly varying function of the GC which can be expanded in a Taylor series truncated at second order. Translated into the present terminology, this implies the approximation of the quotient $H(\varepsilon, \theta, \delta)/\Delta(\varepsilon, \theta, \delta)$ by a second-order polynomial in ε and θ, i.e.

$$H(\varepsilon, \theta, \delta)/\Delta(\varepsilon, \theta, \delta) \cong E(0) - \tfrac{1}{2}[A(\delta)\varepsilon^2 + 2B(\delta)\theta\varepsilon + C(\delta)\theta^2] \tag{32}$$

where the linear terms are again absent due to time reversibility. Straightforward calculation gives for the coefficients in the QA the results

$$E(0) = \langle H\rangle, \tag{33}$$

$$A = 4\langle[H - E(0)]J_z^2\rangle, \tag{34}$$

$$B(\delta) = 2(\langle[H - E(0)]J_yJ_z\rangle\cos\delta - \langle[H - E(0)]J_xJ_y\rangle\sin\delta), \tag{35}$$

$$C(\delta) = \langle[H - E(0)]J_x^2\rangle\sin^2\delta + \langle[H - E(0)]J_y^2\rangle\cos^2\delta - \langle[H - E(0)]J_xJ_y\rangle\sin 2\delta. \tag{36}$$

As in the GOA the coefficient of ε^2 is δ independent. From these formulae it is clear that a second tensor will be of importance. It is obtained from the angular momentum fluctuation tensor by inserting the Hamiltonian devoid of the intrinsic energy $E(0)$, i.e.

$$K_{kl} = \langle[H - E(0)]J_kJ_l\rangle. \tag{37}$$

If the orientation of $\chi(x)$ is chosen such that F_{kl} is diagonal, there is no reason to expect that K_{kl} will share this property. Hence, there is in general no decoupling in the ε and θ angles in the QA. .

At this point it might be worthwhile to clarify the statement 'the intrinsic state defines a specific direction in space' and its relation to the angular momentum fluctuation tensor. In general one will start with an intrinsic state, defined relative to a space-fixed reference system, which does not yield a diagonal angular momentum fluctuation tensor. In fact, each of the rotated states $\mathcal{R}(\varepsilon_0, \theta_0, \delta_0)\chi(x)$ produces different values for the elements F_{kl} according to (29). Thus the numerical values of the angular momentum fluctuation tensor can be considered as the signatures of the directional properties of the intrinsic state. Above we have adopted the convention that, as a first step in the application of the theory, one should diagonalise the matrix $\langle\chi|J_kJ_l|\chi\rangle$ associated with the original intrinsic state. As a result of this computation one can specify three angles ε_0, θ_0 and δ_0 and reorient the intrinsic state by putting $\chi_0(x) = \mathcal{R}(\varepsilon_0, \theta_0, \delta_0)\chi(x)$. From this point onwards all expectation values are taken with respect to $\chi_0(x)$. Therefore, the theory can be developed further using the simplified form of the overlap kernel which results from the fact that the angular momentum fluctuation tensor associated with $\chi_0(x)$ is diagonal. This step in the application of the present scheme is analogous to the explicit definition of body-fixed axes in terms of particle coordinates in the frame transformation theory.

5. Overlap and Hamiltonian matrices

In order to study energy level patterns arising from secular equations like (10), we need to know the dependence of the matrix elements Δ^J_{KL} and H^J_{KL} upon the quantum numbers J, K and L. This necessitates an analytical evaluation of the three-dimensional integrals (23) and (24). Two factors simplify this task. The first one is the diagonal form of the angular momentum fluctuation tensor by appropriately choosing the orientation of the intrinsic state. This leads to decoupled ε and θ integrals. Secondly, the scale factors a and $c(\delta)$ cause the integrands to be essentially zero if ε^2 and θ^2 are much larger than $1/\langle J_k^2\rangle$ (for nuclei and molecules this means much larger than 10^{-2} or 10^{-4}). Therefore we can replace the integrands, excluding the Gaussians, by their asymptotic values for small ε and θ, at the same time extending the integration ranges to $-\infty$, $+\infty$ and 0, $+\infty$, respectively. We take into account the asymptotic form near $\theta = 0$ of the $d^J_{KL}(\theta)$, i.e.

$$d^J_{KK}(\theta) \underset{\theta\to 0}{\sim} 1 - \tfrac{1}{4}\theta^2[J(J+1) - K^2] + \ldots,$$

$$d^J_{KL}(\theta) \underset{\theta\to 0}{\sim} \left[\binom{J+K}{J+L}\binom{J-L}{J-K}\right]^{1/2}\left(-\frac{\theta}{2}\right)^{K-L} + \ldots, \tag{38}$$

for $K > L$ (see e.g. Verhaar 1964). These are good approximations provided the total angular momentum is not too large. Indeed, the $d^J_{KL}(\theta)$ show an increasing number of oscillations if one goes to larger and larger J values. Requiring the first node to be well outside the cut-off range provided by the GOA yields a maximum value of $J \cong \langle J_k^2\rangle^{1/2}$. This implies that the asymptotic forms (38) can safely be used up to $J \cong 10$ or $J \cong 100$ for nuclei and molecules respectively.

Assuming (38) to be valid, all integrations in (23) and (24) can be performed analytically provided one starts with the δ integration. This is not only convenient but also necessary. Indeed, the integrals over δ have to be done exactly, in view of the fact that this angle may take on all values in the range $-\pi$, $+\pi$, still yielding

infinitesimal rotations. Some details of the calculation and the required definite integrals are given in the appendix. We present here the results for the overlap and Hamiltonian matrices to lowest order in $1/\langle J_k^2\rangle$, i.e., the formulae will give accuracy to 2 or 4 decimal places depending on whether one is dealing with nuclei or molecules.

The diagonal and off-diagonal elements of the overlap matrix, to lowest order in $1/\langle J_k^2\rangle$, are given by

$$\Delta^J_{KK} \cong 1 - \frac{1}{4}\left(\frac{1}{\langle J_x^2\rangle}+\frac{1}{\langle J_y^2\rangle}\right)[J(J+1)-K^2]-\frac{K^2}{2\langle J_z^2\rangle}, \tag{39}$$

$$\Delta^J_{KL} \cong \left(\frac{1}{4}\right)^{K-L}\left[\binom{J+K}{J+L}\binom{J-L}{J-K}\right]^{1/2}\frac{(K-L)!}{[\frac{1}{2}(K-L)]!}\left(\frac{1}{\langle J_x^2\rangle}-\frac{1}{\langle J_y^2\rangle}\right)^{(K-L)/2}, \tag{40}$$

for $K>L$ and $K-L$ even. The matrix elements corresponding to odd $K-L$ values are zero, such that a decoupling of odd and even K occurs in the overlap matrix. The above expressions have a completely specified dependence upon the quantum numbers J, K and L, and further contain the eigenvalues of the angular momentum fluctuation tensor, i.e. the $\langle J_k^2\rangle$. Clearly the deviation from orthonormality decreases with increasing values of the $\langle J_k^2\rangle$ but also with increasing $K-L$. These features are due to the combined effect of the GOA and the asymptotic behaviour of the $d^J_{KL}(\theta)$ which cause the overlap matrix to have a peaked structure along the diagonal.

Combination of the GOA and QA gives an approximate form of the Hamiltonian matrix H^J_{KL}. A computationally convenient notation is

$$H^J_{KL} \cong E(0)\Delta^J_{KL} - \tfrac{1}{2}(A^J_{KL}+2B^J_{KL}+C^J_{KL}) \tag{41}$$

where the various contributions A^J_{KL}, B^J_{KL} and C^J_{KL} are obtained by integrating the alphabetically corresponding term in the QA (32) with the reduced GOA (30). The calculation is trivial to perform, but does not yield simple and physically meaningful expressions for the H^J_{KL} even if only the lowest-order terms in $1/\langle J_k^2\rangle$ are retained. The purpose of the next section is to clarify this situation.

However, by grouping the lowest-order contributions to the diagonal elements of the Hamiltonian matrix, it is already apparent that the rotational energies can best be measured relative to a 'zero-point energy' given by

$$E_0 = E(0) - \Delta E, \qquad \Delta E = \frac{1}{2}\sum_k \frac{\langle [H-E(0)]J_k^2\rangle}{\langle J_k^2\rangle}. \tag{42}$$

The contribution ΔE is the analogue of the energy gain obtained in GC descriptions of collective vibrations (Griffin 1957, Brink and Weiguny 1968). In general, one can say that a lowering of the intrinsic energy is obtained by virtue of the variational treatment of the collective motion considered. Clearly ΔE lowers $E(0)$ if the energy of the functions $J_k\chi$ is higher than the intrinsic energy. This is a plausible subsidiary condition on the intrinsic wavefunction. An analogous validity criterion appears in the above-mentioned GC applications.

6. Energy spectra and moments of inertia

A physical interpretation of the approximate projection procedure becomes possible if the secular equation (10) is put in a form which closely resembles the matrix equation resulting from diagonalisation of a rigid rotor Hamiltonian in a basis of Wigner

D-functions. These functions are orthonormal, so that the failure at the stage we arrived at in § 5 is probably due to orthonormality deviations in the overlap matrix. This is a common situation in GC methods where physical interpretation is attempted only after transformation to an orthonormal representation. We refer here to the so-called 'narrowing procedure' introduced by Griffin and Wheeler (1957). The analogue in the present case is the symmetric orthonormalisation of the rotational eigenvalue problem, i.e. (10) is replaced by

$$\sum_L R^J_{KL} d^J_L = E^J d^J_K, \tag{43}$$

$$R = \Delta^{-1/2}(H - E_0\Delta)\Delta^{-1/2}, \qquad d = \Delta^{1/2} c, \tag{44}$$

where $\Delta^{1/2}$ and $\Delta^{-1/2}$ are the square root and inverse square root of the overlap matrix. We have subtracted the zero-point energy so as to obtain 'pure rotational terms'. The fact that symmetric orthonormalisation appears as the key to physical interpretation is not surprising. Indeed this technique has turned up in a variety of interesting problems (Eckart 1935, Löwdin 1970, Louck and Galbraith 1976, Jørgensen 1978). In general, $\Delta^{-1/2}$ has to be computed numerically. However, since the non-orthonormality is small in the case of strong deformation, we can put

$$\Delta = 1 - S \qquad \text{with} \quad S^J_{KL} \ll 1 \tag{45}$$

and use the well known series expansion (Löwdin 1970)

$$\Delta^{-1/2} = 1 + \tfrac{1}{2}S + \tfrac{3}{8}S^2 + \ldots \tag{46}$$

At the level of approximation we are working at we may truncate (46) after the second term and obtain the transformed Hamiltonian matrix in the form

$$R \cong (1 + \tfrac{1}{2}S)H(1 + \tfrac{1}{2}S) \cong H + \tfrac{1}{2}[H, S]_+. \tag{47}$$

The previous, non-interpretable results for the overlap and Hamiltonian matrix can now be combined to evaluate the symmetrically orthonormalised Hamiltonian matrix elements R^J_{KL}. Due to a cancellation of terms, the final results are considerably more simple than the ones before orthonormalisation. We obtain to lowest order

$$R^J_{KK} = \frac{1}{4}\left(\frac{\langle [H - E(0)]J_x^2\rangle}{\langle J_x^2\rangle^2} + \frac{\langle [H - E(0)]J_y^2\rangle}{\langle J_y^2\rangle^2}\right)[J(J+1) - K^2] + \frac{1}{2}\left(\frac{\langle [H - E(0)]J_z^2\rangle}{\langle J_z^2\rangle^2}\right)K^2, \tag{48}$$

$$R^J_{KK-1} = \frac{1}{4}\left(\frac{\langle [H - E(0)]J_xJ_z\rangle}{\langle J_x^2\rangle\langle J_z^2\rangle} + \mathrm{i}\frac{\langle [H - E(0)]J_yJ_z\rangle}{\langle J_y^2\rangle\langle J_z^2\rangle}\right)(2K-1)[(J+K)(J-K+1)]^{1/2}, \tag{49}$$

$$R^J_{KK-2} = \frac{1}{8}\left(\frac{\langle [H - E(0)]J_x^2\rangle}{\langle J_x^2\rangle^2} - \frac{\langle [H - E(0)]J_y^2\rangle}{\langle J_y^2\rangle^2} + 2\mathrm{i}\frac{\langle [H - E(0)]J_xJ_y\rangle}{\langle J_x^2\rangle\langle J_y^2\rangle}\right)$$
$$\times [(J+K)(J+K-1)(J-K+2)(J-K+1)]^{1/2}. \tag{50}$$

These expressions are also obtained if one sets up the matrix of a rigid rotor Hamiltonian in a basis of Wigner D functions. More precisely, the diagonalisation of the matrix R is equivalent to solving the eigenvalue problem for the operator

$$\mathscr{H}_{\text{rotor}} = \tfrac{1}{2}\sum_{kl} \mathscr{J}_k Q^{-1}_{kl} \mathscr{J}_l \tag{51}$$

$$Q_{kl}^{-1} = \frac{\langle [H - E(0)] J_k J_l \rangle}{\langle J_k^2 \rangle \langle J_l^2 \rangle} = \frac{K_{kl}}{F_{kk} F_{ll}}. \tag{52}$$

Here the $\mathcal{J}_k$ are arbitrary, body-fixed angular momentum components and Q^{-1} plays the role of a reciprocal inertia tensor. We can therefore conclude that the spectra, resulting from angular momentum projection as described in the previous sections, are rigid rotor energy level patterns. Alternatively, one may look upon $\mathcal{H}_{\text{rotor}}$ as an effective Hamiltonian. Indeed, (51) and (52) demonstrate that the many-body system described by $\chi(x)$ behaves effectively as a rotating rigid body whose moments of inertia are the eigenvalues of the inertia tensor

$$Q_{kl} = F_{kk} K_{kl}^{-1} F_{ll}. \tag{53}$$

It should be observed that the definitions (52) and (53) are special in the sense that they depend upon the diagonal form of the angular momentum fluctuation tensor. However, it is easily verified that the general formulae read

$$Q^{-1} = F^{-1} K F^{-1} \qquad \text{and} \qquad Q = F K^{-1} F. \tag{54}$$

These are valid independent of any special orientation of the intrinsic state and will yield the correct moments of inertia upon diagonalisation.

7. Discussion

Clearly the most relevant quantity introduced in the present paper is the inertia tensor defined at the end of § 6. In order to distinguish between the inertia tensor I of a classical system of point masses and the present one, we will refer to Q as the *quantal inertia tensor.* This terminology seems appropriate since the eigenvalues of Q are the moments of inertia as obtained via angular momentum projection. Also, Q may be termed quantal since its definition involves the intrinsic wavefunction, the full quantum mechanical Hamiltonian and the quantum form of the total angular momentum components. Although there does not seem to be any obvious connection between the classical and quantal forms of the inertia tensor it is safe to say that the more rigid the system is, the more the two will look alike. Thus, for a quasi rigid molecule, in which the nuclei are confined to the immediate vicinity of their equilibrium positions, we expect $Q_{kl} \cong I_{kl}$. On the other hand, if there is no preferred particle configuration I becomes superfluous whereas the quantal inertia tensor is still able to describe the rotational collective features of the system.

It is clear from the above calculations that of the constituents, F and K, of the quantal inertia tensor only one can be brought into diagonal form by a reorientation of the original intrinsic state. Therefore, in general, Q will not appear in diagonal form. There are, however, situations in which simplifications arise. These occur when the intrinsic state is invariant under the symmetry group of the rigid rotor in the sense that

$$\mathcal{R}(\pi, \boldsymbol{e}_k)\chi(x) = r_k \chi(x) \tag{55}$$

where $r_k = \pm 1$. This implies that both F and K are diagonal and that the moments of inertia are given by the formula

$$I_k = \langle J_k^2 \rangle^2 / \langle [H - E(0)] J_k^2 \rangle. \tag{56}$$

This is a generalisation of the Peierls–Yoccoz (1957) result for axially symmetric systems. In the asymmetric case a Peierls–Yoccoz-type expression is obtained for each spatial direction. If the intrinsic state has an exact or near D_2 symmetry, i.e. if (55) is satisfied exactly or approximately, contributions from rotations near $\mathcal{R}(\pi, e_k)$ should be taken into account while evaluating the overlap and Hamiltonian matrices. These give rise to energy shifts of the same nature as the odd–even shifts studied by Verhaar (1963) in the axially symmetric case. The explicit derivation of these terms are reserved for future publications.

We have here considered angular momentum projection from a fixed intrinsic state. This means we have not taken into account the interaction between rotational and other collective degrees of freedom. For molecules this implies neglecting rotation–vibration coupling, whereas in the nuclear case this situation is equivalent to the Davydov and Filippov (1958) approximation. However, it seems perfectly possible to build a theory including the interaction between overall rotations and, say, molecular vibrations or nuclear surface oscillations if the latter are described using a GC type of approach.

From a computational point of view the present procedure can be looked upon as a step in what is sometimes referred to as a double variation method. Indeed, if initially a set of parameters (linear, nonlinear, GC, ...) in $\chi(x)$ have been optimised, these will no longer be optimal after the angular momentum projection is performed. One can hence start a second cycle of calculations by readjusting the intrinsic parameters, keeping the angular momentum superposition coefficients constant. In this way one alternately treats the internal dynamics and the collective rotations of the system. A consensus between these two types of motion is reached if the above cycle is carried through to self-consistency. Approximate angular momentum projection economises this scheme since the rotational part of the problem can be performed at the mere cost of evaluating the quantal moment of inertia tensor. In fact, one can say that the combination of self-consistent field type calculations with the present scheme seems especially suited to produce high quality wavefunctions.

Finally, we consider the possible didactical merits of the projection technique. We believe this to be of some importance because the classical way to arrive at comparable results, i.e. by using frame transformations, leads to considerable complications and sources of confusion. These are due to the fact that a separation of the Hamiltonian into a rigid rotor, an internal part and coupling terms necessitates a definition of the rotating frame and internal coordinates in terms of particle coordinates. If a choice has been made, the separation can be achieved only by explicitly transforming the operators in H to the internal frame. This involves generalised coordinate transformations which are to be treated carefully upon quantisation, and inevitably leads to the introduction of body-fixed angular momentum components. The latter invariably cause confusion when first encountered because of their anomalous commutation relations (for a review see Herold and Ruder 1979, Herold *et al* 1980). A historical example of these problems is to be found in the molecular physics literature concerned with vibration–rotation Hamiltonians on the basis of the Eckart frame (for a review see Sutcliffe 1980). The present approach using the projection formalism is free of the above drawbacks. Indeed, due to the use of representation theory our methods are strictly algebraic in nature, in contrast to frame transformation theory which requires sophisticated operator techniques and careful consideration of quantisation rules. The technical prerequisites needed to derive the results of this paper are limited to properties of the rotation group and its irreducible representations. Although it

remains a question of taste which procedure to use for teaching purposes, we believe the projection approach to have some didactical advantages.

8. Conclusions

We have presented an approximate angular momentum projection technique for many-body systems with an intrinsic asymmetric deformation. Two steps in the derivation of the results were of crucial importance. The first was the introduction of a new parametrisation of the rotation group. The second involved the use of symmetric orthonormalisation in order to permit a physical interpretation of the rotational secular equations. We were then able to conclude that, at the lowest level of approximation, the angular momentum projection technique yields a rigid rotor spectrum characterised by quantum mechanically defined moments of inertia.

Acknowledgments

We would like to thank Professors M Bouten and P Van Leuven for their continued interest during the course of this work. Financial support from the National Fonds voor Wetenschappelijk Onderzoek, Belgium, the National Science Foundation, USA and the Nato Scientific Affairs Division under grant 1776 are gratefully acknowledged.

Appendix

The analytical evaluation of the integrals over the parameter domain $-\pi/2 \leqslant \varepsilon \leqslant +\pi/2$, $0 \leqslant \theta \leqslant \pi$ and $-\pi \leqslant \delta \leqslant +\pi$ does not present any problems if the asymptotic expansions (38) are used and the extensions of the ε and θ ranges are made. One can always factorise the three-dimensional integrals into two-dimensional ones involving θ and δ and one-dimensional ε integrations. The latter are trivial and involve use of the following results:

$$\int_{-\infty}^{+\infty} \mathrm{d}x \, \exp(-cx^2 + \mathrm{i}bx) = \left(\frac{\pi}{c}\right)^{1/2} \exp\left(\frac{-b^2}{4c^2}\right),$$

$$\int_{-\infty}^{+\infty} \mathrm{d}x \, \exp(-cx^2 + \mathrm{i}bx)x = \mathrm{i}\left(\frac{\pi}{c}\right)^{1/2} \frac{b}{2c} \exp\left(\frac{-b^2}{4c^2}\right),$$

$$\int_{-\infty}^{+\infty} \mathrm{d}x \, \exp(-cx^2 + \mathrm{i}bx)x^2 = \left(\frac{\pi}{c}\right)^{1/2} \left(\frac{1}{2c} - \frac{b^2}{4c^2}\right) \exp\left(\frac{-b^2}{4c^2}\right).$$

The remaining integrations over θ and δ can be performed exactly if one starts with the δ integrals and takes into account the definition of the Bessel functions of imaginary argument (Abramowitz and Stegun 1965):

$$I_m(z) = \frac{1}{\pi} \int_0^{\pi} \mathrm{d}\Phi \, \mathrm{e}^{z \cos \Phi} \cos m\Phi.$$

In the present case the argument z turns out to be proportional to θ^2. The resulting

θ integrals reduce to expressions of the form (Gradshteyn and Ryzhik 1965)

$$\int_0^\infty \mathrm{d}z\, \exp[-zt(t^2-1)^{-1/2}]I_m(z)z^n = \Gamma(m+n+1)(t^2-1)^{(n+1)/2}P_n^{-m}(t)$$

where the $P_n^m(t)$ are the associated Legendre polynomials. The calculation to lowest order involves indices m and n which do not exceed 2 and are therefore of a simple analytical form:

$$P_0^0(t)=1$$

$$P_1^0(t)=t, \qquad P_1^{-1}(t)=\tfrac{1}{2}(t^2-1)^{1/2},$$

$$P_0^2(t)=\tfrac{1}{2}(3t^2-1), \qquad P_2^{-1}(t)=\tfrac{1}{2}t(t^2-1), \qquad P_2^{-2}(t)=\tfrac{1}{8}(t^2-1).$$

We left out a common constant in such a way that the zeroth-order contribution to Δ_{KK}^J equals one.

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