Spectroscopy with Giant Trinuclear Molecules

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Recent experimental investigations are pointing to the existence of a new type of nuclear phenomenon which consists in the formation of a long living molecule during the cold fragmentation of ^{252}Cf. For such long times it is possible that the nuclear molecules rotate and vibrate like atomic molecules making thus possible the study of a new type of spectroscopy. We present in this paper the basic ideas which are leading to the calculation of rotational and vibrational bands of three-clusters molecules.

1. Introduction

Among the various phenomena occurring in Nuclear Physics at low energy, the spontaneous clustering of heavy nuclei in two or more lighter nuclei, represents a remarkable example of quantum phenomena without analogue in classical mechanics. For certain combinations of nuclei a local equilibrium position between repulsive and attractive nuclear and Coulomb will show-up. A nuclear molecule is a system consisting of two or more nuclei bound together on their surfaces in a quasi-bound potential. In the past such exotic nuclear structures were produced in close collisions of ^{12}C by Bromley.[1] Molecular states provided experimental evidence that interacting nuclei were retaining their identity during collisions, thus forming an effective bond which can rotate and vibrate like atomic molecules before flying apart or coalescing into a fused nucleus. It was also supposed that long-living shape isomeric states might be considered also as nuclear molecules due to the pockets in the deformation energies which are developing at large elongations. Also the new type of radioactivity consisting in the emission of heavy nuclei, such as ^{14}C, ^{24}Ne, ^{28,30}Mg, and ^{32}Si predicted by Săndulescu et al.[2] and experimentally discovered by Rose and Jones in 1984[3] can be viewed as an example of nuclear molecules occurring in the fission process. In view of the similarity of the above mentioned phenomena to the cold fission process we undertook the project to study the neutronless fragmentation of ^{252}Cf in two or three fragments by means of a molecular scenario.[4]

Nuclear cold fission is a rare phenomenon consisting in the disintegration of a large nucleus, such as ^{252}Cf, in two or more fragments with a very small dissipation of energy on degrees of freedom, other than the translational motion. Before scission takes place, and after preformation from the mother nucleus is accomplished, there is a transient stage when the clusters are in close vicinity.

In last years the cold fission of ^{252}Cf has been intensively studied in U.S. using the Gammasphere.[5] In the case of the ternary cold fission, when a light cluster is accompanying the ^{10}Be emission, the triple-γ coincidence data contains non-Doppler broadened high energy peaks in coincidence with one-ray in each fragment accompanying ^{10}Be. These peaks are shifted by -6.1 to -26 keV from the 3368 keV energy of the 2_1^+–0^+ transition, a fact which supports the existence of a long-lived nuclear molecule where the three nuclei stick together for a time larger than 1 ps. In such circumstances this exotic quantum system is free to rotate and vibrate like in atomic molecules. In principle the γ rays coming from the deexcitation of these molecular states could be observed in coincidence with the γ rays of the individual nuclei mentioned above, and therefore could provide a conclusive evidence in the search of Giant Trinuclear Molecules.

In what follows we want to review our later works on the collective modes of quasi-molecules occurring in ternary cold fission. We present first the basic ingredients and results for a geometrical model aimed to describe the molecular spectrum of a system composed of an α cluster and two heavy fragments in triangular and linear configuration. Then we comment on the usage of algebraic models for three-cluster systems.

2. Geometrical Approach

As we mentioned above a repulsive core will develop in the nucleus-nucleus interaction at large overlaps. This will determine a typical molecular minima, provided that at least one of them has a non-negligible deformation. *Mutatis mutandis*, the three-body potential, assumed to be the sum of all two-body components,

$$V = V_{12} + V_{13} + V_{23} \ , \tag{1}$$

displays a similar quasimolecular pattern with two minima in the equatorial region and two at the poles of the system. Due to the axial symmetry, the minima in the equatorial region are equivalent, and in fact one could speak about a ring which represents the geometrical locus of the points where the three-body potential attains an absolute minimum. In the case of α or ^{10}Be-like quasi-molecules such minima are formed in all two-body channels.

Very recently we proposed a geometric quantum approach to the three-body problem which leads to the determination of the collective spectrum of a linear or a triangular trinuclear molecule in References 6, 7.

Considering three spherical clusters, whose Cartesian space coordinates are denoted by $\boldsymbol{r}_1$, $\boldsymbol{r}_2$, and $\boldsymbol{r}_3$, the Jacobi coordinates, for which the two heavier clusters 1 and 2 appear explicitly as a subsystem, are introduced by means of the following transformations:

$$\begin{aligned} \boldsymbol{\rho} &= \boldsymbol{r}_2 - \boldsymbol{r}_1 \ , \\ \boldsymbol{\lambda} &= \frac{m_1\boldsymbol{r}_1 + m_2\boldsymbol{r}_2}{m_1+m_2} - \boldsymbol{r}_3 \ , \\ \boldsymbol{R}_{\text{c.m.}} &= \frac{m_1\boldsymbol{r}_1 + m_2\boldsymbol{r}_2 + m_3\boldsymbol{r}_3}{m_1+m_2+m_3} \ . \end{aligned} \tag{2}$$

Choosing for $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ the following ansatz

$$\begin{aligned} \boldsymbol{\rho} &= (\rho\sin a\gamma,\ 0,\ \rho\cos a\gamma) \ , \\ \boldsymbol{\lambda} &= (-\lambda\sin(1-a)\gamma,\ 0,\ \lambda\cos(1-a)\gamma) \ , \end{aligned} \tag{3}$$

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where for triangular configurations

$$a = \frac{\mu_{(12)3}\lambda^2}{\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2} ,$$

whereas for linear configurations

$$a = \frac{1}{2\gamma}\,\mathrm{atan}\left(\frac{\mu_{(12)3}\lambda^2 \sin 2\gamma}{\mu_{12}\rho^2 + \mu_{(12)3}\cos 2\gamma}\right).$$

Considering the case $^{132}\mathrm{Sn} + \alpha + {}^{116}\mathrm{Pd}$ we get $a \approx 1/300$ at the minimum so that we can approximate that γ consists of small deviations from $\pi/2$, i.e. we perform the change of variable $\gamma = \pi/2 - \varepsilon$. We then obtain the following forms for the quantum kinetic energy in the triangular

$$\begin{aligned}\widehat{T} \;=\; & -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} \\ & - \frac{\hbar^2}{2\mu_{(12)3}\lambda^2}\frac{\partial^2}{\partial\varepsilon^2} + \frac{\hbar^2}{2\mu_{12}\rho^2}(\boldsymbol{L}^2 - L_3'^2) \\ & + \frac{\hbar^2}{2\mu_{(12)3}\lambda^2}L_3'^2 - \frac{\hbar^2\varepsilon}{\mu_{12}\rho^2}L_1'L_3' \end{aligned} \tag{4}$$

and linear configurations

$$\begin{aligned}\widehat{T} \;=\; & -\frac{\hbar^2}{2\mu_{12}}\frac{\partial^2}{\partial\rho^2} - \frac{\hbar^2}{2\mu_{(12)3}}\frac{\partial^2}{\partial\lambda^2} \\ & - \frac{\hbar^2}{2}\left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2}\right)\frac{\partial^2}{\partial\varepsilon^2} \\ & + \frac{\hbar^2}{2(\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2)}(\boldsymbol{L}^2 - L_3'^2) \\ & + \frac{\hbar^2}{2\varepsilon^2}\left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2}\right)\left(L_3'^2 - \frac{1}{4}\right). \end{aligned} \tag{5}$$

Before going further we have to make some considerations about energy. In both cases, linear and triangular, we have to stick on some absolute minimum of the potential energy. Therefore we can expand the potential around these triangular and linear minima. We considered expansions up to quadratic terms. Using the above mentioned approximations we get the following expression for the fluctuating part of the potential energy in the triangular

$$\begin{aligned}\widehat{V} \;\approx\; & \frac{1}{2}C_\rho\delta\rho^2 + \frac{1}{2}C_\lambda\delta\lambda^2 + \frac{1}{2}C_\lambda\lambda^2\varepsilon^2 \\ & + C_{\rho\lambda}(\delta\rho\,\delta\lambda\sin\varepsilon - \lambda\delta\rho\varepsilon\cos\varepsilon), \end{aligned} \tag{6}$$

and linear configurations

$$\widehat{V} = \frac{1}{2}C_\rho\delta\rho^2 + \frac{1}{2}C_\lambda\delta\lambda^2 + \frac{1}{2}C_\lambda\lambda_0^2\varepsilon^2 + \frac{1}{2}C_{\rho\lambda}\delta\rho\,\delta\lambda. \tag{7}$$

As one can see the kinetic and potential energy are containing couplings between the different vibrational (specified by the observables ρ, λ, and ε) and rotational (specified by the angular momentum) modes in a non-trivial way. Under the assumption that near the minima's positions the displacements $\delta\rho$, $\delta\lambda$, and ε are not large with respect to the equilibrium values $y_0 = \rho_0, \lambda_0$ and $\varepsilon_0 = 0$, i.e. $y = y_0 + \delta y$, with $\delta y \ll 1$ one can expand in Taylor series all coordinate functions of the kinetic energy and potential energy operators. Restricting to the zeroth-order approximation we obtain the rotational-vibrational spectrum for the triangular case

$$\begin{aligned}E^{(0)}_{IKn_\rho n\varepsilon n_\lambda} \;=\; & \hbar\tilde{\omega}_\rho(n_\rho + \frac{1}{2}) + \hbar\tilde{\omega}_\varepsilon(n_\varepsilon + \frac{1}{2}) + \hbar\omega_\lambda(n_\lambda + \frac{1}{2}) \\ & + \frac{\hbar^2}{2\mu_{12}\rho_0^2}\left[I(I+1) - K^2\right] + \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^2}K^2 \end{aligned} \tag{8}$$

the frequencies being defined as follows

$$\omega_\rho = \sqrt{\frac{C_\rho}{\mu_{12}}}\,, \qquad \omega_\lambda = \omega_\varepsilon = \sqrt{\frac{C_\lambda}{\mu_{(12)3}}}\,, \tag{9}$$

$$\tilde{\omega}^2_{\rho(\varepsilon)} = \frac{1}{2}\left(\omega_\rho^2 + \omega_\varepsilon^2 \pm (\omega_\rho^2 - \omega_\varepsilon^2)\sec 2\eta\right) \tag{10}$$

provided $\tilde{\omega}_\rho > \tilde{\omega}_\varepsilon$. For the linear case we have

$$\begin{aligned}E^{(0)}_{IKn_\rho n\varepsilon n_\lambda} \;=\; & \hbar\tilde{\omega}_\rho(n_\rho + \frac{1}{2}) + \hbar\tilde{\omega}_\lambda(n_\lambda + \frac{1}{2}) \\ & + \hbar\omega_\varepsilon\left(|K| + n_\varepsilon + \frac{3}{2}\right) \\ & + \frac{\hbar^2}{2(\mu_{12}\rho_0^2 + \mu_{(12)3}\lambda_0^2)}\left[I(I+1) - K^2\right] \end{aligned} \tag{11}$$

where the frequencies are defined as follows

$$\tilde{\omega}^2_{\rho(\lambda)} = \frac{1}{2}\left(\omega_\rho^2 + \omega_\lambda^2 \pm (\omega_\rho^2 - \omega_\lambda^2)\sec 2\eta\right) \tag{12}$$

in case $\tilde{\omega}_\rho > \tilde{\omega}_\lambda$ and

$$\omega_\varepsilon = \lambda_0\sqrt{C_\lambda\left(\frac{1}{\mu_{(12)3}\lambda^2} + \frac{1}{\mu_{12}\rho^2}\right)}\,. \tag{13}$$

The parameter η is defined as follows:

$$\begin{aligned}\tan 2\eta \;&=\; -\frac{2C_{\rho\lambda}}{\sqrt{\mu_{12}\mu_{(12)3}}(\omega_\rho^2 - \omega_\varepsilon^2)}\,, \;\; \text{triangular,} \\ \tan 2\eta \;&=\; -\frac{2C_{\rho\lambda}}{\sqrt{\mu_{12}\mu_{(12)3}}(\omega_\rho^2 - \omega_\lambda^2)}\,, \;\; \text{linear.} \end{aligned} \tag{14}$$

Due to the fact that all three clusters have different masses the axial symmetry is broken when the three clusters are laying in a triangular configuration. Then, for the $K = 0$ bands, positive and negative parity states are alternating. For $K \neq 0$ there is no selection rule and for a given angular momentum there is a *parity doublet*, starting at $I = K$ and increasing in steps of one:

$$K \neq 0: \quad K^\pm,\ (K+1)^\pm,\ (K+2)^\pm, \ldots, . \tag{15}$$

Thus, for K= even(odd) positive parity states have spin even(odd) while negative parity states have spin odd(even).

For rotational ground state band of the triangular quasi-molecule the excited state 1_1^- is at 5.4 keV, and the 2_1^+ state at 16.8 keV. The first state of the $K = 1$ band is at 213 keV whereas the 2^+ state of the $K = 2$ band is at 836 keV. The band head ($n_\rho = 1$, $n_\lambda = n_\varepsilon = 0$) is located at 3.85 MeV, a state which in principle can be reached in cold fission.

The rotational spectrum of the linear molecule is approximately two times more compressed than the one corresponding to the triangular configuration, whereas the vibrational band-heads are located at almost the same energies as in the triangular case.

3. Algebraic Approach

Recently inspired from a previous work on baryon structure,[8] we proposed an algebraic model which has the advantage to describe complicated systems, which would require complex procedures in the geometrical model.[9,10] Like in the previous section, we consider spherical clusters and thus the number of degrees of freedom are six and for each relative coordinate we can introduce boson creation and annihilation operators, carrying negative parity. The basic concept of the U(7) model is to introduce a cutoff through the addition of an *s*-boson of positive parity. With this the spherical components of the creation operators are given by

$$p^\dagger_{\rho,m}, \quad p^\dagger_{\lambda,m}, \quad s^\dagger \quad (m = -1, 0, 1). \tag{16}$$

The total number of bosons $N = n_\rho + n_\lambda + n_s$ is conserved, which implies that the total number of p-bosons is restricted between zero and N. Taking all possible double products of a creation with an annihilation operator, we obtain the algebra u(7) with its 49 generators. The basis states are given by

$$| N, (n_\rho, L_\rho), (n_\lambda, L_\lambda); LM_L \rangle \,, \tag{17}$$

with n_ρ and n_λ the number operator of the ρ- and λ-oscillation quanta respectively. The L_ρ and L_λ are the angular momenta of the ρ and λ part, L is the total angular momentum and M_L its projection.

As the model Hamiltonian we use

$$\begin{aligned} H \;=\; & a_L \boldsymbol{L}^2 + A P_1^\dagger P_1 + C P_2^\dagger P_2 + C P_3^\dagger P_3 + D(P_1^\dagger P_2 + P_2^\dagger P_1) \\ & + E(P_1^\dagger P_3 + P_3^\dagger P_1) + F(P_2^\dagger P_3 + P_3^\dagger P_2) \,. \end{aligned} \tag{18}$$

The operators P_i are defined via

$$\begin{aligned} P_1^\dagger \;&=\; p_\rho^\dagger \cdot p_\rho + p_\lambda^\dagger \cdot p_\lambda - R_0^2 s^\dagger s^\dagger \,, \\ P_2^\dagger \;&=\; \sin^2\beta_0 \; p_\lambda^\dagger \cdot p_\lambda - \cos^2\beta_0 \; p_\rho^\dagger \cdot p_\rho \,, \\ P_3^\dagger \;&=\; \sin(2\beta_0) p_\rho^\dagger \cdot p_\lambda - \cos\gamma_0 (\sin^2\beta_0 \; p_\lambda^\dagger \cdot p_\lambda \\ & \quad + \cos^2\beta_0 \; p_\rho^\dagger \cdot p_\rho) \,, \end{aligned} \tag{19}$$

where the parameter $R_0 = \sqrt{\boldsymbol{\rho}_0 \cdot \boldsymbol{\rho}_0 + \boldsymbol{\lambda}_0 \cdot \boldsymbol{\lambda}_0}$ describes the extension of the system.

The parameter β_0 is defined by the relative size of $\vec{\rho}$ and $\vec{\lambda}$, i.e. $\lambda_0 = R_0 \cos\beta_0$ and $\rho_0 = R_0 \sin\beta_0$, and γ_0 gives the angle between the two vectors. Next, the following coherent state is introduced

$$| N R_0, \beta_0 \gamma_0 \rangle = \frac{1}{\sqrt{N!}} (b_c^\dagger)^N \mid 0 \rangle \,,$$

$$b_c^\dagger = \frac{\left[s^\dagger + R_0 \cos\beta_0 p_{\lambda,x}^\dagger + R_0 \sin\beta_0 (\cos\gamma_0 p_{\rho,x}^\dagger + \sin\gamma_0 p_{\rho,y}^\dagger) \right]}{\sqrt{1 + R_0^2}}, \tag{20}$$

where $b_c^\dagger$ is called the *collective boson*. In order to describe the motion around the equilibrium position, we introduce fluctuation bosons orthogonal to $b_c^\dagger$. These are $b_u^\dagger$, describing the breathing mode, $b_v^\dagger$, the butterfly mode, and $b_w^\dagger$ is the mode where the angle γ between the vectors $\boldsymbol{\rho}$ and $\boldsymbol{\lambda}$ is changing (shearing mode). A Bogoliubov treatment is applied, where the $b_c^\dagger$ and b_c are substituted by $\sqrt{N}$ and only leading terms in N are taken into account. The obtained Hamiltonian has the form

$$H_B = \sum_{\alpha_1, \alpha_2} \varepsilon_{\alpha_1 \alpha_2} b_{\alpha_1}^\dagger b_{\alpha_2} \tag{21}$$

with $\alpha_k = u, v, w$. Writing in the second quantization the kinetic and potential energy from the previous section, we were able to give explicit expressions to these frequencies.[10]

Like in the geometrical model we obtain that the ground state band is severely compressed with a distance of two subsequent states of the order of keV. The vibrational modes are of the order of MeV, which indicates a strong separation between rotational and vibrational modes as in the atomic molecules.

4. Conclusions and Perspectives

Based on potential energy considerations we analyzed the molecular collective spectrum of giant trinuclear molecules when all three clusters are spherical. Applications have been considered for the three-body cold breakup of ^{252}Cf when the light cluster, an α particle, is sandwiched between the heavier clusters (linear scenario) or lies above the line connecting the two heavier nuclei (triangular scenario).

We showed that the rotational bands are strongly compressed, the distance between states within a level being of the order of a few keV. The vibrational states are of the order of MeV, implying a strong separation between rotational and vibrational modes.

The compression of the rotational modes and the parity doublets can be used as a signature to look for the formation of three-cluster nuclear molecules. If one observes a transition line from a vibrational mode to, e.g., the ground state band, then it should split into a couple of lines (taking into account spin-selection rules) only a few keV apart, demonstrating the large extension of the system. With the observation of parity doublets, this will indicate the formation of the nuclear molecule.

Additionally, the γ spectroscopy for heavy nuclear molecules is feasible due to the fact that the deexcitation times of the rotational states are $\leq 10^{-19}$ s which are very small compared to the calculated lifetimes of α-accompanied cold ternary fission which range in the interval 10^{-12}–10^{-15} s.

The next step is to introduce the deformation. This has been already done for the linear case in Reference 6. However in the triangular case the geometric Hamiltonian gets very complicated, especially the rotational part describing the relative motion of the center of masses. This is the reason to look to algebraic models based on group theory. Recently we proposed an algebraic model for dinuclear molecules (hypothetically encountered in binary cold fission) which includes also the internal structure of the two clusters.[11] In this model each cluster is described by the Elliott's SU(3) model and the relative motion of the two clusters is given by the Iachello's U(4) model in the dynamical U(3) dynamical chain. Once this project will be completed its extension to the three-cluster case will be straightforward.

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