

THE ROTATIONAL-VIBRATIONAL MODEL OF TRINUCLEAR MOLECULES

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ABSTRACT

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. I present in this paper the basic ideas which are leading to the calculation of rotational and vibrational bands of three-clusters molecules.

keywords: cluster decay, nuclear molecules, rotation-vibration model

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.

Before defining what a nuclear molecule is one have to understand the nature of forces acting between nuclei. Since a nucleus consists of Z positively charges it will interact with another nucleus by means of a repulsive electrostatic force which at large distances behaves like $1/R$, where R is the distance between the two nuclei. The nuclear force is more involved. When the two nuclei are weakly overlapping the nuclear force will be attractive whereas for stronger overlap there is a hard nuclear repulsive core which will prevent the melting of one nucleus into the other. Therefore, for certain combinations of nuclei a local equilibrium position between repulsive and attractive potential walls will show-up. We therefore define a nuclear molecule as 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 is important for the present overview to stress that the concept of nuclear

molecules includes also nuclear systems composed of several clusters. It was also supposed that long-living shape isomeric states might be considered also 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 , $^{28,30}\text{Mg}$ 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 which support the existence of a long-lived nuclear molecule where

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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 gamma-rays coming from the deexcitation of these molecular states could be observed in coincidence with the gamma-rays of the individual nuclei mentioned above, and therefore could provide a conclusive evidence in the search of Giant Trinuclear Molecules.

In this paper we will present a simple description of the spectroscopy of quasi-molecules occurring in ternary cold fission.

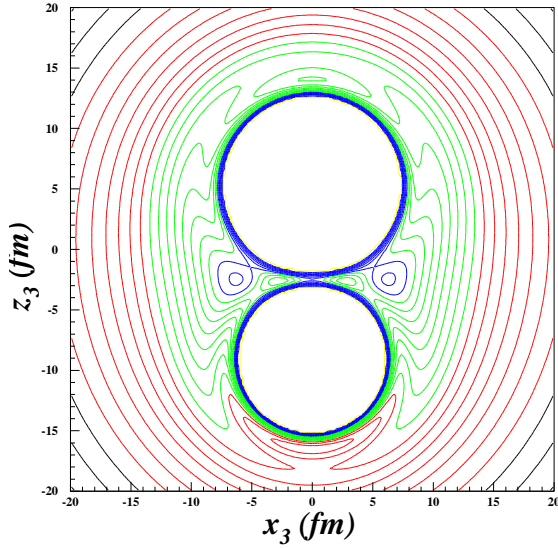


FIG. 1: Contour lines of the total three-body potential. The z_3 -axis defines the fission or molecular axis. Minima off the fission axis, at $x_3 \neq 0$, are defining triangular quasi-equilibrium configurations.

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,

$$V = V_{12} + V_{13} + V_{23} , \quad (1)$$

displays a similar quasimolecular pattern with two minima in the equatorial region and two at the poles of the system (see Fig.1). 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 -like quasi-molecules such minima are formed in all two-body channels, e.g. α - ^{92}Kr , α - ^{156}Nd and ^{156}Nd - ^{96}Kr .

The three-body problem can be handled more easily by separating out the centre-of-mass motion and introducing Jacobi coordinates. The cartesian space coordinates being denoted by r_1, r_2 and 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:

$$\begin{aligned} \vec{\rho} &= r_2 - r_1, \quad \vec{\lambda} = \frac{m_1 r_1 + m_2 r_2}{m_1 + m_2} - r_3, \\ R_{\text{c.m.}} &= \frac{m_1 r_1 + m_2 r_2 + m_3 r_3}{m_1 + m_2 + m_3} \end{aligned} \quad (2)$$

and the kinetic energy can be written

$$H = \frac{1}{2(m_1 + m_2 + m_3)} P^2 + \frac{1}{2\mu_{12}} p_\rho^2 + \frac{1}{2\mu_{(12)3}} p_\lambda^2 \quad (3)$$

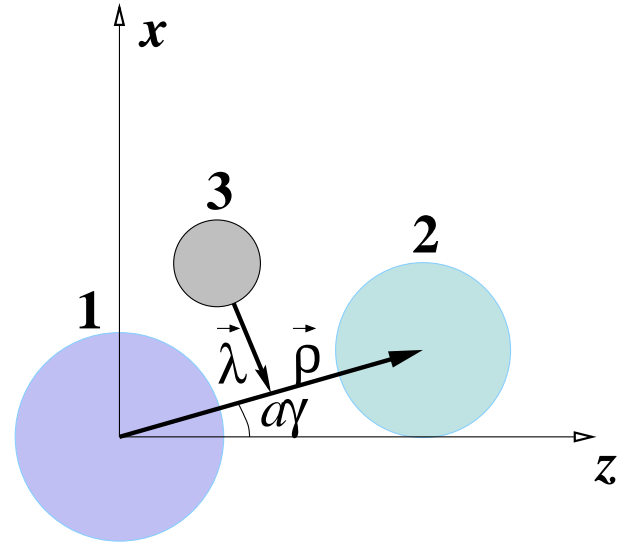


FIG. 2: The three-body problem : the vector $\vec{\rho}$ joining the heavier clusters 1 and 2 makes an angle $\alpha\gamma$ with the z -axis.

The potential was expanded up to quadratic terms around the molecular minima :

$$\begin{aligned} V_{\text{cl}} &= \frac{1}{2} \sum_{i>j=1}^3 C_{ij} (r_j - r_i)^2 = \\ &= \frac{1}{2} C_\rho \delta\rho^2 + \frac{1}{2} C_\lambda \delta\lambda^2 + C_{\rho\lambda} \delta\rho\delta\lambda \end{aligned} \quad (4)$$

Next we define the components of the vectors $\vec{\rho}$ and $\vec{\lambda}$ according to the geometry adopted in Fig.2.

$$\begin{aligned}\vec{\rho} &= (\rho \sin a\gamma, 0, \rho \cos a\gamma), \\ \vec{\lambda} &= (-\lambda \sin(1-a)\gamma, 0, \lambda \cos(1-a)\gamma)\end{aligned}\quad (5)$$

This corresponds to define the intermediate z -axis in between the two vectors. For $a=0$ the vector $\vec{\rho}$ lies along the z -axis. For reasons that will become transparent below we are not choosing $\vec{\rho}$ along the z -axis.

We next specialize our considerations to the case of the system $^{132}\text{Sn} + \alpha + ^{116}\text{Pd}$ which occurs in the α -accompanied ternary cold fission of ^{252}Cf . Picking-up for ρ and λ their values at the minimum configuration and setting $a = \frac{\mu_{(12)3}\lambda^2}{\mu_{12}\rho^2 + \mu_{(12)3}\lambda^2}$, we obtain $a \approx 1/300$. Therefore $a \ll 1$ and several terms of the above expression can be safely left out, as have been done in the previous section with some potential terms. Moreover we can approximate that γ consists of small deviations from $\pi/2$, i.e. we perform the change of variable $\gamma = \pi/2 - \varepsilon$ (this is so because we are not far from the totally symmetric case with $A_1 = A_2$). Applying the Pauli-Podolsky quantization method [6] we obtain

$$\begin{aligned}\hat{T} &= -\frac{\hbar^2}{2\mu_{12}} \frac{\partial^2}{\partial \rho^2} - \frac{\hbar^2}{2\mu_{(12)3}} \frac{\partial^2}{\partial \lambda^2} - \frac{\hbar^2}{2\mu_{(12)3}\lambda^2} \frac{\partial^2}{\partial \varepsilon^2} \\ &+ \frac{\hbar^2}{2\mu_{12}\rho^2} (L^2 - L_3^2) + \frac{\hbar^2}{2\mu_{(12)3}\lambda^2} L_3^2 - \frac{\hbar^2 \varepsilon}{\mu_{12}\rho^2} L_1 L_3\end{aligned}\quad (6)$$

As one can see this form contains couplings between the different vibrational (specified by the observables $\vec{\rho}, \vec{\lambda}$ and ε) and rotational (specified by the angular momentum L') modes in a non-trivial way. At this stage we assume that near the minimum's position the displacements $\delta\vec{\rho}, \delta\vec{\lambda}$ 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 $\delta y \ll 1$. Under this assumption one can expand in Taylor series all coordinate functions of the kinetic energy and potential energy operators

$$\hat{H} = \hat{H}^{(0)} + \sum_i \delta y_i \hat{H}_i^{(1)} + \frac{1}{2} \sum_{ij} \delta y_i \delta y_j \hat{H}_{ij}^{(2)} + \dots \quad (7)$$

Restricting the considerations to the zeroth-order approximation we get

$$\hat{H}^{(0)} = -\frac{\hbar^2}{2\mu_{12}} \frac{\partial^2}{\partial \rho^2} - \frac{\hbar^2}{2\mu_{(12)3}} \frac{\partial^2}{\partial \lambda^2} - \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^2} \frac{\partial^2}{\partial \varepsilon^2}$$

$$\begin{aligned}&+ \frac{\hbar^2}{2\mu_{12}\rho^2} L^2 + \frac{\hbar^2}{2} \left(\frac{1}{\mu_{(12)3}\lambda_0^2} - \frac{1}{\mu_{12}\rho_0^2} \right) L_3^2 \\ &+ \frac{1}{2} C_\rho \delta \rho^2 + \frac{1}{2} C_\lambda \delta \lambda^2 + \frac{1}{2} C_\lambda \lambda_0^2 \delta \varepsilon^2 \\ &- C_{\rho\lambda} \lambda_0 \delta \rho \varepsilon.\end{aligned}\quad (8)$$

Due to the degeneracy of the problem the eigenfunctions of the Hamiltonian must be constructed as linear combinations of the rotation matrices D_{MK}^I . Since L^2 and L_z^I are commuting with \hat{H}_0 only combinations of different K -values for the same I and M are occurring

$$\Psi = \sum_{K=-I}^{+I} F_K^I(\vec{\rho}, \vec{\lambda}, \gamma) D_{MK}^I(\theta). \quad (9)$$

The non-perturbed spectrum will then read

$$\begin{aligned}E_{IKn_\rho n_\lambda n_\varepsilon}^{(0)} &= \hbar \tilde{\omega}_\rho (n_\rho + \frac{1}{2}) + \hbar \tilde{\omega}_\varepsilon (n_\varepsilon + \frac{1}{2}) \\ &+ \hbar \omega_\lambda (n_\lambda + \frac{1}{2}) + \frac{\hbar^2}{2\mu_{12}\rho_0^2} [I(I+1) - K^2] \\ &+ \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^2} K^2.\end{aligned}\quad (10)$$

It is important to stress that 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*:

$$K \neq 0: \quad K^\pm, (K+1)^\pm, (K+2)^\pm, \dots, \quad (11)$$

In Fig.3 we presented the rotational states with energy smaller than 1 MeV for the g.s. band and the band with $K=1$ and 2 of the GTM $^{132}\text{Sn} + \alpha + ^{116}\text{Pd}$. The excited rotational 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 heads of the vibrational bands are laying at much higher energy, and were not plotted on this figure. For example the band head ($n_\rho = 1, n_\lambda = n_\varepsilon = 0$) is located at 3.85 MeV, a state which, however, could be reached in cold fission. We also compared on the same figure the results obtained in the nonperturbed case and taking

into account the second-order corrections

$$\begin{aligned} \delta\hat{H}^{(2)} = & \frac{\hbar^2}{2\mu_{(12)3}\lambda_0^3} \left(2 - 3\frac{\delta\lambda}{\lambda_0}\right) \delta\lambda \left(\frac{\partial^2}{\partial\varepsilon^2} - L_3'^2\right) \\ & + \left[C_\lambda\lambda_0\delta\lambda - \frac{3\hbar^2}{4\mu_{12}\rho_0^2} \left(1 + 2\frac{\mu_{(12)3}\lambda_0^2}{\mu_{12}\rho_0^2}\right) \right] \varepsilon^2 \\ & - \frac{\hbar^2}{2\mu_{12}\rho_0^2} \left[\left(2 - 3\frac{\delta\rho}{\rho_0}\right) \frac{\delta\rho}{\rho_0} (L^2 - L_3'^2 - 1) \right] \\ & + \frac{\hbar^2}{2\mu_{12}\rho_0^2} \left[2 \left(1 - 2\frac{\delta\rho}{\rho_0}\right) \varepsilon L_1' L_3' \right], \end{aligned} \quad (12)$$

and compute the spectrum in the stationary perturbation theory. It is easy to infer from here that taking only the zeroth-order Hamiltonian is a satisfactory approximation.

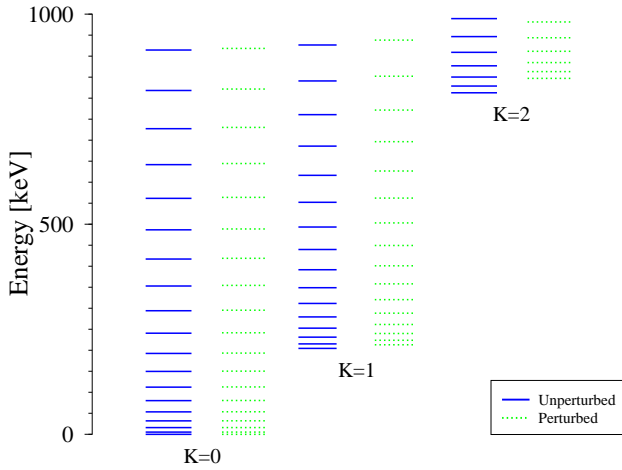


FIG. 3: The first rotational bands $K = 0, 1, 2$ in the zeroth-order approximation (full lines) and with the account of the first order perturbation (dashed lines).

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 life-times of α -accompanied cold ternary fission which range in the interval 10^{-12} s - 10^{-15} s.

The rotational bands are strongly compressed, the distance between states within a level being of the order of a few keV. For the ground state band ($K = 0$) positive and negative parity states are alternating. This is also true for excited vibrational states with $K = 0$. The positive parity states have spin even while the negative states have spin odd. The vibrational states are of order of MeV, implying a strong separation between rotational and vibrational modes. For $K \neq 0$ we obtain *parity doublets* starting with $I = K$ and increasing in steps of one.

The compression of the rotational states and the parity doublets can be used as a signature to look for the formation of three-cluster nuclear molecules [7]. 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.

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