

Nuclear Clusters and Covalently Bound Nuclear Molecules

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Light $N=Z$ nuclei show pronounced clustering for excited states close to the decay thresholds. This fact is expressed in the *Ikeda-Diagram*. In neutron-rich nuclei, low lying states close to the neutron drip-line show also pronounced clustering with α -particles and heavier closed shell clusters. For the valence neutrons a concept based on molecular orbitals can be used for their single particle wave functions. Covalent binding between α -particles due to valence neutrons and a related shallow local potential is now known to be responsible for particular structures in light neutron rich nuclei, like dimers in Be-isotopes, and trimers in C-isotopes. A local potential as between two α -particles can be defined between other clusters, for example between α -particles and ^{16}O , giving rise to intrinsic reflection asymmetric molecular structures in ^{21}Ne as parity doublets. An *Extended Ikeda Diagram* can be constructed based on these concepts with clusters and covalent neutrons.

§1. Introduction

We can look back into 30 years of studies of clustering phenomena in nuclear physics. There has been a steady evolution of the field, with a particularly strong push in recent years due to the study of neutron rich exotic nuclei. The earlier work was focused on light nuclei with $N=Z$ and their reactions, which has been on the forefront of research, in particular due to the observation of the heavy-ion resonances. Many theoretical concepts have been clarified in the last decades, specifically the equivalence of the shell model description with the cluster model approaches, once antisymmetrisation is considered in its full consequence (see also Ref. 1)).

The shell model approaches made big progress because they were able to include larger and larger model space. However, the need for cluster models can be illustrated by the fact that the most extensive shell model calculations²⁾ do not produce the famous second 0^+ -state of ^{12}C . The long standing problem how to define a cluster has been a line of thought all this time.¹⁾ One approach is perfectly illustrated by the diagram^{3),4)} proposed by Ikeda in 1968. In Fig. 1 this diagram is shown. The concept suggests that states of a particular cluster-(sub)structure appear close to the threshold for the decay of the composite system into these “fragments”. This can actually be observed with rising excitation energy starting from the bound states of normal nuclei, as well as function of decreasing binding, when approaching the dripline. The main physical effect of the diagram resides in the properties of the nucleon-nucleon interaction, which gives rise to saturation and the stabilisation of clusters with particular “magic” numbers, and their mutually reduced interaction.

The study of scattering of light nuclei, which has been started 30 years ago is

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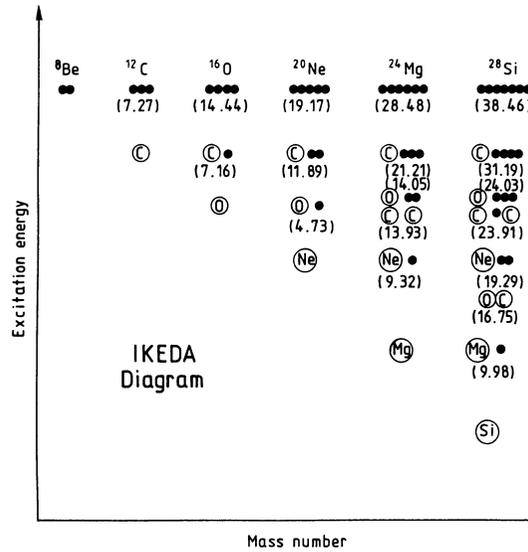


Fig. 1. The Ikeda-diagram for light α -cluster nuclei. The threshold energies are indicated.

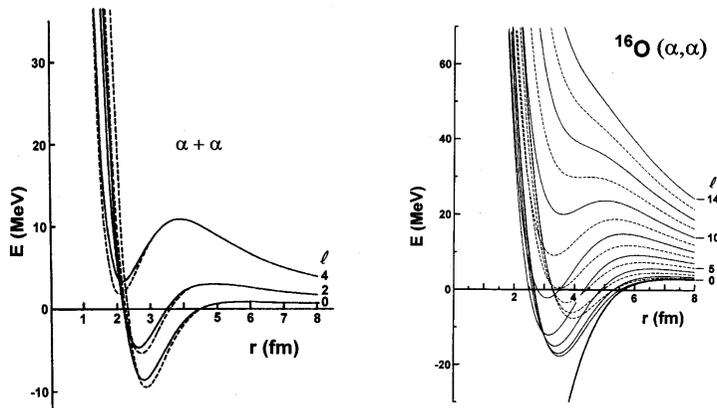


Fig. 2. Two examples of shallow local potentials⁶⁾ for the interaction of α -particles with α -particles: the case of ${}^8\text{Be}$, and with ${}^{16}\text{O}$: the case of ${}^{20}\text{Ne}$.

still the subject par excellence of cluster physics, because it involves the important aspect of nuclear dynamics, antisymmetrisation, inelastic couplings and the always present problem of the *cluster-cluster potentials*. The problem of antisymmetrisation in heavy-ion scattering is consistently described by Y. Tang and D. R. Thompson in the Maryland Conference¹⁾ in 1975, and by Friedrich⁵⁾ in 1981, and many others.^{6), 7)} With the concept of phase equivalent potentials and the supersymmetric transformation introduced in 1987 by Baye,⁸⁾ we are now able to discuss local potentials for the cluster-cluster interaction in molecular states. In these the bound states of the deep potentials, which are forbidden by the Pauli-Principle are removed. These local potentials can be used in the discussion of the covalent molecular structures, where antisymmetrisation between clusters and valence particles will be neglected.

These potentials have a repulsive core and are usually L -dependent.

The formation of covalent molecular structures relies on special properties of the nuclear potential, namely the occurrence of a “molecular” potential, with attraction at large distances and repulsion at small distances. There are two quite conspicuous examples: Figure 2 shows the cases of α -particles interacting with α -particles and with ^{16}O . The curves are actually the result of a reduction (using a supersymmetric transformation⁸⁾) of the “true” potential to a phase equivalent shallow potential. For the case of the $\alpha+\alpha$ potential the molecular potential created by Ali and Bodmer⁹⁾ for ^8Be (dashed curves) is seen to coincide well with the supersymmetric local potential. For a more detailed discussion I refer to the recent survey of the literature.⁶⁾ These potentials, which are repulsive at small distances are needed in order to build covalently bound nuclear cluster structures discussed in the next section.

§2. Molecular orbitals for valence neutrons

Over the last decades the molecular orbital model^{10)–16)} for neutrons has been applied to nuclear collisions, in which single nucleons, mostly neutrons are exchanged between two nuclear cores. This has been done for low energy reactions and for cases, where strongly bound cores (typically alpha-cluster nuclei) and valence nucleons can be defined in a clear way. In a collision process this approach corresponds to a choice of basis states, which takes the two center structure of the nuclear reaction into account and the couplings appear as radial and Coriolis couplings.¹¹⁾ It can be seen as an equivalent approach to the coupled reaction channel calculations based on the asymptotic basis states of the separated nuclei. A comparison for one example can be found in Ref. 17). For the stationary cases, particularly for the beryllium case, many authors have dealt with the molecular orbital approach.^{18), 19), 21)}

In the molecular orbital model and in the two-center shell model (in this case not only valence nucleons, but all nucleons are considered), a correlation diagram has to be drawn, as it is well known from atomic physics (see also Ref. 20), p. 328). The diagram merges at small distances with the Nilsson-diagram of the deformed compound nucleus. The molecular orbitals are classified according to the well known quantum numbers of molecular valence states: the K -quantum number for the projection of the spin, the σ and π orbitals for the $M = 0$, and $M = 1$ projections of the orbital angular momenta L , the parity, and the *gerade* and *ungerade*-symmetry due to the identity of the two molecular cores. With this correlation diagram we are able to discuss²¹⁾ the structure of the isotopes ^9Be , ^{10}Be and ^{11}Be .

Today we are able to define rotational bands in all three isotopes of ^{9-12}Be as compiled from different transfer reactions measured at HMI-Berlin by Bohlen et al.²²⁾

2.1. The beryllium isotopes

For ^{9-12}Be we consider²¹⁾ the population of the molecular orbits for a distance corresponding to the potential minimum, or in the Nilsson model to a deformation of $\beta_2 = 0.6-0.7$. At the distance, where the α - α potential has its minimum, the $K = 3/2^-$ orbit crosses the $K = 1/2^+$ orbit and becomes the lowest state at the

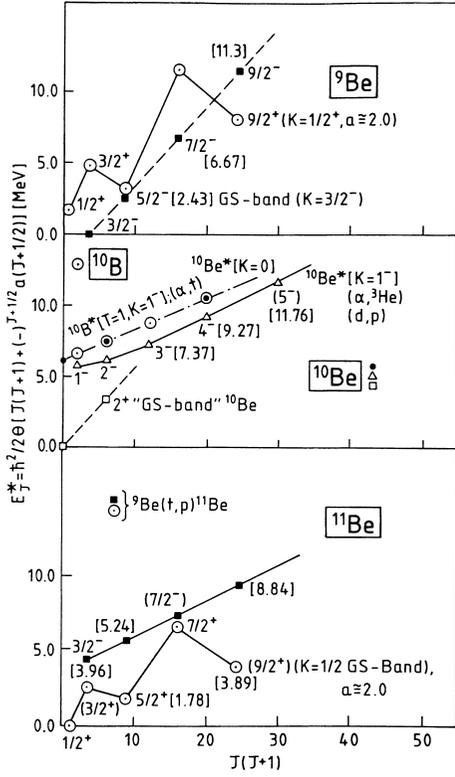


Fig. 3. Energy levels of the isotopes of ${}^9\text{Be}$, ${}^{10}\text{Be}$ and ${}^{11}\text{Be}$ grouped into rotational bands and plotted as function of angular momentum $J(J+1)$.

of the $K = 3/2^-$ rotational band, shown in Figs. 3 and 4, which have been completely

smaller distances. Thus the levels of ${}^9\text{Be}$ are completely reproduced by this scheme. In the systematic survey of the structure of the beryllium isotopes²¹⁾ all (!) known states of the isotopes ${}^9\text{--}{}^{12}\text{Be}$ have been grouped into rotational bands with molecular configurations. Today we are able to extend the rotational bands in all three isotopes of ${}^{10}\text{--}{}^{12}\text{Be}$ to the highest spins, as compiled in Fig. 4 from different transfer reactions measured at HMI-Berlin by Bohlen et al.²²⁾

These molecular states must be considered as true super-deformed shape isomers. This aspect is also observed in a dramatic way through the selectivity for the population of states in ${}^{11}\text{Be}$. We have studied the population of states²²⁾ in ${}^{11}\text{Be}$ by, (i) single nucleon transfer on ${}^{10}\text{Be}$ and, (ii) two-neutron transfer on ${}^9\text{Be}$. Whereas for the single neutron transfer reaction on ${}^{10}\text{Be}$ only three low lying single particle states (with the configurations $p_{1/2}$, $p_{3/2}$ and $d_{5/2}$) are observed, the two-neutron transfer populates strongly the higher spin members

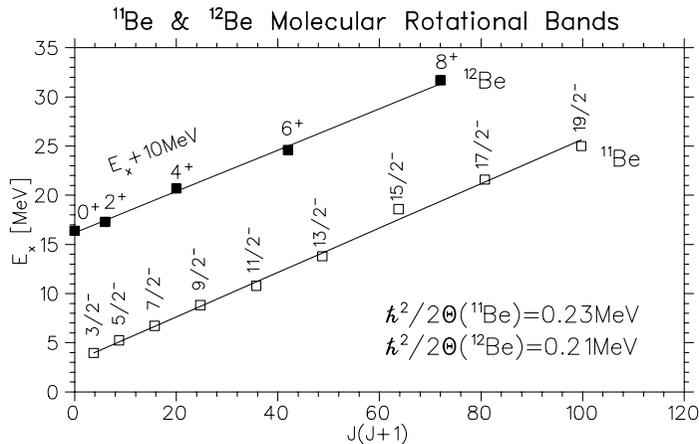


Fig. 4. Excitation energies of molecular bands in ${}^{11}\text{Be}$ (the $K = 3/2^-$ band) and ${}^{12}\text{Be}$ plotted as function of spin $J(J+1)$.

absent in the one neutron transfer spectrum.

The structure of the rotational bands in the $^{10-12}\text{Be}$ -isotopes is thus determined by the “driving forces” of the evolution of the π and σ orbitals as function of distance. Using the correlation diagram we recognise that these are the covalent neutrons in configurations with $(\pi)^1$ and $(\sigma)^x$, which results in a particular stability and a large deformation due to the properties of the $(\sigma)^x$ -bond. The moment of inertia of these bands is thus similar to that of the excited $K = 0^+$ band in ^{10}Be .

We may try to understand the maximum spin of $19/2$ for ^{11}Be in a deformed oscillator-shell model by counting the spins of the individual three valence nucleons: in fact the maximum spin of the valence neutrons would be only $[(3/2(p_{3/2}) + 8/2 \text{ (for } 2 \times 5/2))] = 11/2$; the total spin must be obtained by adding the maximum spin of the ^8Be -system, which is known to be 4, giving the total sum of $19/2$. Here, as remarked by Hess et al.,²³⁾ the highest spin would have been blocked by the Pauli principle, however, this blocking does not occur in the pure cluster model, where all antisymmetrisation effects are taken care by the proper choice of the local interaction potentials as discussed in the Introduction.

The proposed molecular properties of the states in Be-isotopes have found striking confirmation from “model independent” calculations (“computer experiments”) using the method of Antisymmetrised Molecular Dynamics (AMD) of Kanada-En’yo and Horiuchi.^{24)–26)} Particularly impressive are the density distributions obtained for the ground state 0^+ and second 0^+ state of ^{10}Be , which have been obtained by projection on spin and parity.²⁶⁾ The formation of covalent bonds with the $(\sigma)^2$ and $(\pi)^2$ configurations is very conspicuous in the density distributions.²⁶⁾

§3. Reflection asymmetric cases: Parity doublets in ^{21}Ne

In order to study the structure of ^{21}Ne in a model which consists of one neutron

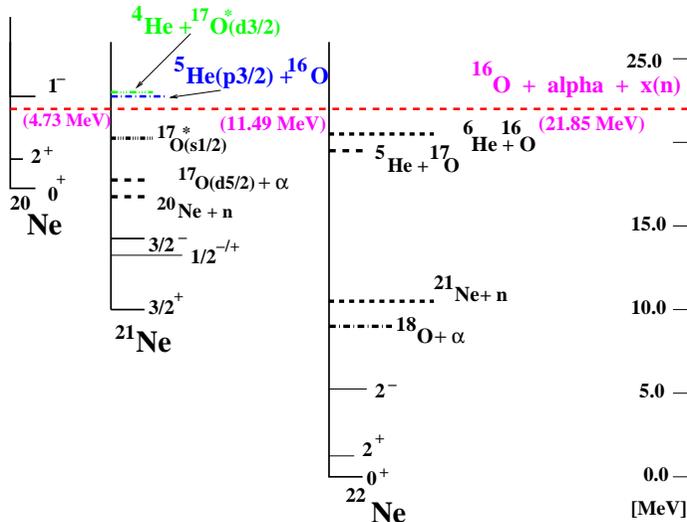


Fig. 5. Illustration of the thresholds (aligned to the same level) for the Neon isotopes for the $\alpha + (x \text{ neutrons}) + ^{16}\text{O}$ -cluster model. Some J^π values of band heads in ^{21}Ne with their K -quantum numbers are indicated.

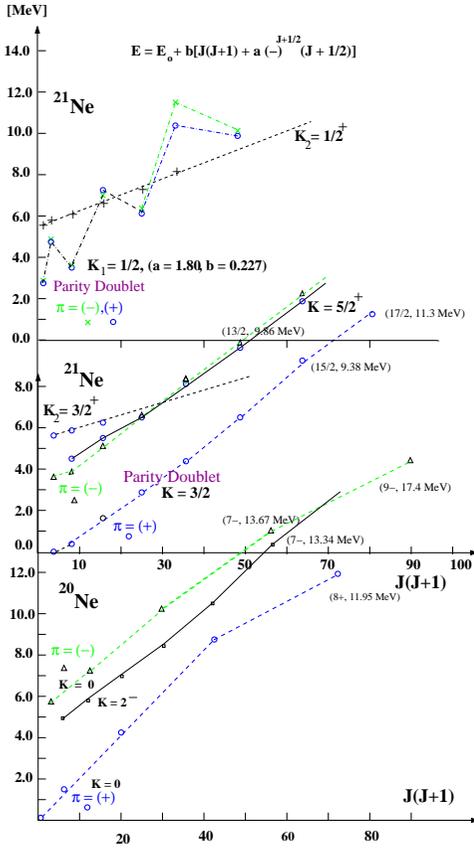


Fig. 6. Plot of the excitation energies of states in ^{20}Ne and ^{21}Ne showing rotational molecular parity doublets forming rotational bands, as well as “normal”(quadrupole deformed) $K = 1/2, 3/2$ and $5/2$ bands. This compilation places all low lying negative parity states in ^{21}Ne as belonging to parity-doublets, as expected from the structures shown in Fig. 7. The coefficients a and b are given for the band structure of the $K = 1/2$ parity doublet.

ric atomic molecules, as cited by Herzberg.²⁰⁾ The interesting question of the size of the signature splitting and of the purity of these configurations must be answered by additional experiments. For a complete spectroscopy the following rotational bands should be present in ^{21}Ne : a) Two parity doublets related to the octupole (molecular) shape as bands with $K = 1/2$ and $3/2$, with $(-)$ and $(+)$ parity. b) The remaining states should be arranged into bands, which are due to “normal” *reflection symmetric* Nilsson model configurations. Here the lowest orbitals are $[202, 5/2]$, $[211, 1/2]$ and $[202, 3/2]$. For the $[220, 1/2]$ and $[211, 3/2]$ Nilsson orbits we expect in addition two bands of positive parity with $K = 1/2, 3/2$, which may mix with the two positive parity bands cited

bound in a *two-center system with two unequal cores*, namely α and ^{16}O , we have to look into the structure of an asymmetric top with $K \neq 0$, with an intrinsic violation of parity, as illustrated in Fig. 7. I directly apply (see Ref. 27)) the molecular orbital (LCNO) model mentioned in the previous sections. Some important features can be deduced from an energy diagram, as shown in Fig. 5, which shows the positions of the single particle orbits of the neutron at the two centers, namely for α and ^{16}O . A *remarkable coincidence occurs: the $p_{3/2}$ resonance of ^5He (at 890 KeV) is almost degenerate with the $d_{3/2}$ -state (also a resonance at 941 KeV) in ^{17}O* . We note that this particular situation is known as the *quasi-resonance condition* for the sharing of valence particles in molecular science.

For the two-centre states in ^{21}Ne we take these two $j = 3/2$ orbits and construct the $K = 1/2$ and $K = 3/2$ states. These configurations are schematically shown in Fig. 7. There is no reflection symmetry, therefore states with definite parity are constructed by using superpositions, which produce a definite signature and parity.²⁸⁾ The outcome of these considerations is that there must be in the $(\alpha + ^{16}\text{O} + \text{neutron})$ -cluster model, *two bands* with $K = 1/2$ and $3/2$, each *with the two parities* $(+)$ and $(-)$. This situation is also well known in asymmet-

$$\{ | \bigcirc \bigcirc \rangle \pm | \bigcirc \bigcirc \rangle \}_{\frac{1}{\sqrt{2(1+\Delta)}}} = \Phi(^{20}\text{Ne})$$

$$\{ | \bigcirc \infty \rangle \pm | \infty \bigcirc \rangle \}_{\frac{1}{\sqrt{2(1+\Delta_n)}}} = \Phi(^{21}\text{Ne})$$

$$\{ | \bigcirc \infty \rangle \pm | \infty \bigcirc \rangle \}_{\frac{1}{\sqrt{2(1+\Delta_{2n})}}} = \Phi(^{22}\text{Ne})$$

$$\{ | \infty \bigcirc \infty \rangle \} = \Phi(^{26}\text{Mg})$$

"NUCLEAR WATER"

$$\{ | \infty \bigcirc \infty \rangle \} = \Phi(^{28}\text{Mg})$$

Fig. 7. Schematic illustration of the structure of molecular shape isomers in the Neon and Magnesium isotopes based on the $(\alpha + ^{16}\text{O})$ -cluster model plus some x -covalent neutrons. For the odd mass isotopes parity-doublets of bands are expected. The splitting of the bands will be proportional to the nonorthogonality Δ .

above. The new arrangement of states is shown in Fig. 6, and discussed in detail, with some relevant transition probabilities in Ref. 27).

§4. Chain-states: The case of ^{13}C

For the chain states of carbon isotopes, the inclusion of a third centre will in the first order, as in physical chemistry, introduce a further splitting in the basis states and a lowering of the lowest state relative to the three(four)-body thresholds. Starting with a $(^9\text{Be}+\alpha)$ structure, with the occurrence of intrinsically reflection asymmetric states,^{27),28)} we expect parity doublets. In a larger survey,²⁹⁾ the existence of two molecular parity split rotational bands in ^{13}C has been established, and they are illustrated in Fig. 8. Another important aspect, however, will enter if we consider trimers as in atomic physics, due to the underlying structure as $\alpha+\alpha+\alpha+n$. A model for a three-centre structure of the ^{13}C chain state is also formulated in Ref. 29). There we make use of the molecular orbital formulation known in physical chemistry as the Hückel method.³⁰⁾

The states of such structure should have large α -strength. It has been shown,²⁹⁾ that the states selected for Fig. 8 are strongly populated in reactions like $^9\text{Be}(\alpha,\alpha)^9\text{Be}$, $^9\text{Be}(\alpha,n)^9\text{Be}$, $^9\text{Be}(^6\text{Li},d)^{13}\text{C}$, etc. Fortunately, the $^9\text{Be}(\alpha,n)^{12}\text{C}$ reaction is very interesting for astrophysical problems, and has been measured a number of times.

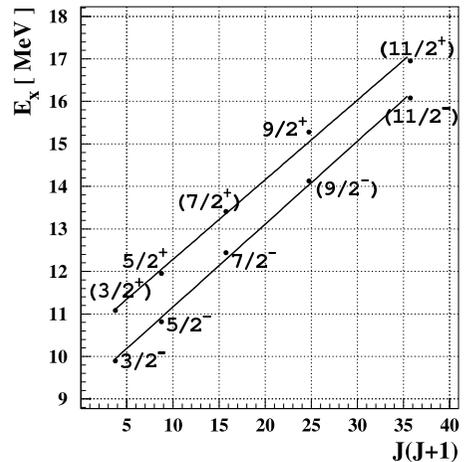


Fig. 8. Proposed rotational α -cluster bands in ^{13}C . their energies are plotted as function of their assumed spins, $J(J+1)$, as rotational bands with K -quantum numbers $3/2^\pm$, as indicated.

More recent work based on the explicit use of multi-center molecular configurations has been done for Be-isotopes and for C-isotopes by Itagaki et al.^{31), 32)}

§5. Summary and conclusions

Applying the observed results and extending the cluster model with the covalent structures of valence neutrons, we can construct more extended molecules, whose band heads can be estimated by using the information from the Neon-isotopes. The existence of an intrinsic symmetric molecule of “Nuclear Water”, $(\text{He})_2\text{O}$, can be predicted. The structure is schematically shown in Figs. 7 and 9. These states are true shape isomers with complicated deformations, which can *not* be easily described by a few terms in the Legendre-function expansions for nuclear shapes. A search for γ -transitions from such configurations appears as a future interesting challenge for nuclear spectroscopy. Radioactive beam facilities will give access to these nuclei and thus to new regions of deformations in light nuclei and clustering will play the dominant role in this field.

Using the cluster plus valence neutrons model we can use concepts of the Ikeda diagram, with the extension already formulated by the author in 1996, Ref. 21). The covalent binding of neutrons produces a new diagram of Fig. 9, a *Covalent Molecular Binding diagram*: “covalently bound molecular structures will appear in neutron rich

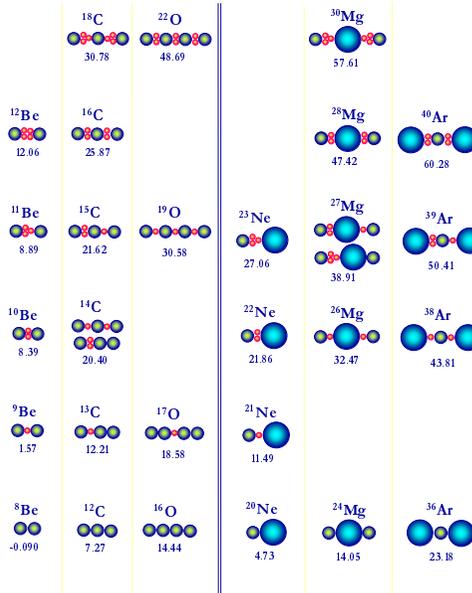


Fig. 9. Illustration of the structure of molecular shape isomers in neutron rich isotopes of nuclei consisting of α - and ^{16}O -clusters, plus some covalently bound neutrons. For the odd mass isotopes each K -quantum number gives rise to a parity doublet of bands. The splitting of these bands will be proportional to the probability to “shift” from one configuration to the other, given by the nonorthogonality Δ as illustrated in Fig. 7.

nuclei close to the thresholds for decomposition into neutrons, α -clusters, and other clusters”.

In the construction of this diagram in Fig. 9, I restricted myself to α -clusters and ^{16}O -clusters, because for these the appropriate local potentials are well defined. The establishment of these structures by particle- and γ -spectroscopy poses a very interesting challenge for the next decade.

Acknowledgements

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