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Author: Bill Rae

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The Hunt for the Nuclear Molecule

Ever since the discovery of the ^{12}C + ^{12}C resonances in the early 60's nuclear physicists have attempted to describe many heavy ion resonance phenomena in terms of nuclear molecules (or quasi-molecular states). The idea is very attractive and is obviously based on an analogy with the atom and atomic molecules. But until recently there has been no consistent explanation of the origin of the ^{12}C + ^{12}C resonances and other heavy ion resonances. Nor has there been convincing evidence that we are dealing with a ^{12}C–^{12}C molecular structure.

Recently there has been considerable progress due to the introduction of concepts and ideas from work on superdeformation. Indeed one might say that the resonances result from superdeformation rather than being the result of the formation of a molecule. In fact both of these statements might be true but it does require some redefinition of what we mean by a molecule in nuclear physics.

In atomic physics a molecule is formed between atoms with incomplete shells of atomic electrons. The shell structure in the composite system is totally different and if the correct combination of atoms is formed the shell structure in the composite system is such that when electrically neutral the system becomes a "closed shell" system, i.e. there is a significant gap between the last filled level in the molecule and the first empty level in the molecule. This latter aspect is true in the nuclear case also — the stability results from the shell structure in the composite system. But in the nuclear case we find that if we interpret the structure in terms of individual (spherical or normal) nuclei the molecule is formed form *closed* shell nuclei such as the α-particle and ^{16}O and ^{40}Ca!

The reason for this is to be found by studying the axially symmetric deformed harmonic oscillator. For many years I (Bill Rae) have pointed out the arithmetic relationships between the magic numbers at integer deformation ratios and spherical magic numbers. Recently Nazarewicz has rediscovered this mathematically and has proven that the systems at n:1 deformation can be described in terms of n spherical oscillators — or in terms of combinations of n spherical clusters. Thus axially deformed light nuclei can be considered as superdeformed nuclei or as molecular (or cluster) states built up of alphas, ^{16}O and ^{40}Ca nuclei!

For oblate shapes and certain triaxial shapes there is not the same bilateral viewpoint and superdeformation is often the only sensible description although the alpha cluster model can describe all shapes. For triaxial and oblate nuclei if a cluster description is valid then the clusters are not generally spherical nuclei but other deformed nuclei such as ^{12}C.

Recently most progress has been made in the study of the axially symmetric prolate deformations which seem to be the most rigid or stable and for which a molecular description is most valid but there is much work to be done on triaxial and oblate nuclei although the evidence for these is to be found in low energy scattering data since these structures are less stable at high energies and high spin.

Another feature that the prolate axially symmetric nuclei possess is that they are all *linear* molecules and the number of clusters is equal to the deformation ratio n:1. So for example 3:1 shapes involve 3 clusters while 2:1 involve 2 clusters. The systematics are so simple that you can invent any linear molecule made up of alphas and ^{16}O nuclei and it will correspond to some superdeformed nucleus which may be observable. (I think the only restriction is that only two type of cluster can be used but I am not sure about this for the deformed shapes with non integer deformations eg 1.5:1.) There is no evidence as of today for linear molecules involving ^{40}Ca except possibly the alpha cluster states in ^{44}Ti (^{40}Ca+α – 1.5:1 I think).

There are some well known examples:

^{8}Be $= \alpha - \alpha$

^{12}C excited 0^+ state at 7 MeV $= \alpha - \alpha - \alpha$

Excited rotational bands in ^{16}O starting at 18 MeV which decay to ...

^{8}Be + ^{8}Be $= \alpha - \alpha - \alpha - \alpha$

^{20}Ne ground state = ^{16}O$- \alpha$

A less well established example is in ^{32}S the ^{16}O–^{16}O resonances which should correspond to a 2:1 superdeformed state in ^{32}S. It is not clear whether the band head is actually bound with respect to the ^{16}O–^{16}O channel and that the observed resonances do not in fact correspond to a 1p-1h or 2p-2h excitation (the first vibrational state). But the known resonances that must correspond to this shape are very broad.

Using these systematics some of the more dramatic molecules that are predicted are:

α – ^{16}O – α in ^{24}Mg (3:1)

^{16}O – α – ^{16}O in ^{36}Ar (3:1)

^{16}O-^{16}O-^{16}O in ^{48}Cr (3:1)

$\alpha - \alpha - \alpha - \alpha - \alpha - \alpha$ in ^{24}Mg (6:1)

α – ^{16}O – α – α in ^{28}Si (4:1)

α – α – ^{16}O – α – α in ^{32}S (5:1)

This is not an exhaustive list and you can add your own. However is there evidence for any of these?

Let us start with the first example. As a graduate student I studied alpha transfer reactions at Harwell. I continued this work at Daresbury many years ago.

If you do alpha transfer onto ^{12}C you see a very strong dramatic rotational band. I now believe this corresponds to a triaxial superdeformed state in ^{16}O that has a ^{12}C–α structure (see below). Similarly if you do alpha transfer onto ^{16}O you see a very dramatic rotational band - the ground state band of ^{20}Ne which is an ^{16}O-alpha molecule. But when we did alpha transfer onto ^{20}Ne we saw nothing dramatic in ^{24}Mg - no band that stands out as in the other two cases - is there no α–^{16}O–α state in ^{24}Mg?

Well recently we (Charissa) did this experiment at Berkeley but we looked at a very specific decay channel for ^{24}Mg the ^{12}C–^{12}C channel - and we saw a *very* dramatic rotational band - nay a group of rotational bands - the α–^{16}O–α molecular states! Unfortunately the referees minds are so closed to new ideas that they could not see how important this experiment is!

But we do have some things to explain. Why look at the ^{12}C–^{12}C decay channel and why several bands? Lets take the first point. We have for many years used the Harvey model to predict how nuclear states might fission. If we apply the Harvey model to the α–^{16}O–α state we can predict that ^{12}C–^{12}C is a possible fission path for this molecule. Also I have demonstrated this using simple geometrical models with spheres representing alpha particles (see Daresbury annual report). There are many other decay channels predicted including α– ^{20}Ne and ^{8}Be - ^{16}O. We have an experiment approved at Berkeley to study the latter and this will be an important experiment.

Why several closely spaced bands? Well here we can again use the work of Nazarewicz. He shows that certain linear molecules are very soft to octupole deformation. These are the 2:1 molecules in which each cluster is different (eg ^{16}O–α) and the 3:1 molecules in which there are two small clusters and one large (e.g. α–^{16}O–α). Surprisingly the one with two large and one small are not octupole soft! I have also argued this from a more standard viewpoint using results from Nilsson Strutinsky calculations which predict similar instability in the same regions. Thus the multiple bands actually may result from octupole vibrations of the molecule and this is consistent with TDHF calculations, standard HF calculations and our cluster calculations (Marsh and Rae).

Interestingly although we have these octupole vibrations the cluster model predicts that negative parity states are unstable! and should not exist! I do not understand this fully but this is another reason why the ^{8}Be experiment at Berkeley is important since the ^{12}C + ^{12}C channel is only open for positive parity states (two identical bosons!).

Based on our results it is natural to associate the resonances we see with the ^{12}C + ^{12}C resonances discovered 30 years ago and it is this association that has got us into old prejudices with the referees! But I think the association is clear and thus we have at last a consistent interpretation of these resonance. They do indeed correspond to a molecular configuration but not a ^{12}C–^{12}C molecule (see later) but an α–^{16}O–α molecule!

Having discovered this α–^{16}O–α molecule is was very fortunate and surprising to realise that we had also discovered another molecule - the ^{16}O–α–^{16}O molecule at Daresbury in our attempt to study the ^{12}C–^{12}C resonances there. We have been working on the ^{24}Mg + ^{12}C breakup reaction for some years. With the much better data from the new array several things became clear.

The resonances we were populating in ^{24}Mg were narrow with higher spins than we had expected (4^+, 6^+ and 8^+ instead of 2^+).

They correlated with the ^{12}C + ^{12}C resonances and even our own Berkeley data.

The reaction mechanism was certainly not direct!

The latter came from data which were taken at different energies due to the machine not performing well at high voltages. The statistics are poor but I feel there is sufficient evidence to rule out direct. We have taken data at many more energies recently and this will be crucial in determining the reaction mechanism and we must get this data analysed quickly.

But what could be going on? Application of the Harvey model to the ^{16}O–α–^{16}O molecule indicates that it can break up into ^{12}C + ^{24}Mg. I proposed this idea some two years ago at a Charissa meeting. At that time it was viewed very sceptically. But there has been more evidence recently. The first was that there were many ^{16}O–^{16}O–α coincidences on tape! This is what we would expect from such a molecule. I predicted that ^{24}Mg on ^{16}O should not show breakup. The early evidence is that it does not. But it is crucial that this data is analysed carefully and quickly to get the backgrounds down in the ^{16}O target data to make this point conclusively and this is a very important experiment. Finally I have found some ^{20}Ne + ^{16}O $\rightarrow$^{12}C + ^{24}Mg resonance data at lower energies in the literature which is consistent with cluster model calculations and the l-value (resonant l-value) deduced from our data.

To follow this up further we have just completed an experiment to measure the resonant l-value in ^{24}Mg breakup on ^{12}C at a lower energy to compare with the cluster model calculations. Again an early conclusive result is important. One of the surprises is that this molecule is stable up to l values of around $30\hbar$ and excitation energies (above the band head) of over 50 MeV! It is clear that we must also make a more direct connection between the breakup work and the ^{20}Ne + ^{16}O resonances at lower energy. To do this we propose to submit a proposal to study the resonances in this channel up to the breakup energies but starting with ^{24}Mg + ^{12}C $\rightarrow$^{20}Ne + ^{16}O (Oxford proposal).

In conclusion the evidence for the ^{16}O–α–^{16}O molecule looks good but much work has to be done. In particular with the new detector system the ^{16}O–^{16}O coincidences should be measured. Indeed an excitation function showing a resonant yield in that channel would be very important.

Recently I was invited to Argonne to look at data for ^{12}C + ^{12}C $\rightarrow$^{12}C*(0^+) + ^{12}C*(0^+) taken by Russell Betts, Alan Wuoasma and Martin Freer. This shows a beautiful resonance at 33 MeV in the centre of mass and is very convincing evidence for the 6 alpha linear chain state in ^{24}Mg . The reason for this is that the excited 0^+ state in ^{12}C is thought to be the three–α linear chain state. also the energy of the resonance agrees well with the cluster model predictions (Marsh and Rae). This is an experiment we had approved for Daresbury but due to the needs of thesis material for graduate students and the pressures of the last year we had not been quite ready to do it.

This is a very important experiment though and there is much to be done because of it. First it indicates that these chain structures are stable up to 6:1 (ie 6 clusters in chain)! The width of the resonance is less than 3 MeV so these structures are relatively stable. It is also important since the only way into this 6–α chain state is through the small (4%) mixture of the linear chain state in the ^{12}C ground state. This means that we can think of ^{12}C in its ground state as a linear chain 4% of the time. This *will-* be very important for making other linear chain states – let your imagination run wild – but the cross sections will be a factor of 20 down at least! Hence we need more efficient detector systems and we need a detector system for ^{12}C*. Argonne used the Edinburgh double sided 5cm $\times$ 5cm design and this is clearly excellent for the ^{12}C* experiments!

There is yet more to be done. The angular distributions for the above 6–α reaction are very unusual. The resonance is apparently only observable at 90 degrees in the centre of mass. There is a factor of 5 enhancement of the oscillation at 90 degrees. Alan Merchant and I have reproduced this but we need up to 20 partial waves to do it! This is what I would expect. The moment of inertia is very large and the resonances are wide so the different partial wave resonances overlap and add coherently to produce an *aligned deformed nuclear density* in the *laboratory* at 90 degrees to the beam!! We can fit this crudely but the angular range of data taken at Argonne (70—110) is too small to get a full fit and a good understanding. We need normalised data over a much wider range and including zero degrees which is possible. Argonne hope to extend their data in November but possibly not including zero degrees. I suggest we go for zero degrees and very forward angles with some data at 90 for relative normalisation purposes. This could be potentially a very important experiment. Never before has deformation been observed in the laboratory!

Another prediction of these systematics is an ^{16}O–^{16}O–^{16}O molecule. We have also been working on this in the cluster model. Harvey predicts that this molecule can decay to two ^{24}Mg nuclei in their ground state and resonances are known in the ^{24}Mg + ^{24}Mg system. The cluster calculations predict this molecule to be *very* stable. It can support spins of up to $60\hbar$ and excitation energies above bandhead in excess of 100 MeV. This is in *total* disagreement with the liquid drop model which predicts maximum spins of the order of $40\hbar$. Several laboratories including Argonne are going to be looking for this molecule. It is an important experiment in the systematics and we should put every effort in trying to be first this time. We need to look for ^{16}O–^{16}O coincidences in the ^{24}Mg + ^{24}Mg reaction and we need to show that it is resonant with beam energy. My intuition is to go to the higher energy possible at Daresbury, viz. 180 MeV. Argonne and others will probably go for around 110MeV! We have had a quick look there and saw nothing but the detectors were not optimal. I think you need to get over all the Coulomb barriers with energy to spare and the ^{16}O nuclei might be preferentially produced in the 3^- at 6.13 MeV which has a much more tetrahedral alpha cluster structure as in ^{24}Mg + ^{12}C.

Where next? The choices are many. We need someone to systematically work through all the linear molecules with up to 6 or 8 clusters and work out the Harvey predictions and beam target possibilities. We must remember that ^{12}C has a linear chain state in the ground state 4% of the time. So $^{16}\text{O} + {}^{12}\text{C} \rightarrow \alpha\text{–}{}^{16}\text{O–}\alpha\text{–}\alpha$ is a possibility and I will follow this up with Neil Fletcher at Florida State for a possible collaboration in 1993/4. But ^{20}Ne + ^{12}C(chain) is another obvious choice, as is ^{20}Ne + ^{20}Ne, ^{20}Ne +^{16}O all Berkeley experiments!

Is there any 4–α chain state in the ^{16}O ground state? If we assume the same mixing matrix element as in ^{12}C, the difference from the point of view of first order perturbation theory is the energy denominator — 7 MeV in ^{12}C, 18 MeV in ^{16}O. This gives a probability of 0.6% - a factor of 7 down. This puts the ^{16}O + ^{16}O $\rightarrow$ 8–α chain state cross section down to 15 μb/49 = 0.3 μb at the peak. But it would be very clean like the 6 alpha data and the signature of 4 ^{8}Be nuclei would also be very clean! An experiment for a large array! (The ^{16}O + ^{12}C cross-section at the resonance peak is only down by a factor of 7 ie 2 μb. Thus a 7 alpha chain may be easier to go for first!)

We should also be considering ^{40}Ca but there are fewer experiments possible there. Two are ^{40}Ca + ^{16}O and ^{40}Ca + ^{40}Ca. To clean the data up we need to look at some very different exit channels predicted by Harvey. Looking too close to the entrance channels will be complicated by many other direct reaction processes. ^{40}Ca + ^{20}Ne is not as interesting because the 3:1 shapes cannot be made out of 3 different nuclei – two must be the same for a integer:1 superdeformed state but for half integer values this restriction is relaxed. All the integer ratio superdeformed states involving ^{40}Ca will also only involve ^{16}O for the lighter systems. For half integer deformations alpha particles may also be present.

Another exciting prospect is the study of decay from one molecule to the next. Obviously many of these are obvious. Each molecule can emit a single cluster to make another molecule. But it will not in general be possible to observe complete alpha decay chains when this involves the breakup of a ^{16}O cluster. Intermediate superdeformed states may not exist and even if they do exist they are likely to be much less stable and not exist at the higher spins and excitation energies. But I have demonstrated via the geometrical models that in many cases ^{12}C decay is a possibility. Indeed it so happens that the ^{16}O–^{16}O–^{16}O molecule can ^{12}C decay to the ^{16}O–α–^{16}O molecule which can ^{12}C decay to the α–^{16}O–α molecule which can ^{12}C decay to ^{12}C. This is a feature of all molecules with ^{16}O clusters since replacement of the ^{16}O cluster by an alpha always results from a ^{12}C decay and so the superdeformed state is guaranteed to exist! Only the energetics need then be considered. You can also show this with the Harvey model.

Indeed our ^{24}Mg + ^{12}C data show the ^{16}O–α–^{16}O molecule decay by ^{12}C emission to the α–^{16}O–α molecule and that is why we see the ^{12}C–^{12}C resonances in the decay of this molecule. This is the first observation of the exotic ^{12}C decay of a superdeformed state! If we see the ^{16}O–^{16}O–^{16}O molecule we should look for this ^{12}C decay to the ^{16}O–α–^{16}O which may then decay to α–^{16}O–α or ^{24}Mg(gs) by the emission of a ^{12}C nucleus.

In conclusion I think there are still many linear chain states to be investigated which involve ^{16}O and alpha nuclei. Many of these are reachable with existing beams (if we go to Berkeley) and at reasonable cross-sections. There is the possibility of more exotic molecules with ^{40}Ca to be explored. But in many ways more detailed experiments will be just as important. It is going to be important to measure both

α, ^{16}O and ^{12}C widths for these states — clearly people like Schiffer will not be convinced if widths are not measured and compared with theory. Alan and I hope to start work on the prediction of alpha and ^{16}O widths for the linear molecules — this we think is achievable. ^{12}C and ^{24}Mg widths are very much more difficult but are much smaller we suspect. So I would encourage you to think about these experimental problems while we start to tackle the theoretical problems. There is also much work to be done on very deformed linear chains where the resonances are overlapping - this is a very exciting new area of nuclear structure physics!!

I will now discuss the non axially symmetric superdeformed states. Not so much is known or understood about these as yet but there is one family that is reasonably extensive. These are my so called planar nuclei — made of a single plane of alpha clusters - they involve both superdeformed oblate states such as the gs of ^{12}C and the as yet unconfirmed 1:3 oblate state in ^{24}Mg (5 alphas in a ring with one alpha in the centre).

The family includes the ground state of ^{12}C, the triaxial "kite" state in ^{16}O (the 6.03 MeV 0^+ band with ^{12}C–α structure), a 5 alpha state in ^{20}Ne (0^+ around 11 MeV, 8^+ at 17.5 MeV, other members not known) which decays to $^{16}O(6.03)0^+$ or ^{12}C + ^{8}Be. There are two planar states predicted in ^{24}Mg. One the 1:3 oblate state and the other which looks like two coplanar ^{12}C nuclei. This is indeed a ^{12}C–^{12}C molecule but because ^{12}C is oblate this molecule is triaxial. The experimental evidence for this state is probably the band of resonances discovered in ^{12}C + ^{12}C inelastic scattering by Cormier. These resonances have all the correct properties including the fact that the deformations seem to be aligned as deduced from measurements of the gamma ray alignments.

Until recently I thought there was no evidence for the oblate state in ^{24}Mg. But with the recent discovery of the 6 alpha state the resonances in $^{12}C(gs)$ + $^{12}C^*(0^+)$ first discovered by Brian and confirmed at Argonne now do not correlate with the 6 alpha resonance. The oblate state in ^{24}Mg should be able to decay to $^{12}C(gs)$ + $^{12}C^*(0^+)$ (see Harvey) and so this may be evidence of that. It cannot be the band based directly on that configuration since this is not predicted to go to such high excitation (and spin?) but its possibly a 1p-1h or 2p-2h excitation of the band.

There is a prediction in both the cluster model and Nilsson Strutinsky models of a planar state in ^{28}Si. (Six alphas in a ring and one in the middle.) They may be others in heavier nuclei.

There are many triaxial superdeformed states. One family is intermediate between the ^{16}O–α and ^{16}O–^{16}O 2:1 states. It includes members such as ^{16}O–^{8}Be (transverse) and the ^{16}O–^{12}C (transverse) prolate state in ^{28}Si. Between the ^{16}O–α–^{16}O and ^{16}O–^{16}O–^{16}O states I have found a ^{16}O–^{8}Be–^{16}O (transverse ^{8}Be) state and there may be a ^{16}O–^{12}C–^{16}O state — but these are not so stable and breakup at quite low spins and excitation energies.

In the heavier nuclei there is a ^{16}O–^{24}Mg–^{16}O state in ^{56}Ni which is stabilised by octupole deformation. Harvey predicts that can decay to ^{28}Si + ^{28}Si and there are known resonances in this system. The cluster model predicts that this state should become more octupole deformed as it rotates - especially Y_{31} deformation (banana shape). But we still have to carefully crank this structure up to see if it will reach the spins measured in ^{28}Si + ^{28}Si. I hope Alan will tackle this after his experience with ^{48}Cr.

There is much to be done to correlate existing data on heavy ion resonances with these superdeformed states. This requires a systematic search through the literature for experimental data followed by a systematic search with the alpha cluster model. I hope that Pete Simmons might tackle this after we look at the ^{24}Mg + ^{12}C excitation function.

So in conclusion here there are many things to look for. The experiments we have done on ^{24}Mg + ^{12}C $\rightarrow^{16}$O+^{8}Be+^{12}C may identify the ^{16}O–^{8}Be states. We have data on ^{28}Si ^{12}C+^{16}O. The ^{24}Mg on ^{16}O at lower energies might reveal the $\rightarrow^{16}$O–^{8}Be–^{16}O states. The ^{12}C(gs) + ^{12}C*(0^+) may be evidence for the oblate state in ^{24}Mg. But there is work to do in ^{28}Si (planar state) and on the ^{28}Si + ^{28}Si resonances $\rightarrow^{16}$O + ^{24}Mg + ^{16}O. This would be a nice follow up to ^{48}Cr $\rightarrow^{16}$O + ^{16}O + ^{16}O, but the band may not go up high enough in energy for this decay channel to open up significantly.

Overall I think we are just at the beginning of the study of *nuclear molecules.* We have just begun to uncover these structures and we are just beginning to build the sort of detector arrays that will be required for their detailed study. It is clear that the Edinburgh style detectors are excellent for multiple alpha breakup studies. The sort of telescopes we are considering (Charissa) are better for heavy ions — but a combination of both will be needed for many of these studies. There is much to be done theoretically in terms of predicting excitation energies, moments of inertia, decay widths and angular distributions. There are many experiments that are crucial to the systematics — we have much data already that must be analysed quickly and thoroughly to support these ideas and there are several very crucial and important experiments in which we want to be first to publish. Thus there is little time since there is strong competition afoot. We need to encourage those with the technology and those with the energy needed to join our collaboration and we need to get the next version of the array built, up and running and producing data within months rather than years!

Finally when our new array is ready we might consider looking for gamma decays of some of these bands. Many years ago a group looked for the gamma decay from the ^{12}C + ^{12}C resonances using a crystal ball and particle detectors. You need to populate a resonance and look for a gamma decay to another resonance in coincidence with the particle decay from the final resonance. Its a triple coincidence experiment. Unfortunately, or fortunately the previous experiment yeilded a negative result. However it is my opinion that they looked for a decay from a Cormier (triaxial) resonance to an α–^{16}O–α type ^{12}C-^{12}C resonance. This is an obvious experiment to repeat but two changes need to be made: (i) Look at two α–^{16}O–α resonances, and (ii) use Eurogam rather than the crystal ball. Also one might consider the ^{16}O–^{8}Be particle decay as well as ^{12}C–^{12}C. We need to talk to Mike Bentley. Also maybe Alan can calculate some gamma widths to give us an idea of the cross sections we expect. □End