

# ATOMIC CLUSTER SCIENCE: INTRODUCTORY NOTES

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This article is the introduction to the volume of proceedings of the “International Symposium Atomic Cluster Collisions: fission, fusion, electron, ion and photon impact” (a Europhysics Conference) held in St. Petersburg, Russia, July 18-21, 2003 (ISACC 2003) as a satellite meeting of the XXIII International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC 2003, Stockholm, Sweden, July 23-29, 2003). A brief introduction to atomic cluster physics, the interdisciplinary field, which developed rather successfully during recent years, is presented. A review of recent achievements in the detailed *ab initio* description of structure and properties of atomic clusters and complex molecules as well as the methods of their study is given. The main trends of development in the field are discussed and some of its new focuses are outlined.

## 1. Introduction

The “International Symposium Atomic Cluster Collisions: fission, fusion, electron, ion and photon impact” (a Europhysics Conference) was held in St. Petersburg, Russia, July 18-21, 2003 (ISACC 2003) as a satellite meeting of

the XXIII<sup>rd</sup> International Conference on Photonic, Electronic, and Atomic Collisions (ICPEAC 2003, Stockholm, Sweden, July 23-29, 2003). This international Symposium promoted the growth and exchange of scientific information on the structure and properties of atomic cluster systems studied by means of photonic, electronic and atomic collisions. Particular attention during the symposium was devoted to dynamical phenomena, many-body effects taking place in cluster systems, which include problems of fusion and fission, fragmentation, collective electron excitations, phase transitions and many more. Both experimental and theoretical aspects of atomic cluster physics uniquely placed between atomic and molecular physics on the one hand and solid state physics on the other, were discussed at the symposium.

During the last decade it was recognized, both experimentally and theoretically, that complex molecules and atomic clusters (ACs) often possess unique properties, which make them a new object of physical research, rather different from both a single atom and from the solid state (see Refs. 1, 2 and references therein). The knowledge of the detailed electronic and ionic structure of single complex molecules and nano-clusters can be essential for various practical applications, such as the formation of new materials, nano-structures, in the design of drugs and biologically active species as well as for the understanding of fundamental issues, such as the functioning of quantum and thermodynamic laws in nano-scale systems or mechanisms for the formation of complex multi-atomic systems, self-assembly and functioning.

The demand for understanding of the principles of assembly and functioning of complex multi-atomic systems such as bio-molecules or nano-clusters is tremendous, because of the potential use of this knowledge for purposes of microelectronics, of biochemistry, the drug industry etc. The problems of self-organization, of self-assembly and of the functioning of complex multi-atomic aggregates and their interactions have been addressed both theoretically and experimentally in a large number of papers from different perspectives (for a review, see Refs. 1, 2). Often, these problems can be reduced to the problem of the interaction of a limited number of atoms within a complex molecule or even to the interaction of a single atom or ion with a certain fragment of a complex molecule (an active center responsible for a certain function) or a cluster structure. Thus, in order to achieve a real breakthrough in the field, one needs to learn how to handle both theoretically and experimentally (i.e. to be able to manipulate experimentally and predict theoretically) properties of multi-atomic systems containing about 100 atoms, or maybe a little less or a little more than this limit. With this

knowledge in hand, one can then move towards a detailed *ab initio* understanding of the properties of larger multi-atomic systems, bio-molecules (proteins, DNA), which typically consist of rather small fragments (amino acids or bases) whose structure and interactions do not involve more than 100 atoms.

These structures are, nowadays, subjects of very intensive theoretical and experimental studies in many physical, chemical and biological laboratories and institutions worldwide. A variety of methods have been used to investigate these objects (see, e.g., Refs. 1–9 and this book). Due to these efforts a vast amount of physical, chemical and biological data on the properties of complex multi-atomic systems and their interaction with the environment have been accumulated. However, it can be stated that until now there is no consistent theoretical approach, based solely on the fundamental principles of quantum physics, which might allow one, not only to explain systematically the known experimental data, but also, and this is quite essential, to predict new properties of the objects and new phenomena related to them. Nearly all theoretical approaches developed so far can be termed ‘phenomenological’ in the sense that each one of them substitutes the full quantum-mechanical description of the dynamics of constituents of a multi-atomic structure with a model theory which uses a set of parameters deduced from the experimental data. Each of these models is able to reproduce a limited number of particular properties of a complex multi-atomic system of a particular type, since the sets of the parameters involved are not of a universal nature. Thus, the model theories have severe restrictions: in each case they can explain but few of the experimental data. From them, often, one can hardly draw general conclusions or produce predictions of the properties of other structures. A more accurate description of the electronic and ionic structures, internal dynamics and interaction with external objects and fields has to be elaborated. The development of such an approach, which is multifaceted and includes not only theoretical investigations based on the first principles of quantum many-body theory but also implies a great amount of experimental work and computing, is currently the subject of joint efforts by specialists in various fields of physics and chemistry.

Theoretical approaches for the description of complex multi-atomic systems built on *ab initio* principles, model approaches and experiments will create real breakthroughs in understanding essential properties of complex multi-atomic formations, nano-clusters, bio-molecules and the mechanisms of their assembly and functioning. This will open up new possibilities for

cheap computer experiments for modelling complex multi-atomic systems possessing unique properties, for example, biologically active molecules. This knowledge is demanded in various applications ranging from micro-electronics to micro-biology and medicine.

The complete theoretical description of nano-scale systems consisting of about 100 atoms is extremely difficult.<sup>4</sup> So far, an *ab initio* many-body quantum mechanical description accounting for all electrons in the system can be used effectively for systems of a few tens of atoms<sup>5,6</sup> rather than hundreds. The computer power required for such calculations<sup>5,6</sup> grows exponentially with increasing molecular or cluster size. Therefore, one needs to invoke various simplified model approaches in order to describe complex systems of sufficiently large size.<sup>4,7,8</sup> However, often, the predictability of large systems by such approaches varies dramatically, particularly in the cases when modelling of complex molecules or nano-clusters neglects quantum effects.

Therefore, a careful choice of the model and accurate accounting for many-body and quantum phenomena are very important, as is demonstrated by various examples in this book. Thus, the high predictive power of a model can be achieved on the basis of detailed comparison of the predictions of the model and *ab initio* approaches with each other and with experiment for relatively small systems, consisting of tens of atoms, and by the extrapolation of the model postulates towards larger scale systems.<sup>4-6</sup>

In order to illustrate these ideas and some of the topics and focuses of this book, in the next sections, we briefly discuss fission, fusion and collision processes involving ACs as well as some general aspects of AC science.

## 2. Atomic Cluster Science

A group of atoms bound together by interatomic forces is called an atomic cluster. There is no qualitative distinction between small clusters and molecules, except perhaps that the binding forces must be such as to permit the system to grow much larger (in principle: with no upper limit to size) by stacking more and more atoms or molecules of the same type if the system is to be called a cluster. As the number of atoms in the system increases, ACs acquire more and more specific properties making them unique physical objects different from both single molecules and from the solid state.

In nature, there are many different types of AC: van der Waals clusters, metallic clusters, fullerenes, molecular, semiconductor, mixed clusters, and

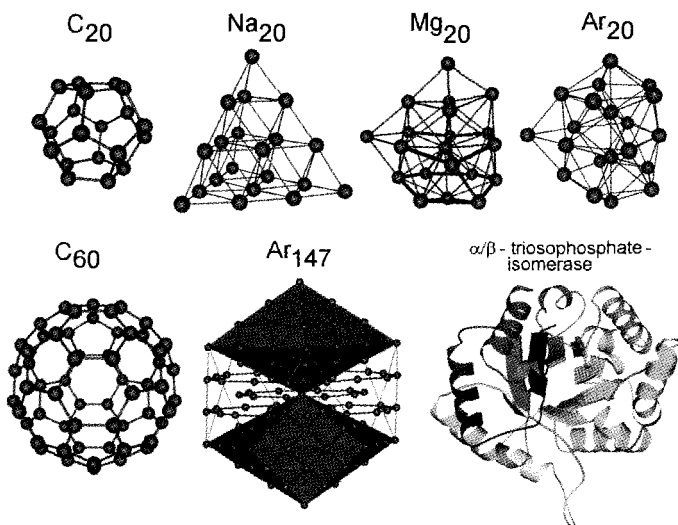


Fig. 1. The different nature of interatomic forces results in different principles for their organization within clusters and complex molecules. Geometries of the presented ACs have been calculated in Refs. 4–6 and 9 the structure of the protein globule ( $\alpha/\beta$ -triosphosphate-isomerase) is taken from Ref. 3.

their shapes can depart considerably from the common spherical form: arborescent, linear, spirals, etc. Usually, one can distinguish between different types of clusters by the nature of the forces between the atoms, or by the principles of spatial organization within the clusters. Clusters can exist in all forms of matter: solid state, liquid, gases and plasmas.

In Fig. 1, we present images of a few clusters in order to show a big variety of cluster forms existing in nature. We also show the structure of the  $\alpha/\beta$ -triosphosphate-isomerase globule aiming to stress that complex molecules such as proteins can be treated as clusters of subunits and that each of the subunits is a cluster on its own.

The novelty of AC physics arises mostly from the fact that cluster properties provide a better understanding of the transition from the single atom or molecule to the solid state limit. Modern experimental techniques have made it possible to study this transition. By increasing the cluster size, one can observe the emergence of the physical features in the system, such as plasmon excitations, electron conduction band formation, superconductivity and superfluidity, phase transitions, fission and many more. Most of

these many-body phenomena exist in solid state but are absent for single atoms.

The science of clusters is a highly interdisciplinary field. ACs concern astrophysicists, atomic and molecular physicists, chemists, molecular biologists, solid-state physicists, nuclear physicists, plasma physicists, technologists all of whom see them as a branch of their subjects but cluster physics is a new subject in its own right.

Significant progress achieved in the field over the past two decades ushered in the understanding of ACs as new physical objects with their own distinctive properties. This became clear after such experimental successes as the discovery of the fullerene  $C_{60}$ , of the electronic shell structure in metal clusters, the observation of plasmon resonances in metal clusters and fullerenes, the observation of magic numbers for various other types of clusters, the formation of singly and doubly charged negative cluster ions and many more. A complete review of this field can be found in review papers and books, see e.g. Refs. 1, 2, 10–15 and the present book.

### 3. Distinctive Properties of Atomic Clusters: Cluster Magic Numbers

ACs, as new physical objects, possess some properties, which are distinctive characteristics of these systems. The cluster geometry turns out to be an important feature of clusters, influencing their stability and vice-versa. The determination of the most stable cluster forms is not a trivial task and the solution of this problem is different for various types of cluster. The stability of clusters and their transformations is a theme which does not exist at the atomic level and is not of great significance for solid state but is of crucial importance for AC systems. This problem is closely connected to the problem of cluster magic numbers.

The sequence of cluster magic numbers carries essential information about a cluster's electronic and ionic structure. Understanding the magic numbers of a cluster is pretty well equivalent to understanding its electronic and ionic structure.<sup>4</sup> A good example of this kind occurs for sodium clusters. In this case, the magic numbers arise from the formation of closed shells of delocalised electrons, one from each atom (see Refs. 10, 14 and references therein). Another example is the discovery of fullerenes, and in particular the  $C_{60}$  molecule,<sup>16</sup> by means of the mass spectroscopy of carbon clusters.

In Fig. 2, we present the mass spectra measured for Na and Ar clusters (see<sup>10,12</sup> and references therein), which clearly demonstrate the emergence

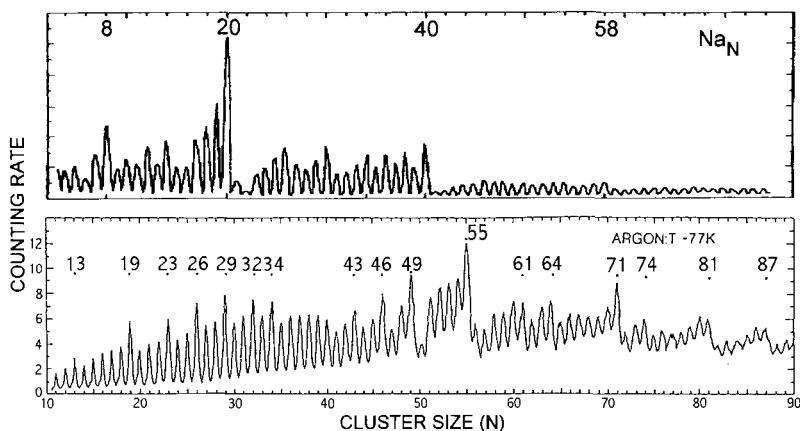


Fig. 2. Mass spectra measured for Ar and Na clusters (see Refs. 10,12,15 and references therein). The intense peaks indicate enhanced stability.

of magic numbers. The forces binding atoms in these two different types of clusters are different. The argon (noble gas) clusters are formed by van der Waals forces, while atoms in the sodium (alkali) clusters are bound by the delocalized valence electrons moving in the entire cluster volume. The differences in the inter-atomic potentials and pairing forces lead to significant differences in structure between Na and Ar clusters, their mass spectra and their magic numbers.

In Fig. 3, we present and compare the geometries of a few small Na and Ar clusters of the same size. It is clear from Fig. 3 that different principles of cluster organization result in different geometries of the alkali and noble gas cluster families.

Such differences can easily be explained. The van der Waals forces lead to enhanced stability of cluster geometries based on the most dense icosahedral packing. The most prominent peaks in mass spectra of argon clusters correspond to completed icosahedral shells of 13, 55, 147, 309 etc atoms. The origin of these magic numbers can be understood on the basis of the classical equations. The origin of the sodium cluster magic numbers is different and is based on the principles of quantum mechanics. In this case the cluster magic numbers 8, 20, 34, 40, 58, 92 etc correspond to the completed shells of the delocalised electrons:  $1s^2 1p^6 1d^{10} 2s^2 1f^{14} 2p^6$  etc. This feature of small metal clusters make them qualitatively similar to atomic nuclei for which quantum shell effects play the crucial role in determining their properties.<sup>17</sup>

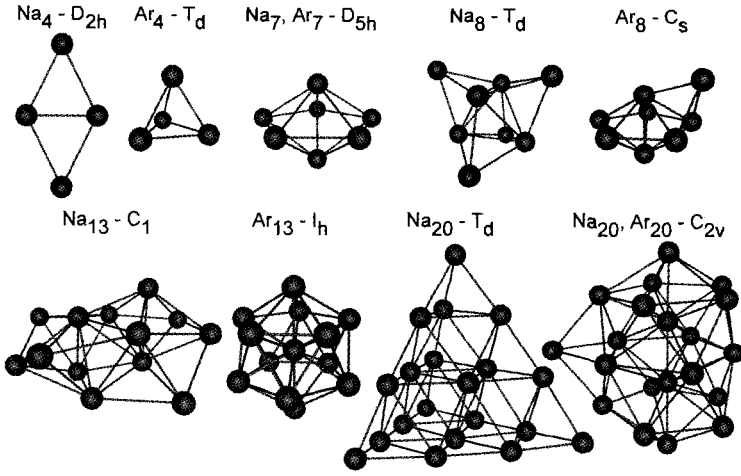


Fig. 3. Geometries and the point symmetry groups of some Na and Ar clusters calculated in Refs. 4, 5.

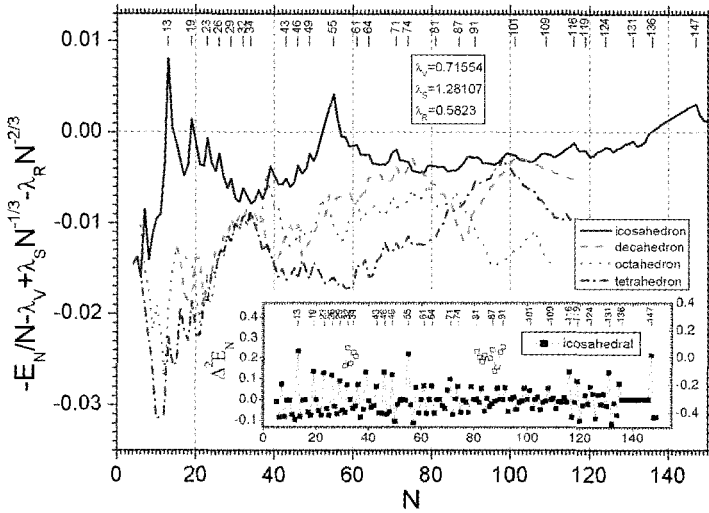


Fig. 4. Binding energies and their second differences for Ar clusters calculated in Ref. 4.

The enhanced stability of cluster systems can be characterized by computing the second differences in cluster binding energies. In Fig. 4 we present Ar clusters binding energies and their second differences calculated in Ref. 4. The correspondence of the peaks in Fig. 4 to those in the Ar clusters mass spectrum shown in Fig. 2 is readily established.

Finally, let us stress the obvious connection between AC physics and physics and chemistry of large molecules, such as proteins or DNA, which in fact can be treated as large clusters of amino acids or bases. The characteristic size of a fragment (amino acid or base) in such clusters is of the order of a few tens of atoms, i.e. the size of a small cluster. It is obvious that the knowledge gained from the AC studies is relevant for the biomolecular investigations and vice versa. A bunch of interesting phenomena can arise at the juncture of the two fields. For example, fusion of ACs with bio-molecules can create new objects which can be handled as easily as ACs or possess some specific properties and characteristics of ACs, but at the same time carry all essential features of bio-molecules and participate in bio-processes.

#### 4. Collisions Involving Atomic Clusters

The properties of clusters can be studied by means of photon, electron and ion scattering (see Course 9 by A.V Solov'yov in Ref. 2 and this book). These methods are the traditional tools for probing properties and internal structure of various physical objects.

Interesting phenomena arise in elastic collisions of electrons with ACs. For example, the diffraction of fast electrons by the fullerene  $C_{60}$  molecule was predicted and later observed.<sup>18</sup> The diffraction pattern in the electron elastic scattering cross section carries important information on the electron density in the vicinity of the fullerene's surface.

Electron excitations in metal cluster systems have a profoundly collective nature (see Ref. 11 and references therein). They can be pictured as oscillations of electron density against ions, the so-called plasmon oscillations. This name is carried over from solid state physics where a similar phenomenon occurs. Collective electron excitations have also been studied for single atoms and molecules. In this case the effect is known under the name of the shape or giant resonance. The name giant resonance came to atomic physics from nuclear physics, where the collective oscillations of neutrons against protons have been investigated.<sup>17</sup>

The interest of plasmon excitations in small metal clusters is connected

with the fact that the plasmon resonances carry a lot of useful information about cluster electronic and ionic structure. By observing plasmon excitations in clusters one can study, for example, the transition from the pure classical Mie picture of the plasmon oscillations to its quantum limit or detect cluster deformations by the value of splitting of the plasmon resonance frequencies.

The plasmon resonances can be seen in the cross sections of various collision processes: photabsorption and photoionization, electron inelastic scattering, electron attachment, bremsstrahlung (see Course 9 by A.V Solov'yov in Ref. 2). Both surface and volume plasmons can be excited. In electron collisions and in the multiphoton absorption regime, plasmons with large angular momenta play an important role in the formation of the cross sections of these processes.<sup>19</sup>

In Fig. 5, we present experimentally measured and theoretically calculated cross section for the photoabsorption of some Na and Mg clusters.<sup>20</sup> The cross sections are resonantly enhanced owing to the excitation of plasmon oscillations in the target cluster.

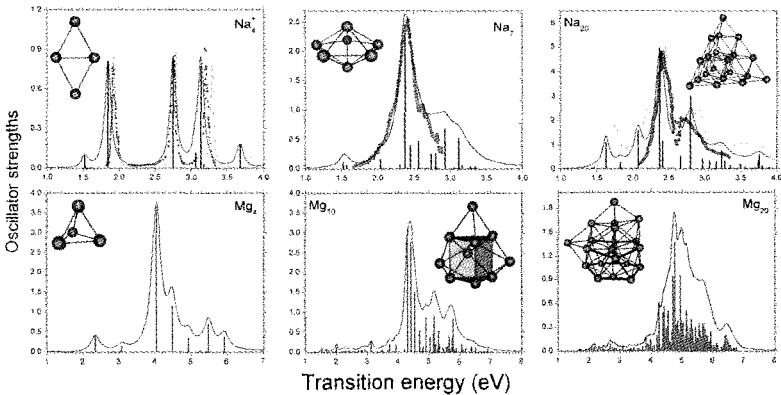


Fig. 5. Photoabsorption spectra of some Na and Mg clusters.<sup>20</sup>

Plasmon excitations in clusters decay via the Landau damping mechanism, while the relaxation of single electron excitations in clusters occurs via the interaction with the vibrations of ions, i.e. via the electron-phonon interaction (see Course 9 by A.V Solov'yov in Ref. 2).

Collisions involving ACs raise many more interesting physical problems.

For example, in collisions one can study phase transitions (solid-liquid or liquid-gas) in mesoscopic systems or the cluster multifragmentation process.

Another problem is linked closely to the problem of plasmon excitations in metal clusters. With increasing cluster size, the electronic energy levels of the single constituent atoms become grouped together, tending to form the conduction band, valence band etc. In this situation, the problem of localisation-delocalisation of the valence electron density in the cluster arises. This is known as the first order Mott phase transition. Plasmon excitations can be used as a probe of the Mott transition in ACs.

## 5. Fission Instability of Multiply Charged Clusters.

Multicharged ACs become unstable towards fission. The process of multicharged metal clusters fission is qualitatively analogous to nuclear fission. The fission instability of charged liquid droplets was first described by Lord Rayleigh in 1882 within the framework of classical electrostatics.<sup>21</sup> Reviews of recent work on metallic cluster fission, can be found in Refs. 2, 7, 13, 14.

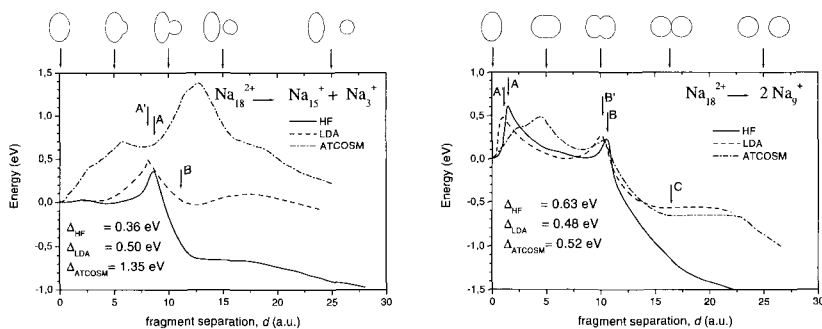


Fig. 6. Fission barriers for the asymmetric and symmetric fission channels of  $Na_{18}^{2+}$ :  $Na_{18}^{2+} \rightarrow Na_{15}^{+} + Na_3^{+}$  and  $Na_{18}^{2+} \rightarrow 2Na_9^{+}$  calculated in Ref. 7.

The fission process of ACs is interesting because it reveals the obvious parallel of AC studies with nuclear physics, where the fission process of nuclei has been studied for many decades.<sup>17</sup> The experiments on cluster fission provide a very good opportunity to test various concepts, approximations and AC models. Fission convincingly demonstrates the importance of the correct accounting for quantum and many body phenomena in the

description of multi-atomic systems. Dynamical aspects of the AC fission problem are also of great interest, because, contrary to nuclear physics, in the fission of ACs all the forces in the system are known and thus one can develop the full dynamical description of the process.

To illustrate the fission of charged metal clusters we plot in Fig. 6 the fission barriers for the symmetric and asymmetric fission channels of  $Na_{18}^{2+}$ :  $Na_{18}^{2+} \rightarrow Na_{15}^+ + Na_3^+$  and  $Na_{18}^{2+} \rightarrow 2Na_9^+$ . The barriers plotted in Fig. 6 have been calculated in Ref. 7 within the two-center LDA and Hartree-Fock jellium model and compared with the asymmetric two-center-oscillator shell model (ATCOSM). Figure 6 demonstrates the evolution of cluster shape during the fission process, the importance of cluster deformations, many-electron correlation and shell effects.

## 6. Fusion Process of Atomic Clusters.

The formation of a sequence of cluster magic numbers should be closely connected to the mechanisms of cluster formation and growth. It is natural to expect that one can explain the magic numbers sequence and find the most stable cluster isomers by modelling mechanisms of cluster assembly and growth, i.e. the fusion process of ACs.<sup>4</sup>

The problem of magic clusters is closely connected to the problem of searching for global minima on the cluster multidimensional potential energy surface. The number of local minima on the potential energy surface increases exponentially with the growth of cluster size and is estimated<sup>2,4</sup> to be of the order of  $10^{43}$  for  $N = 100$ . Thus, searching for global minima becomes an increasingly difficult problem for large clusters. There are different algorithms and methods of the global minimisation, which have been employed for the global minimisation of AC systems (see Refs. 2, 4 and references therein). These techniques are often based on Monte-Carlo simulations.

Alternatively, the algorithm based on dynamic searching for the most stable isomers in the cluster fusion process has been recently proposed.<sup>4</sup> The calculations performed with this new algorithm demonstrated that this approach is an efficient alternative to the known techniques of cluster global minimisation. The big advantage of the fusion approach consists in the fact that it allows one to study not just the optimized cluster geometries, but also their formation mechanisms.

In the recent work,<sup>4</sup> the fusion algorithm was formulated in a most simple, but general form. In the most simple scenario, it was assumed that

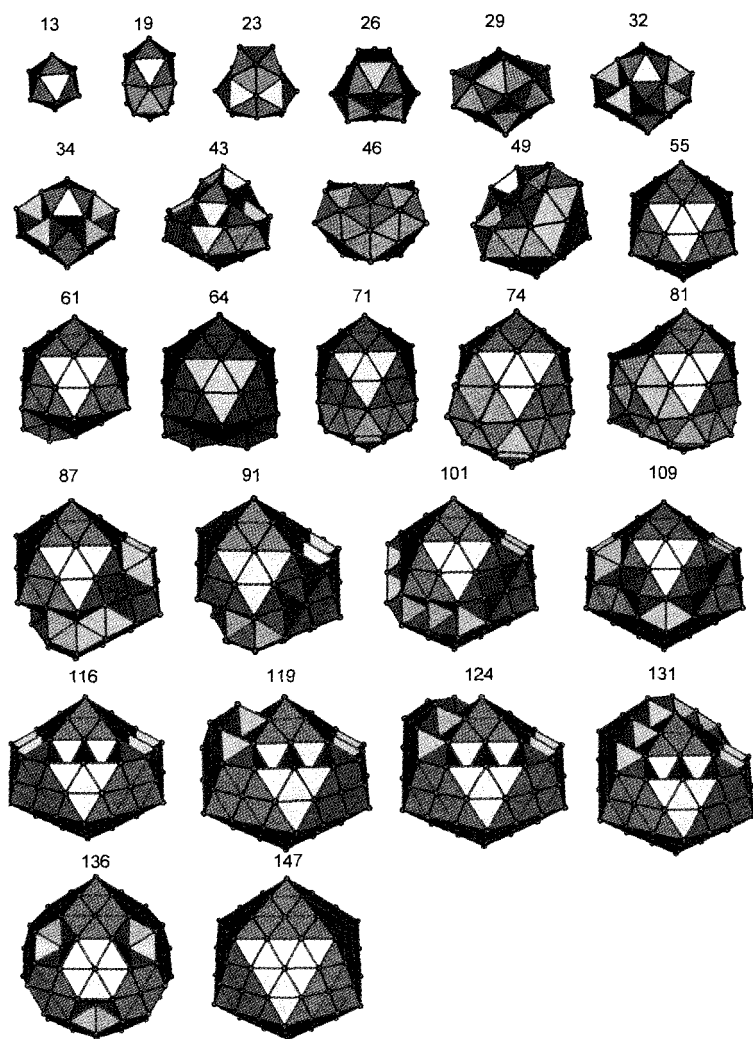


Fig. 7. Images of the Lennard-Jones global energy minimum cluster isomers.<sup>4,9</sup> The mass numbers of the pictured clusters correspond to the magic numbers of the noble gas (Ar, Kr, Xe) clusters.

atoms in a cluster are bound by Lennard-Jones potentials and the cluster fusion takes place atom by atom. In this process, new atoms are placed on the cluster surface in the middle of the cluster faces. Then, all atoms in the system are allowed to move, while the energy of the system is decreased.

The motion of the atoms is stopped when the energy minimum is reached. The geometries and energies of all cluster isomers found in this way are stored and analysed. The most stable cluster configuration (cluster isomer) is then used as a starting configuration for the next step of the cluster growing process.

Starting from the initial tetrahedral cluster configuration and using the strategy described in Ref. 4 cluster fusion paths have been analysed up to the cluster sizes of more than 150 atoms. We have found that in this way practically all known global energy minimum structures of the Lennard-Jones clusters can be determined. Figure 7 shows the images of the Lennard-Jones global energy minimum cluster isomers.<sup>4</sup> The mass numbers of the clusters represented correspond to the magic numbers of the noble gas (Ar, Kr, Xe) clusters.

So far, the cluster fusion algorithm has been applied to the noble gas clusters which are based on the LJ type of the inter-atomic interaction. However, the fusion process can be generated in a similar way for systems, like metal clusters, held together by quantum forces. This technique can also be used for the simulation of the fusion process of complex bio-molecules (proteins and DNA) or for the study of protein folding. It would be interesting to see to which extent the parameters of inter-atomic interactions can influence the cluster fusion process and the corresponding sequence of magic numbers or whether the clusterization in nuclear matter consisting of alpha particles and/or nucleons is possible. Studying cluster thermodynamic characteristics with the use of the technique developed is another interesting theme which is left open for future considerations.

## **7. Conclusions**

In recent years, AC physics has made very significant progress, but a large number of problems in the field are still open. The transition of matter from the atomic to the solid state implies changes of organization which turn out to be a good deal more subtle and complex than was originally supposed. Different types of clusters, composite clusters, various size ranges, cluster geometries, complex molecules (including biological), clusters on a surface and in plasmas, all provide additional themes which make this field of science very rich and varied. Collisions involving ACs, mass spectroscopy and laser techniques provide tools for experimental studies of the AC structure and properties.

However, what are the experimental limitations? Where should the the-

ory go next? Where does the future lie? Could clusters one day become the smallest devices or be used to make the smallest devices? Could one manipulate cluster isomers for the production of new materials and nanostructures? What is the difference between a cluster and a bio-molecule or a virus? Could molecules as complex as proteins or DNA and their functions be understood on the basis of classical mechanics or does one ultimately need to invoke the quantum theory? What are the principles of the self-organization of matter, of self-assembling and functioning on the nanoscale? We merely mention such intriguing questions here, but we hope that at least some of them will be resolved during the future development of the topics described in this book.

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