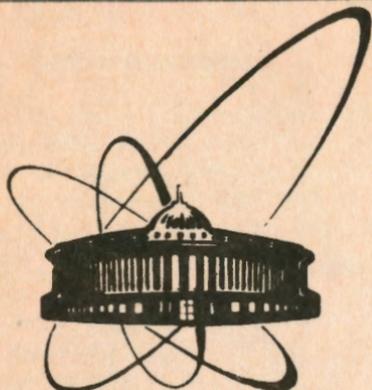


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ОБЪЕДИНЕННЫЙ
ИНСТИТУТ
ЯДЕРНЫХ
ИССЛЕДОВАНИЙ
ДУБНА

E4-92-529

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OVERLAP OF METAL CLUSTERS
AND NUCLEAR STRUCTURE PHYSICS

The talk at the 4th International Spring Seminar on Nuclear Physics
«The Building Blocks of Nuclear Structure»
(Amalfi, Italy, May 18-22, 1992)

1992

Обсуждаются свойства металлических кластеров (МК). Как стало недавно известно, МК, представляющие собой связанные системы, состоящие из атомов некоторых металлов, имеют оболочки такого же типа, как в атомах и ядрах. Последующие исследования выявили поразительное сходство многих свойств МК и атомных ядер: оболочки, квадрупольная деформация, деление, гигантские резонансы и т.д.. В результате, многие идеи и методы ядерной физики (в первую очередь теоретические) могут быть использованы после некоторой модификации для изучения МК. С другой стороны, МК имеют свои собственные уникальные особенности (возможность образования систем с числом частиц из 20000, супероболочки, смешанные МК). Физика МК, находящаяся на стыке атомной и ядерной физики, твердого тела, квантовой химии и т.д., является весьма многообещающим направлением как с точки зрения фундаментальных результатов, так и практических приложений.

Работа выполнена в Лаборатории теоретической физики ОИЯИ.

Препринт Объединенного института ядерных исследований. Дубна 1992

The properties of metal cluster (MC) are discussed. The MC representing the bound systems of atoms of some metals have recently been discovered to possess shells of the same type as in atoms and nuclei. Subsequent investigation has shown the striking similarities of many features of MC and nuclei: shells, quadrupole deformation, fission, giant resonances and so on. As a result, a lot of ideas and methods (first of all the theoretical ones) of nuclear structure may be applied after some modification in the MC field. MC have also their own very specific properties, such as supershells, the opportunity of form very large clusters with the number of atoms up to 20000, mixed clusters and etc.. The MC physics is relevant to many different branches (nuclear structure, atomic physics, solid body, quantum chemistry, etc.) and seems to be a very promising field for both fundamental results and practical applications.

The investigation has been performed at the Laboratory of Theoretical Physics, JINR.

1 Introduction

The MC physics in its modern status has arisen in 1984 when MC of some metals (first of all, alkali and noble ones) have been shown to manifest a very unusual property that was unknown before for atomic clusters. Namely, it has been discovered that these clusters have the shells with the same magic numbers as in nuclei and atoms (see refs.^{1,2} and refs. therein). Simultaneously, shells of the nuclear type in MC have been predicted on the basis of the self-consistent calculations by W.Ekardt³. This discovery meant that apart from atoms and nuclei that had been investigated for a very long time there is another quantum system with an analogous mean field.

Since 1984 a large progress in the study of MC was made. But due to a multidisciplinary character of this field the information on MC turned out to be distributed over a lot of different journals. Moreover, this information requires some special education for its understanding. There are two reviews^{2,4} on MC and the other two^{5,6} are to be published. The first one² provides a description of some general features of MC and of the experimental facilities but, on the other hand, covers only the first steps of the MC activity. In the second review⁴, the connection of MC with some other kinds of clusters is analyzed. A qualitative systematics of clusters is proposed. It is shown that clusters can be used as steps providing a unique chance to investigate all the way from one atom to a solid body and to understand at what step the cluster becomes the embryo of bulk matter embracing its main properties. The third review⁵ is devoted to collective excitations in MC.

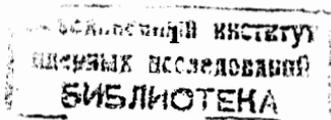
The aim of my talk is to present a short and simple guide for nuclear physics specialists in the MC field starting from the very beginning. I will try to cover all the most interesting modern directions of this field without its consideration in detail. Throughout the talk the permanent comparison of the properties of MC and nuclei will be done to display possible applications of nuclear physics experience in the MC field. This talk is based on a more general review⁶.

2 The basic properties of MC

As has been mentioned above, a MC is the bound system consisting of the atoms of some metal. MC can be divided into two subsystems: valence electrons (being outside the closed shells in atoms) and positive charged ions. The latter can be replaced in many cases, especially for sodium clusters, by a uniform rigid sphere of positive charge with a bulk density (jellium approximation).

The main characteristic of valence electrons, the density $n(\mathbf{r})$ in the ground state, is usually calculated by minimization of the Kohn-Sham density functional⁷:

$$E\{n(\mathbf{r}), \tau(\mathbf{r})\} = 1/2 \int \tau(\mathbf{r}) d\mathbf{r} - 3/4(3/\pi)^{1/3} \int n(\mathbf{r})^{4/3} d\mathbf{r} - \int \frac{0.44n(\mathbf{r})}{7.8 + (4\pi n(\mathbf{r}))^{1/3}} d\mathbf{r} + 1/2 \iint \frac{n(\mathbf{r})n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}d\mathbf{r}' \quad (1)$$



$$+ \int V_j n(\mathbf{r}) d\mathbf{r} + E_{jj},$$

where the terms are distinguished in order, a kinetic energy, a Coulomb exchange term in the Slater approximation (the Pauli principle), a correlation term of Lang-Kohn type⁸, a direct Coulomb interaction of valence electrons with each other and with the jellium, the jellium Coulomb energy. Also,

$$\tau(\mathbf{r}) = \sum_{i=1}^N |\nabla \phi_i|^2, \quad (2)$$

$$n(\mathbf{r}) = \sum_{i=1}^N |\phi_i|^2, \quad (3)$$

where ϕ_i is the electron single particle wave function.

Using the variational principle it is easy to obtain from (1)-(3) the expression for the mean field⁹:

$$v(n(\mathbf{r}), \mathbf{r}) = V_j(\mathbf{r}) + \int \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}' + \frac{dv_{ex-corr}}{dn}, \quad (4)$$

where the exchange-correlation term has a form

$$v_{ex-corr} = -3/4(3/\pi)^{1/3} n(\mathbf{r})^{4/3} - \frac{0.44n(\mathbf{r})}{7.8 + (\frac{3}{4\pi n(\mathbf{r})})^{1/3}}. \quad (5)$$

It is worth noting that the first two terms in (4) nearly compensate each other, which leads to a dominant rule in the mean field of the exchange-correlation term. Also, a rather consistent tail of the electron cloud outside the jellium sphere (spill out effect) should be noted. The spill out effect is the larger, the smaller the MC. It influences noticeably almost all the properties of MC.

It is easy to show that just valence electrons determine the quantum properties of MC⁴. Indeed, using Heisenberg's uncertainty relation

$$\Delta x \cdot \Delta p \geq \hbar, \quad (6)$$

one can obtain estimates for a zero-order momentum $\Delta p = \hbar/\Delta x$ and zero-point energy $\Delta E = (\Delta p)^2/2m$, the same expressions for both valence electrons and ions. Then, e.g., for sodium cluster N_{20} which consists of 20 monovalent atoms and has a diameter of 11.5 Å (this value determines the uncertainty in position of both electrons and ions) we have

$$\begin{aligned} - \text{for electrons} & \quad \Delta E_e \geq 0,16 \text{ eV}, \\ - \text{for ions} & \quad \Delta E_i \geq 0,04 \cdot 10^{-4} \text{ eV}. \end{aligned}$$

It is seen that zero-point energies for electrons and ions are quite different since the electron mass is much smaller than the ion mass. The typical temperature of MC is 300 – 800 K that corresponds to $kT = 0.03 - 0.08$ eV. Therefore, only electrons determine the quantum behavior of MC. The thermal energy of ions is much larger than their zero-point energy and, as a result, they move like classical particles.

The radius of MC is written as

$$R = r_{WS} \cdot N^{1/3}, \quad (7)$$

where N is the number of atoms; r_{WS} is the bulk Wigner – Seitz radius determined from the expression $n^+ = (4/3\pi r_{WS}^3)^{-1}$ with n^+ to be a bulk density. In sodium clusters, for example, $r_{WS} = 3.93$ a.u. (atomic units are 27.2 eV for energies and 0.529 Å for lengths, respectively). Then, exp. (7) gives for sodium clusters with $N = 20 - 200$ the radii 5.6 – 12.2 Å.

There are different methods of production of MC². Supersonic nozzle sources are widely used. The main principle of this source is the adiabatic expansion of the nearly saturated metal vapor mixed with a carrier inert gas into a vacuum through a narrow nozzle. The expansion and corresponding cooling of the metal vapor results in a creation of clusters (a process like condensation). Rather low temperature and high speed of MC promote the forming of a rather well-focussed cluster beam.

3 Comparison of MC with other systems

It is worth comparing a MC with an atom, a nucleus, a bulk matter and a molecule. The analysis of differences and similarities between these systems can clarify why MC is more similar just to a nucleus.

MC – Atom.

– *Similarity:* In both the systems the main interaction is Coulomb.

– *Differences:* a) When passing from hydrogen to uranium, the atom demonstrates the typical property of quantum gas: the mean density of electrons increases by almost two orders of magnitude while the size of the atom remains to be approximately the same. Vice versa, an increase in the number of atoms in MC does not change noticeably the density of MC. b) In atom the positive charge is concentrated in the centre of the system while in MC the density of positive ions is more or less uniform. In charged (ionized) MC the additional charge is placed at the surface.

MC – Nucleus.

– *Similarities:* a) Both valence electrons in a MC and nucleons in a nucleus are nearly free. b) In both the systems the mean density is almost constant as a function of a particle number. c) The charge density in a nucleus and the density of positive ions in a neutral MC are almost uniform. d) In both the systems the surface effects are very important. e) Both the systems with open shells are deformed.

– *Differences:* a) Different kinds of interaction. b) Nucleus has a considerable positive charge while MC can be both neutral and positive charged. c) Spin-orbit interaction in MC can be neglected. d) There is no yet a reliable evidence of pairing in MC.

MC – Bulk matter.

– The geometry of ionic structure of MC is spherical- or spheroidal-shaped while the geometry of the corresponding bulk matter is mainly cube-shaped. The larger MC, the closer the geometries of MC and bulk matter. In sodium and potassium clusters the bonding of valence electrons is found to be weak, nonlocalized and nondirectional. As a result, the ionic core of these clusters can be approximated by jellium. For other MC in some cases the effects of ionic structure should be taken into account. In this case the pseudopotential technique is used.

Since the MC is a relatively small system, the surface effects in MC and bulk matter, e.g., giant resonances of a surface type, should be rather different.

MC — Molecules.

— In principle, MC can be considered as a particular large molecules consisting of the same kind of atoms. Although the forces in MC and molecules have the same Coulomb interaction's origin their final form are different.

Particular properties of MC.

— a) MC is a metallic drop. So, the screening to the external fields in the interior of MC takes place. b) MC can include much more constituents (atoms) than nuclei contain nucleons and atoms — electrons. Due to this property MC, unlike nuclei and atoms, have the supershells. c) There are mixed MC which include the admixtures of atoms of other elements.

The above comparison shows that MC have some similarities with atoms, nuclei, bulk matter and molecules and, simultaneously, possess their own specific features. Most important for our purpose is that some properties of MC (nearly free valence electrons, the constant density, the uniform distribution of positive charge, the surface effects and the deformation) lead to very close analogies between MC and nuclei. As a result, a lot of the models and approaches derived in nuclear structure physics may be used also in the MC field.

4 Discovery of shells in MC and subsequent perspectives

As has been mentioned above, the discovery in MC of shells of the nuclear type has played a crucial role in the MC field. In figure 1(a) the experimental mass spectrum representing the abundance of sodium clusters in a beam as a function of N , the number of atoms in a cluster, is presented¹. Lower, in figure 1(b), the second difference

$$\Delta_2(N) = E(N+1) + E(N-1) - 2E(N), \quad (8)$$

calculated for the Nilsson single particle potential is shown¹⁰. In exp. (8) $E(N)$ is the total energy of MC with N valence electrons (or atoms since the sodium atoms are monovalent). The second difference (8) is known to display a peak if a gap in the single particle spectrum takes place, which is just the case for a stable system.

Figure 1 shows that clusters with $N = 8, 20, 40$ and 50 are most stable and, therefore, these values of N can be considered as magic numbers. The most striking point is that these magic numbers are the same as in nuclei. Some discrepancy taking place for larger magic numbers is explained by the negligible character of the spin-orbit interaction in MC. The analogous size effects have been revealed in measurements of the ionization potential and static dipole polarizability as well².

It should be noted that magic numbers are also known for non-metallic clusters, e.g., for noble gas and carbon clusters⁴. In these clusters all electrons are localized. So, the magic numbers in non-metallic clusters reflect a geometrical structure of MC. They do not coincide with the magic numbers in nuclei. The main merit of the discovery of magic numbers in MC is that the shells of non-geometrical nature were found in clusters for the first time.

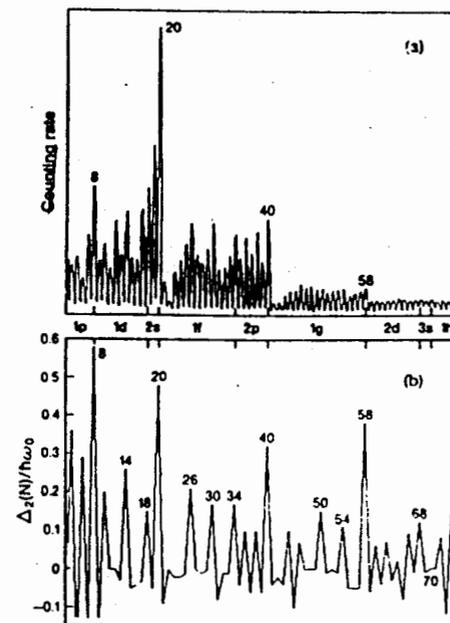


Figure 1. (a) Experimental abundance spectrum of sodium clusters¹. The number N is shown for selected clusters. (b) The second difference calculated for the Nilsson potential¹⁰.

During last years a lot of new information on MC has been obtained. Lower, the sketch of the main perspectives in this field is presented.

— 1). Now MC with N up to 20000 can be produced¹¹⁻¹⁵. Thus, for the first time, we have a unique chance to study a continuous transition from a single atom to a bulk matter.

— 2). 23 shells were found in MC^{14,15} — much more than is known in nuclei. Moreover, an essentially new effect, supershells, has been predicted^{12,13} and later observed¹⁴ in MC. Supershells can exist only in sufficiently large systems with $N \geq 1000$ ^{12,13}. So, neither nuclei, nor atoms are appropriate in this sense. MC are now a single real system where supershells take place.

— 3). MC with open shells were found to be deformed. Numerous manifestations of quadrupole deformation (particular peaks in the abundance spectrum¹⁰, a splitting

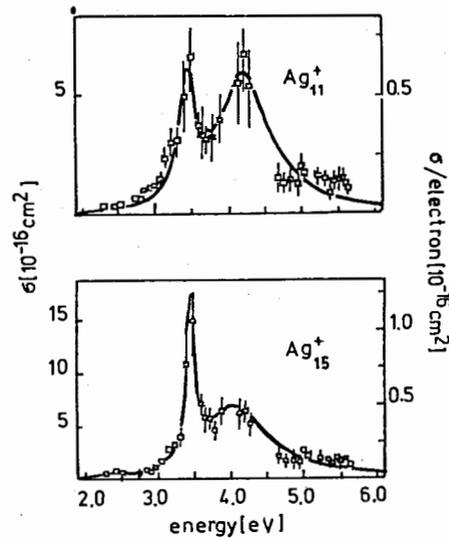


Figure 3. The GDR in the deformed clusters Ag_{11}^+ and Ag_{15}^+ ¹⁸. The experimental data and the calculations.

for example, the origin of pairing is ascribed to the attractive exchange-correlation term³¹.

The study of pairing in MC is also interesting in connection with the competition of pairing and deformation in MC²⁹.

— 6). The investigation of giant resonances is one of the most intensive directions in the MC physics^{5,6,9,16–18,23,24,34–49}. There is now a plenty of experimental data on the GDR in MC^{16–18,35–37}. As an example, the experimental results¹⁸ for the GDR in the deformed clusters Ag_{11}^+ and Ag_{15}^+ are presented in figure 3.

The GDR was found in different kinds of MC: spherical and deformed, neutral and charged, small and large. The main part of the experimental data was obtained in photoabsorption reaction (see, e.g., refs.^{16,17}). The theoretical investigations also are mainly concentrated on the GDR^{5,6,9,16,23,24,34,38–49}. Although there is a similarity between the nature of the GDR in MC and in nuclei, the GDR in MC has some particular features. For example, the fragmentation of the GDR in MC is, on the average, larger than in nuclei that is explained by a long-range character of Coulomb forces⁴⁶. There is a large difference in fragmentation of the GDR in neutral and charged clusters⁴⁴. Also, the response of MC to external electromagnetic fields should differ from their counterpart in nuclei due to fact that MC unlike a nucleus is a metallic drop.

There are no now the experimental data on the other giant resonances in MC. On

the other hand, there are interesting theoretical predictions, e.g., of the low-lying M1 "scissors" mode in deformed MC³⁹. The analogous mode is well known in nuclei⁵².

— 7). The MC produced at the experiment have usually rather large temperature (300-800 K)^{2,4}. In comparison with nuclei and atoms MC give a new possibilities for investigation of temperature effects^{12,13,48,50,51}. Indeed, because of the large difference between masses of electrons and a cluster MC may serve as a huge heat bath for valence electrons. Secondly, the temperature provides necessary conditions for the supershells in MC^{12,13}. And, at last, the temperature seems to influence the critical size of MC after which a new set of magic numbers appears¹¹. This new set reflects the geometrical structure of cluster.

— 8). There are mixed MC which contain the admixtures of atoms of other elements². Mixed clusters have a rich physics and seem to be promising for the practical applications, e.g., for the creation of new materials.

— 9). Because of the close similarity between MC and nuclei the numerous theoretical models and approaches widely used in nuclear physics can be applied after some modification in the MC field. Now this activity is very popular. In the MC physics there are known the applications of the Nilsson potential^{10,16,39}, the BCS method^{29,30}, the vibrating potential model³⁹, the sum rule approach^{38–42}, the random phase approximation^{39,43–46}, the self-consistent response function method^{23,48}, the fluid dynamics models^{9,49}, the IBM type models^{22,29}, the liquid drop model⁵³, the Strutinsky shell correction model^{20,28}, the relativistic models⁵⁴, the shell model calculations²⁴ and so on.

Probably, in some time the opposite process will take place: models and ideas from the MC field will be used in nuclear physics.

5 Conclusions

The MC field is very young. Nevertheless, this field has already involved many specialists from different branches of physics. The main reason is that MC, on the one hand, promise an exciting fundamental results and practical applications and, on the other hand, give possibility to use in the new field the available experience.

The close similarity of MC with nuclei opens interesting perspectives for the application in the MC field of a great theoretical potential of the nuclear physics. Without any doubt we can get in this way really fundamental results which could, in turn, be useful not only for MC but for nuclear physics too.

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Received by Publishing Department
on December 16, 1992.