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COMPOSITE NANOSTRUCTURES WITH METAL COMPONENTS

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The paper provides an overview of the current state of theoretical and experimental investigations of the properties (mainly optical) of composite nanostructures that contain metal components, as well as of small solid metal clusters. The paper describes traditional metal nanoshells and nanotubes as well as more complex nanosystems obtained in recent years – nanorice (ellipsoidal nanoshells), nanoeggs and nanocups (nanoshells with non-concentric core), and multi-layer nanoshells – so-called nanomatryoshkas. In the work, the description of the specific properties of these nanosystems and of the existing approaches to their investigation is given. The main aspects of the technical applications of the composite nanostructures with the metal components are described.

Keywords: *METAL NANOSYSTEMS, OPTICAL CONDUCTIVITY, PLASMON RESONANCE, NANOSHELLS, METAL NANOTUBES, NANORICE, NANOEGGS, NANOCUPS, MULTILAYER NANOSHELLS.*

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1. INTRODUCTION

Physics of nanostructures, i.e. objects with nanosize in one, two, or three dimensions, is a new promising direction in condensed-matter physics. As known, properties of nanostructures differ from the corresponding properties of massive bodies [1]. This is connected with some factors, namely, with substantial contribution of surface phenomena to the cluster properties (in nanostructures, fraction of surface atoms is comparable with fraction of volume atoms), impossibility of collective excitation with the wavelengths exceeding the cluster size (this essentially changes its thermodynamic properties), etc. In particular, dependence of the material constants, for example, conductivity [2], on the size and shape appear in such objects.

Metal clusters, nanowires, and thin films [1, 3] are of interest among nanostructures of different types from the point of view of the application in technics. Small metal particles have been studied theoretically [4, 5] and experimentally [6, 7] for a rather long time [1, 3, 8, 9]. More and more perfect methods have been developed recently to obtain small metal particles and their ensembles [10-12].

As known, the difference in the properties of small particles and massive bodies appears, in particular, when one (at least, one) of the characteristic dimensions of the system becomes equal to the characteristic parameter of the physical phenomenon which has dimension of length [1, 3]. Thus, the electron mean free path is such parameter in metal nanostructures, and the characteristic dimension of metal cluster or thickness of thin metal film can be, for example, the characteristic dimension. For example, in island films (thin films which are not continuous, but represent the population of small clusters – islands) one can observe the electron and photon emission during introduction of power into these films [1, 3, 13-18], when the characteristic dimension of an island becomes of the order of the electron mean free path. In this case, the matter is about relatively low powers, at which in bulk metal nothing of the kind is observed. This is explained by the attenuation of the electron-lattice interaction for the sizes of metal cluster which are less than the characteristic electron mean free path [17-25]. Intensity of the electron-phonon power interchange [25] for such clusters is two orders less than for bulk metals [26, 27]. Thus, hot electrons appear in a cluster during introduction of powers sufficient for the heating of electron subsystem only [28] that allows to explain not only the above mentioned photon and electron emission, but also other anomalous properties of island films, such as luminescence, electron emission, and nonlinearity of the volt-ampere characteristics [29]. New regularities (in comparison with massive metal objects) are also observed in the light absorption by small metal particles [30, 31], in general, in their optical properties [5] (in particular, sharp dependence of the absorption on the particle shape and electromagnetic wave polarization [2] appears). It is also known [4] that in small metal particles another behavior of the plasmon resonance in comparison with bulk metals is observed. Light absorption in small metal cluster depends substantially on the polarization of the electromagnetic wave incident on a shell [2]; for bulk metals such effects are not observed. A class of metal nanosystems, which have nanosizes in only two directions, – metal nanowires [32] – is studied separately; optical properties of such systems are investigated both experimentally (see, for example, [33]) and theoretically (see, for example, [4, 34]).

With the development of nanotechnologies, researchers have obtained the composite nanostructures, i.e. non-uniform nano-objects. Composite nanoparticles – in particular, those which contain metal elements – are intensively studied both theoretically and experimentally. If continuous nano-objects display a number of unusual properties, which condition their practical application, then various configurations of composite nano-objects display more unique properties and provide much more possibilities for the control of the properties of materials based on them.

The given work represents an overview of the physics of composite metal nanostructures (i.e. composite nanostructures which contain metal elements) at the moment.

2. CONTINUOUS METAL CLUSTERS

Continuous metal nanosystems are intensively studied theoretically. Recent investigations show, in particular, that optical properties of small metal particles (in contrast to massive ones) are substantially determined by their shape [2, 35-37]. Thus, absorption power in small metal particles of the same volume but different shape can differ by orders of magnitude [37]. Such regularities lead to the need of the study of the optical properties separately for particles of different shape. Thus, there are publications devoted to the optical properties of continuous metal clusters in the form of a ball [38, 39], cylinder [4, 40], ellipsoid [2], and parallelepiped [41], as well as to thin plane-parallel films. A number of papers ([2, 4, 34, 38, 39], etc.) and some monographs (for example, [1, 3]) are devoted to the theoretical investigation of the optical properties of different-shaped metal clusters. Effects connected with the influence of the shape and size of metal nanoparticle on its optical properties are studied, for example, in [2, 42, 43].

When studying the light absorption in metal nanosystems one should consider separately two absorption components – electrical (connected with the electric vector of the electromagnetic wave) and magnetic (connected with the magnetic vector). As theoretical investigations show [2], depending on the particle size and shape and electromagnetic wave frequency as well, electric absorption can be both much more and much less than the magnetic one. For metal nanosystems both electric and magnetic absorption are studied [2, 4, 40, 44, 45]. Optical properties of continuous metal particles are investigated theoretically using both the standard kinetic theory [2] (by the solution of the Boltzmann kinetic equation for conduction electrons in metal) – for nanosystems in the form of a sphere [46, 47] and cylinder [48, 49] and – the quantum-microscopic [4] approach depending on the temperature and ratio between characteristic cluster sizes and electron mean free path.

When speaking about the ensembles of fine particles, for example, island films, obviously that in order to obtain optical properties of such systems it is necessary to take into account the interaction between separate particles. (Thus, mutual influence of islands on the value of the local field due to the dipole interaction is considered in [50, 51] for island metal films.) However, exact calculation is complicated problem even for two identical metal spheres [52, 53] and especially for the system of such spheres [54]. In the case of the ensembles of nanoparticles, correlation effects, in general, should be taken into account. But for the ensemble with rather low concentration of nanoparticles, which have spherical symmetry (spherical metal particles and the below described nanoshells and nanomatryoshkas), or for the rarefied ensemble of nonsymmetric nanoparticles of the similar size and orientation, the problem of approximate description of the ensemble presents no difficulties, if optical properties of one particle are known. Last years, production of such ensembles became possible even for nanoparticles of complex configuration. Moreover, for the theoretical description of the optical properties of an arbitrary ensemble investigation of isolated nanoparticle is necessary. These factors, as well as the fact that production and experimental investigation of isolated metal nanoparticles of different configurations became possible last years, make calculations of optical conductivity of separate nanoparticle (to which the most of theoretical works are devoted) to be necessary and reasonable. Investigation of the optical properties of the ensemble of continuous metal nanoparticles is carried out, for example, in [55] (reflection of an infrared radiation from the layer of fine metal particles).

3. SYMMETRIC COMPOSITE METAL NANOSTRUCTURES

Synthesis and application of nanostructures of different configurations and, correspondingly, with different properties are the result of the development of nanotechnologies. Composite nanostructures are also synthesized and used last years. Small composite clusters with dielectric core and thin metal shell, which have nanosizes in three dimensions – the so-called nanoshells [56-60] (obtained in the laboratory typical nanoshells are close to symmetric ones, see Fig. 1) and fullerene nanotubes covered by thin metal layer [61, 62] – the so-called metal nanotubes (Fig. 2) belong to the above mentioned structures. We have to note that carbon nanotubes with metal properties are also called metal nanotubes. Hollow metal nanoshells [63, 64] were obtained and studied in the laboratory as well. Prospects for technical applications of such nano-objects are mainly connected with the possibility to control their properties, in particular, optical properties, more flexibly than for continuous nano-objects. This flexibility is connected with the possibility to change the characteristic dimensions of metal shell – internal and external – independently. (Optical properties of such systems are mainly determined by metal shell, dielectric core does not contribute substantially.) Moreover, intensity of the resonance scattering in typical nanoshells is much more than in typical continuous metal nanoparticles [65].

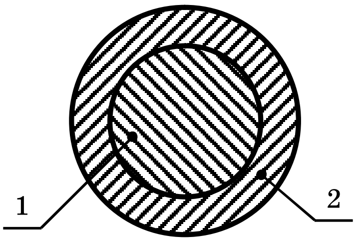

Fig. 1 *– Nanoshell in section: 1 – dielectric core; 2 – metal shell*

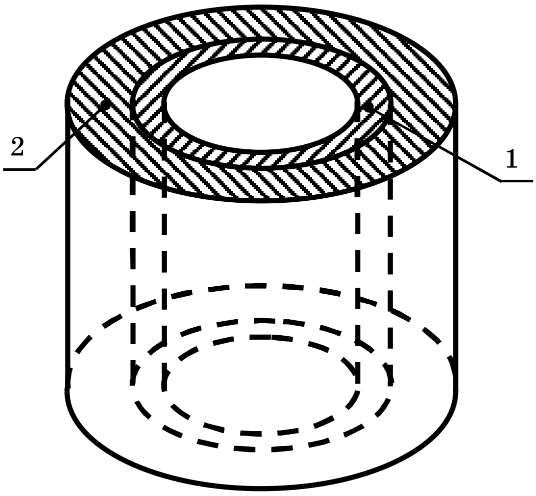

Fig. 2 *– Metal nanotube: 1 – carbon nanotube; 2 – metal shell*

As known, metal nanoparticles and nanowires are used in engineering due to their unique controlled optical properties mainly. Thus, they effectively absorb light on the given wavelength. Composite metal nanostructures allow extension of the working range of wavelengths in comparison with traditional continuous metal nanoparticles. Frequency of the plasmon resonance in composite metal nanostructures can be also changed in a wider range than for the corresponding continuous nanoparticles [58, 59]. Such flexibility in the control of the properties, in particular, optical properties, makes nanoshells and metal nanotubes to be especially promising for technical applications.

Besides the production technology of the objects with the specified optical properties, nanoshells have also other applications in engineering. Thus, sensitivity of their properties to the environment is used in chemical and biological sensors. Currently, nanoshells are most actively used in medicine [60, 65]. Due to the possibility of flexible control of the plasmon resonance frequency the most promising for medical applications are nanoshells whose plasmon resonance lies in the transparent region of biological tissues, from 700 to 1100 nm [60]. So, introduction of nanoshells into the tissue or blood with further fixation of their position using translucence in this range allows, for example, to study the tissue morphology [66]. Moreover, while using drugs as a nanoshell core, it is possible to destroy shell by a laser pulse with the wavelength belonging to this interval. Therefore, concentration necessary for the treatment is generated not in a whole patient organism, but in the right region.

First nanoshells were obtained in the middle of 90th [67] and intensively studied, initially experimentally [56-59]. In first nanoshells obtained in the laboratory dielectric core consisted of Au_2S, and shell – of gold [67]. Such nanoshell has significant restrictions on the configuration and, as a result, on the possibilities of the control of the plasmon resonance position [60], but the next obtained class of nanoshells wit the core of silicon dioxide and gold shell [68] removes the majority of these restrictions [60]. For such nanoshell of the radius of, for example, 60 nm with the change in the shell thickness from 5 nm to 20 nm, absorption peak of nanoshell is shifted from 1000 nm to 700 nm, for example, in works [57, 60].

Both experimental [57] and theoretical [56, 58] investigations of composite metal nanosystems are focused, first of all, on the study of the plasmon resonance. But in the frequency ranges, which are far from the plasmon resonance, contribution of individual transitions to the light absorption becomes the dominant one that makes necessary the investigation of single-electron components for a complete description of the optical properties of such nanoobjects. Calculation of the single-electron component of electrical absorption in quantum formalism (both for composite and continuous metal clusters) is usually based on one of two equivalent expressions for the components of diagonal form of the conductivity tensor

$$\hat{\sigma} = \begin{pmatrix} \sigma_1 & 0 & 0 \\ 0 & \sigma_2 & 0 \\ 0 & 0 & \sigma_3 \end{pmatrix}$$

(in such a way that coordinate axes coincide with the main tensor axes) in the Cartesian coordinate system (x_1, x_2, x_3): notation through the coordinate representation

$$\sigma_j = \frac{\pi e^2 \omega}{V_s} \sum_{(i,f)} \left| < i \mid x_j \mid f > \right|^2 f(E_i)(1 - f(E_f))\delta\left(E_f - E_i - \hbar\omega\right), \tag{1}$$

here $j = 1, 2, 3$; V_s is the volume of metal shell; ω is the electromagnetic wave frequency; i and f are the designations of the initial and final electron states, respectively; E_i and E_f are the electron energy in the initial and final states, respectively; $f(E)$ is the energy distribution function, summation is realized by all initial and final states; and notation through the momentum representation

$$\sigma_j = \frac{\pi e^2}{m_e^2 \omega^3 V_s} \sum_{(i,f)} \left| < i \left| \frac{\partial V}{\partial x_j} \right| f > \right|^2 f(E_i)\left(1 - f(E_f)\right) \delta\left(E_f - E_i - \hbar\omega\right), \tag{2}$$

here $V(\vec{r})$ is the potential energy of electron in the shell (see, for example, [4]). Works which study the contribution of individual transitions to the light absorption usually consider one of two opposite limiting cases. The first case corresponds to the situation when distances between quantum electron levels are very small in comparison with the energy of light quantum, and sum by discrete levels is replaced by an integral. The second limiting case corresponds to the situation when distance between energy levels is of the same order with the energy of light quantum, and only two-three levels are taken into account. Nevertheless, there are much more electron levels in a typical shell. On the other hand, one can not neglect the quantum effects connected with the discreteness of electron energy in the shell. The reason is in the fact that energy levels for thin shell become quasi-one-dimensional and distance between them increases. Effects of the oscillating dependence – both optical and electrical properties – on the thickness, which are connected with the discreteness of electron spectrum, were observed even for continuous metal nanowire [33]. For thin shells, obviously, such effects are much stronger. There are theoretical works which study the single-electron optical properties and describe such quantization effects – for spherical nanoshells and cylinder metal nanotubes [69] and for nanoshells in the form of the ellipsoid of revolution [70]. In these works in order to take into account discreteness of electron levels at calculation of the sum by electron states instead of the replacement of this sum by the integral, the following exact mathematical summation formula – the Poisson formula – is used:

$$\sum_{n=1}^{\infty} g(n) = \int_0^{\infty} dn \left(g(n) + 2\sum_{s=1}^{\infty} g(n)\cos(2\pi s n) \right), \tag{3}$$

here g is an arbitrary function of a natural argument n. Resultant expressions for the conductivity, indeed, are the oscillating functions of the frequency of incident light, and oscillation frequency and amplitude depend on the shell thickness (when the latter decreases, both oscillation frequency and amplitude increase).

There also theoretical works [44, 45] which study magnetic absorption in such systems. In this case one uses the kinetic approach for the description of the interaction between conduction electrons and electromagnetic wave. Usually, in works which study magnetic component of absorption in composite nanosystems, one find the solution of the Boltzmann kinetic equation for conduction electrons in shell metal, which are under the action of the vortex electrical field arising at application of the alternating magnetic field of electromagnetic wave. Description of the magnetic component of absorption is necessary to obtain the complete picture of the optical properties of such systems, since, as it was mentioned above, electrical component of absorption can be of the same order as the magnetic one or even much more than it [2].

As we have mentioned earlier, when studying composite metal nanosystems, attention is paid, first of all, to the phenomenon of plasmon resonance and system behavior in the frequency range near the resonant peaks. This is connected with the fact that the majority of technical applications of such systems is based on the phenomenon of plasmon resonance and sharp change in the optical properties in the corresponding frequency range. Intensive study of the behavior of both continuous and composite metal nanoparticles in the vicinity of the plasmon resonance led to the appearance of a new field of the research – plasmonics [65]. Plasmon resonance has been actively studied both experimentally and theoretically in composite metal nano-objects of different configuration. Thus, plasmon resonance in nanoshells of spherical form was studied in works [56-59], in nanoshells of ellipsoidal form – in work [71], in composite nanosystems of an arbitrary form – in [72]. It is possible to control the position of the plasmon resonance in nanoshells within wide boundaries (in particular, whole visible and infrared parts of the electromagnetic spectrum) changing the ratio of the dielectric core radius to the external radius of the nanoshell. Dependence of the position of the plasmon resonance in nanoshells on the core-to-shell radius ratio at the fixed chemical composition of the core and shell is studied, in particular, in [58]. Theory of plasmon hybridization developed, for example, in works [72, 73] is used for the theoretical description of the plasmon resonance in nanoshells and similar nanocomposites. According to this theory, plasmon arising on the external surface of the metal shell (spherical plasmon) and hollow plasmon arising on the internal surface of the shall interact with each other. The result of this interaction – hybridization – is the appearance of two plasmon modes with other (which are different from the output frequencies of spherical and hollow plasmons) frequencies, namely, bonding and antibonding which determine the plasmon properties of the system. In this case, only spherical and hollow plasmons with the same multipole index can be hybridized in symmetric nanoshell. And since for small nanoshell (whose typical size is much less than the wavelength) only dipole plasmons [72] can be excited, in the nanoshell spectrum, respectively, only two plasmon resonances (dipole bonding and dipole antibonding) are observed. Under some conditions, absorption peaks connected with these two plasmons are rather close; and we can speak about one absorption peak connected with the plasmon resonance (which is observed in many works). Frequencies of two resultant plasmons can be controlled in a wide range by the change in the external and internal shell radiuses at the fixed chemical composition of both shell and core.

We note that modern technologies allow to obtain and study both isolated nanoshell and ensemble of nanoshells with similar sizes (both core and shell) and orientation (for asymmetric nanoshells). This makes reasonable investigation of the optical properties of one nanoshell; and study of such systems is devoted to this problem.

4. NANOPARTICLES WITH SYMMETRY VIOLATION

Up to now, researchers have confined themselves by the case of symmetrical composite nano-objects – spherical nanoshells, cylindrical nanotubes, etc. – with one shell. However, new classes of composite nanosystems – nanoshells and metal nanotubes with lesser degree of symmetry – are described and studied in recent works. In particular, nanorice, nanoeggs, nanocups, and metal nanotubes with elliptical section belong to such systems. Nanorice [74] synthesized during last decade is the nanoshell in the form of considerably extended ellipsoid of revolution (large-to-small semi-axes ratio is equal to 4-7 for typical nanoparticles), see Fig. 3. Nanorice combines properties of local fields of high intensity typical for metal nanowires and nanotubes with flexible control of the position of plasmon resonance inherent for nanoshells. Presence of two plasmon resonances – longitudinal and transverse which are correspondingly controlled by the longitudinal and transverse sections of nanorice is the unique property of nanorice.

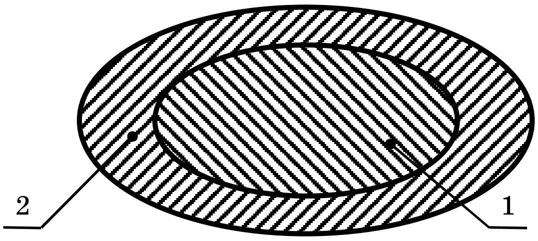

Fig. 3 – Nanorice in section: 1 – dielectric core; 2 –metal shell

Nanoegg [75-77] is the nanoshell with non concentric core, i.e. with the core shifted relative to the geometric center of the external boundary of the shell (see Fig. 4). Picture of the plasmon resonance in nanoegg is much more complicated than in usual symmetric nanoshell. If for symmetric nanoshell hybridization occurs only between spherical and hollow plasmons with the same angular momentum (and, as a result, because of the confinement of the dipole boundary one can observe only two plasmons in nanoshells), then at symmetry violation spherical and hollow plasmons of all multipole indexes interact with each other generating bonding and antibonding plasmons. Such additional interaction leads to stronger hybridization, larger shift of the position of the plasmon energy, and, the most important, to the appearance of mane plasmons in the vicinity of the dipole boundary [76].

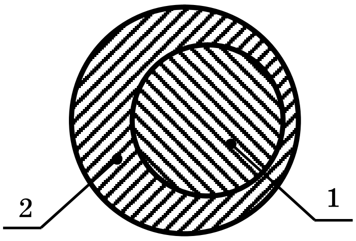

Fig. 4 – Nanoegg in section: 1 – dielectric core; 2 –metal shell

Nanocups [77-80] are similar to nanoeggs; they are nanoshells with non concentric core as well. However, shifting of the core in nanocups is larger than the shell thickness; and core partly falls outside its boundaries. Thus, shell in such nanoparticles does not cover the core in full forming a “cup” (see Fig.5). Many peculiarities inherent to nanoeggs [77, 78] are observed in the optical properties. Moreover, nanocups have unique feature especially promising for technical applications; they can be used as effective nanoantennas for reorientation of light incident on them in the direction which depends on the nanocup orientation. This property is conditioned by the possibility of generation in nanocup of the “magneton” plasmon (magnetic inductive resonance) with the frequency from the optical range and intensity which is comparable with the intensity of usual “electrical” plasmon [79].

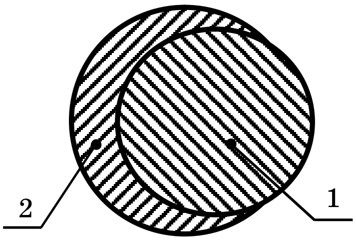

***Fig. 5** – Nanocup in section: 1 – dielectric core; 2 –metal shell*

5. MULTILAYER NANOSTRUCTURES

Besides usual symmetric nanoshells which consist of the dielectric core and one metal shell, multilayer nanoshells – the so-called nanomatryoshkas – become the subject of research in last years. Multilayer nanoshell is the composite nanoparticle consisting of the core and two or more shells of other substances [81-83]. Currently, nanomatryoshkas which contain metal and dielectric layers, for example, with Au-SiO_2-Au structure [81], are mainly under investigation. An absorption peak in multilayer nanoshells is shifted toward larger wavelengths in comparison with usual nanoshells. This makes nanomatryashkas promising for technical applications, since it is considerably difficult to obtain traditional nanoshells with plasmon resonance on these frequencies. Also, there is work devoted to the investigation of plasmon resosnance in nanomatryoshka with non concentric core, i.e. in multilayer nanoegg [84].

6. CONCLUSIONS

Thus, we have considered the present situation in physics of composite nanostructures with metal elements, basic approaches for description of such nanosystems, as well as main aspects of their technical applications. We have described the unique properties of such nanosystems providing wide possibilities for technical applications. As seen, the appearance of more and more complicated composite nanosystems offers the prospects for the production of sensors, materials with the specified properties, etc. This makes physics of composite nanostructures to be a promising area of physics of nano-objects, and investigation of such nanostructures becomes topical. Physics of composite nanostructures has been actively studied, and overview of the state-of-the-art presented in the work can be useful for researches.

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