New controllable electrophysics and optics of granulated thin films: the 4D laser-induced topological nanoclusters

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Abstract. The modeling of functional characteristics (electrophysics + optics) of topologically different nanocluster system deposited on solid surface is considered. Both energetic electronic levels and mobility of electrons are strongly depended on the cluster shape, their spatial distribution, and can be modified in a necessary way for possible applications in the photonics thin film elements.

Introduction
Laser-induced nanostructures and thin films with controllable topology depend on the laser pulses duration, and may be associated with the 4D-laser technology fabrication of new structures and materials. In our early experiments, we used a multilayer film formation by two-stage dynamic nonlinear process: first, laser-induced nanoparticles arise in colloidal solution by ms/μs-laser pulses (10⁶ – 10⁷ W/cm²) acting on the solid target in liquid; and second, the creation of thin films due to laser deposition of nanoparticles from colloid on a solid surface under cw-radiation [1,2]. By such technique we obtained a dramatic enhancement (in several orders) of electro- conductivity due to the variation of topological peculiarities of an induced nanocluster thin film system (cf. [2]). The process may be interpreted as a nonlinear phase transition in topological structure with bifurcations in functional properties.

In fact, the interaction effects of solid targets with laser pulses of different durations for obtaining various nanocluster structures can be viewed as the possibility of synthesizing 4D-objects by selected mechanisms. The result depends not only on the stationary topological/geometric parameters of the system, but also on the dynamic interactions in the system, which lead to different final stable structures. This is due to the fact, that the specific mechanisms of nanostructuring are activated for different durations of laser pulses. Therefore, time plays the role of a control parameter responsible for phase transitions, which can be seen in association with the spatial parameters when nanostructures of various dimensions arise, i.e. from quantum dots (0D) to 3D nanostructures. In addition, for short laser pulses we have non-equilibrium/transient phase transitions over the steady-state pressure-temperature (PT) phase-diagram according to the laser trajectory of nonlinear heating. Although the conventionality of this consideration is obvious (the equilibrium phase diagram cannot be used for
non-stationary processes), it allows us to discuss the current trends and clarify the basic physical picture [3]. The modeling of such processes, responsible for the controlled production of different types of topological structures, is a necessary preliminary stage for the possibility of verifying new physical principles to fabricate the thin film elements with controllable functional characteristics by femto-nanophotonics technique (cf [3]). We discuss the problem in present paper by the computer simulation approach for both electronic energetic states and electric current parameters of nanostructures.

1. Topological energetic states in nanocluster system

The simplest model of a nanocluster within its shell model is the representation for the first shells as a spherical potential well with infinite walls (we count energy from the bottom of the potential well). The radius of the sphere \( R_0 \) depends on the number of valence electrons of the Ne nanocluster as follows [4]: 
\[
R_0 = r_s d_0 N^{1/3} \epsilon,
\]
where \( r_s \) is the Wigner-Seitz parameter (inversely proportional to the cubic root of the electron density). In the case of potential surface deformation, it is assumed that the volume of the nanocluster does not change (topological structure) and its size is determined by the root-mean-square radius.

The stationary Schrödinger equation for the wave function \( \psi \) in this case (potential energy \( U = 0 \)) has the form:
\[
\mathbf{\nabla}^2 \psi + \frac{2m^*}{\hbar^2} E\psi = 0,
\] (1)
where \( m^* \) is the effective electron mass (proportional to the square of the nanocluster radius and depending on the dispersion relation for the nanocluster ensemble), \( \theta, \phi \) are the angles of the spherical coordinate system. Solution (1) has the following form:
\[
\psi(R_0, \theta, \phi) = C_{j_l l}(k_{n_l r} R_0) Y_{l m}(\theta, \phi),
\] (2)
where \( Y_{l m}(\theta, \phi) = \frac{1}{\sqrt{2\pi}} e^{im\phi} \Theta_{l m}(\theta) \) is a spherical function with separate dependencies on \( \theta \) and, \( \phi, j_l \).

\( j_l(z) = \sqrt{\frac{\pi}{2 R_0}} J_{l + 1/2}(z) \) is the spherical Bessel function. Moreover, \( j_l(k_{n_l r} R_0) = 0 \), i.e. we have the \( n \)th root of the Bessel function, \( k_{n_l r} R_0 \) is the wave vector for the transition between the shells \( n \rightarrow t \). The energy levels of electrons in this potential are as follows:
\[
E_{n_l}^{(0)} = \frac{\hbar^2 k_{n_l r}^2}{2m^*}.
\] (3)

The parameters of the deformation of the nanocluster potential are determined by the distribution of the charge density over the volume of the nanocluster. By analogy with the shell theory of the atomic nucleus (cf. [4]), the potential deformation is associated with the anisotropy of nanoparticles (electron distribution in the case of a nanocluster or nucleon in a nucleus), under which it is necessary to take into account the orbits of electrons that are not in the filled shells; the configuration of the filled shells is considered spherically symmetric. In the simplest case, we assume that the nanocluster is a uniformly charged ellipsoid of revolution with semiaxes \( a > b \). At the same time, the distribution density of electrons and holes is constant inside the ellipsoid, but is equal to 0 outside it (the “core” model with a sharp edge) – Fig. 1 [5].
Figure 1. Diagram of electron energy levels of a cluster with a undeformed spherical potential with a sharp edge (nomination – by standard quantum numbers).

Deformation of the energy potential in a nanocluster with a variable shape. A feature of the nanocluster structure is the need to take into account the different dependences of the electron density on the surface ($\sim R_0^2$) and in the volume ($\sim R_0^3$) of the nanocluster. We will now be interested only in the surface states of electrons (dominate for small $R_0$), since the electronic states in the volume of the nanocluster are assumed to be completely filled.

Consider the deformations of the spherical surface potential due to the change in the shape of the nanocluster in the following form:

$$R(\theta, \phi) = \begin{cases} R_0 \left(1 + k_1 \cos(\theta) \cos(p_1 \phi)\right) + k_2 \left(-1 \right)^{p_2 \left(\frac{p_2 - 1}{2}\right) \mod(p_2 + 1, 2) \sin(p_2 \theta) + \mod(p_2 + 1, 2) \cos(p_2 \theta)\right), & p_1 \neq 0, \\ R_0 k_2 \left(-1 \right)^{p_2 \left(\frac{p_2 - 1}{2}\right) \mod(p_2 + 1, 2) \sin(p_2 \theta) + \mod(p_2 + 1, 2) \cos(p_2 \theta)\right), & p_1 = 0. \end{cases}$$

(4)

where $p_2 = 0, 1, 2, \ldots$ is the number of zenith (in "longitude") distortion; $p_1 = 0, 1, 2, \ldots$; $p_2$ is the number of azimuthal distortion (in “latitude”), $k_1$ and $k_2$ are the magnitude of the corresponding distortion. Several selected results in accordance with formula (4) are shown in Fig. 2a. In the case the energy levels are split as follows – see Fig. 2b.

Figure 2. (a) Several topological structures controlled by the key parameters (see text). (b) Spectra of deformed nanoclusters. The split energy levels are shown for different distortions when $k_1 = k_2 = \alpha$ and variable magnitudes of $p_1, p_2$. Numerical parameters are: valent electron density $N_e = 40$; Wigner-Seitz parameter ($\sim 1/N_e^{1/2}$) $r_s = 2.1$; electron effective mass $m_e^* = 1.4m_e = 0.708M_e$.

Let’s move to the new coordinates associated with the boundary conditions, i.e. we deform the coordinate system so that the potential in the new coordinates is again a sphere [4].

$$\tilde{r} = r \left(1 + k_1 f_{p_1}(\theta, \phi) + k_2 f_{p_2}(\theta, \phi)\right)^{-1}, \tilde{\theta} = \theta, \tilde{\phi} = \phi,$$

(5)

where the parameter $f$ determines the number of corresponding distortions in angular variables. Laplacian in deformed coordinates (5) takes the following form:

$$\Delta u = \tilde{\Delta} u - \frac{f(f+2)}{(1+f)^2} \Delta u - \cot\theta \frac{\partial f}{\partial \theta} \frac{\partial u}{\partial \theta} + \frac{1}{r^2(1+f)^3} \left( \frac{\partial^2 u}{\partial \theta^2} + \tilde{r} \left( \frac{2}{1+f} \frac{\partial^2 u}{\partial \theta \partial \phi} \right)^2 - \frac{\partial^2 u}{\partial r \partial \theta} - 2f \frac{\partial f}{\partial \theta} \frac{\partial^2 u}{\partial r^2} \right) +$$

$$+ \frac{1}{r^2(1+f)^3(\sin\theta)^2} \tilde{r} \left( \frac{\partial^2 u}{\partial \phi^2} \right)^2 + \tilde{r} \left( \frac{2}{1+f} \frac{\partial^2 u}{\partial \phi \partial \theta} \right)^2 - \frac{\partial^2 u}{\partial \phi \partial \theta} - 2f \frac{\partial f}{\partial \phi} \frac{\partial^2 u}{\partial \phi \partial \theta} \right).$$

(6)

Linearization with respect to the small deformation parameter of a spherical nanocluster.

Linearize potential perturbations by a small parameter of deformation $\alpha$.
\[
\frac{1}{(1+f)} \approx 1 - f, \quad \frac{1}{(1+f)^2} \approx 1 - 2f, \quad \left( \frac{\partial f}{\partial \theta} \right)^2 \approx 0.
\]

In the new coordinates in the approximation of small deformations, the Hamiltonian takes the following form:

\[
\delta \hat{H} = \delta \hat{H}_0 + \delta \hat{H}_1 + \delta \hat{H}_2 + \delta \hat{H}_3,
\]

where we have:

\[
\delta \hat{H}_1 = -2 \left( k_1 f_{p_1} + k_2 f_{p_2} \right) \hat{H}_0 + \frac{\delta \hat{H}_2^2}{\hat{F}},
\]

\[
\delta \hat{H}_2 = -2 \left( k_1 f_{p_1} + k_2 f_{p_2} \right) \hat{H}_0 + \frac{\delta \hat{H}_3^2}{\hat{F}}.
\]

In the first approximation of perturbation theory, axial deformations do not contribute to energy. Therefore, we first consider axially symmetric deformations, i.e. we consider \( p_1 = 0 \). Then the disturbed part of the Hamiltonian will look like:

\[
\delta \hat{H} = -2f \hat{H}_0 + \frac{\hbar^2}{2m^*} \frac{\partial^2 \hat{F}}{\partial \hat{F}} \left\{ \sin^2 \frac{\theta}{\hat{F}} \frac{\partial \hat{F}}{\partial \cos \theta} \right\} + 2 \left\{ \sin^2 \frac{\theta}{\hat{F}} \frac{\partial \hat{F}}{\partial \cos \theta} \right\} \frac{\partial \hat{F}}{\partial \cos \theta}.
\]

Relations (8a – 8d) completely determine the energy picture for an isolated cluster in this approximation. Further, we consider according to the perturbation theory [4]:

\[
E_{n,l,m} = E_{n,l}^{(0)} + \delta E_{n,l,m}.
\]

The energy correction is determined by the diagonal matrix element of the perturbed part of the Hamiltonian in the basis of the unperturbed Hamiltonian:

\[
\delta E_{n,l,m} = \int d\phi \int_0^{2\pi} \int_0^{\hat{F}} \psi_{n,l,m}^*(r, \theta, \phi) \delta \hat{H} \psi_{n,l,m}(r, \theta, \phi) r^2 \sin \theta dr d\theta d\phi.
\]

The last term in (8d) makes a small contribution to the diagonal terms of the perturbation operator (cf. [6]), therefore

\[
\delta E_{n,l,m} = -2E_{n,l}^{(0)} \int d\phi \int_0^{2\pi} \int_0^{\hat{F}} \sin \theta \psi_{n,l,m}^*(r, \theta, \phi) \left\{ \left( k_1 f_{p_1} + k_2 f_{p_2} \right) \right\} \psi_{n,l,m}(r, \theta, \phi) r^2 \sin \theta dr d\theta d\phi.
\]

The expression (11) in the case of \( p_2 = 2; \ p_1 = 0 \) leads to the relation:

\[
\delta E_{n,l,m} = \frac{16 E_{n,l}^{(0)} (l+1)l!}{(2l+1)^2(1-m)!} \left\{ \left( l+1-m \right) \left( 1-m \right) \right\} + \frac{1}{(2l-1)^2(1-m)!} \frac{1}{(2l+1)(1-m)!}
\]

The resulting splitting of the energy levels (for Fig. 1) due to the different magnitude of deformations (\( \alpha \)) of the initially spherical cluster are shown in Fig. 3. Significant changes in the energy spectrum are seen, which makes it possible to speak of the topological spectral characteristics of nanoclusters of various types.

![Figure 3](image_url)

**Figure 3.** The splitting of energy levels (fine structure) with different strains for \( k_1 = 0, \ k_2 = \alpha \): (a) \( p_2 = 2; \ p_1 = 0 \); (b) \( p_2 = 4; \ p_1 = 0 \); (c) \( p_2 = 6; \ p_1 = 0 \). In the linear approximation. Options: \( N_\gamma = 40, r_\gamma = 2.1, m_\gamma = 1.4 m_e = 0.708 \text{MeV} \).

**Model of a nanocluster as a harmonic oscillator.** Hamiltonian of a harmonic oscillator in the case of spherical symmetry (r coordinate) is written in the form [4]:

\[
H = -\frac{\hbar^2}{2m_e} \nabla^2 + \frac{m_e \omega^2 r^2}{2}.
\]
In this case, \( m^* e \) is the effective electron mass (it is in our case proportional to the square of the nanocluster radius), the oscillator frequency is determined by the relation:

\[
\omega = \frac{\sqrt{2E_f}}{\sqrt{m^*(E_f)R_0^2}},
\]

where \( E_f \) is the Fermi level, \( R_0 \) is the radius of the sphere.

Then the Schrödinger equation with the wave function \( \psi \) and the Laplacian in spherical coordinates is represented in the standard form:

\[
\nabla^2 \psi = \frac{2m^*\hbar^2}{\hbar^2} \left( E - \frac{m^*\omega^2 r^2}{2} \right) \psi = 0, \quad \psi(\infty, \theta, \phi) = 0.
\]

The solution of equation (16) is reduced to the expression:

\[
\psi(r, \theta, \phi) = C \frac{r^l}{l^2_\mu} \mu^{-1/4} \exp \left( -\frac{r^2}{2l^2_\mu} \right) L^\mu_n \left( \frac{r^2}{l^2_\mu} \right) Y_{l,m}(\theta, \phi),
\]

where \( Y_{l,m}(\theta, \phi) = \frac{1}{\sqrt{2\pi}} e^{im\phi} \Theta_{lm}(\theta) \) is the ball function, \( l^\mu_n \) is the adjoined Laguerre polynomial, \( \Theta_{lm}(\theta) = \sqrt{\frac{(2l+1)(l-m)!}{2(l+m)!}} P^m_l(\cos(\theta)) \), where \( P^m_l \) is associated Legendre polynomials, and the normalization of the wave function is determined by the parameter \( |C|^2 = \frac{n!}{l^2_\mu \Gamma(2\mu+n+1)} \) with the parameter \( \mu = (2l+1)/4 \), \( l^2_\mu = \hbar/m^*\omega, \Gamma \) – is the gamma function. The energy levels of electrons in such a potential depend on the quantum numbers \( n, l \):

\[
E^{(0)}_{n,l} = \hbar \omega \left( 2n + l + \frac{3}{2} \right),
\]

where the combination \( N = (2n + l) \) we will call the main quantum number, which determines the number of excited oscillatory modes (the number of the electron shell in the cluster).

In this model, the energy levels are degenerated by the orbital (\( l \)) quantum number – for the first two levels we have: \( E_{13} = E_{21}, E_{14} = E_{22}, E_{12} = E_{20} \).

Consider the surface deformation potential of the following type:

\[
R(\theta, \phi) = R_0 \left( 1 + f(\theta, \phi) \right),
\]

in which the function \( f(\theta, \phi) \) determines the specific type of deformation of the sphere. Then the harmonic potential can be represented by the relation:

\[
V = \frac{m^*\omega(r)^2 r^2}{2},
\]

where \( \omega(r) = \frac{1}{(1+f)} \omega_0 \).

In this case, the perturbation operator takes the following form (deformation of the cluster shape) determines the form of the interaction potential:

\[
V(r) = -\frac{f(2+f)}{(1+f)^2} V_0.
\]

Let us turn as well to new coordinates, in which the shape of the distorted cluster appears again spherical.

Then, the wave functions of the considered, for example, terms with initially degenerate energy levels \( E_{13} \) and \( E_{21} \), respectively, in linear combination can be presented as:

\[
\psi = c_1 \psi_1 + c_2 \psi_2.
\]
\[(H + V)\psi = E\psi.\]  \tag{22}

Substituting (21) into (22) and we get
\[c_1(E_1 + V - E)\psi_1 + c_2(E_2 + V - E)\psi_2 = 0,\]  \tag{23}
where \(c_{1,2}\) – some coefficients for which we obtain two algebraic equations
\[c_1(E_1 + V_{11} - E) + c_2V_{12} = 0,\]
\[c_1V_{21} + c_2(E_2 + V_{22} - E) = 0.\]  \tag{24}

It follows from the properties of the hermiticity of the perturbation operator that \(V_{12} = V_{21}^*\), and according to the compatibility condition of equations (24), their determinant should be zero:
\[
\begin{vmatrix}
E_1 + V_{11} - E & V_{12} \\
V_{21} & E_2 + V_{22} - E
\end{vmatrix} = 0.
\]  \tag{25}

Expanding the determinant, we have for splitting the initially degenerate energy levels:
\[E = \frac{1}{2}(E_1 + E_2 + V_{11} + V_{22}) \pm \frac{1}{4}(E_1 - E_2 + V_{11} - V_{22})^2 + |V_{12}|^2.\]  \tag{26}

For definiteness, the deformation of the harmonic potential will be assumed to be quadrupole:
\[f = \alpha P_2 (\cos \theta)\] with the deformation parameter \(\alpha\) and the Légandre polynomial \(P_2\). In Fig. 4 it is shown the cluster energy spectrum for the case.

**Figure 4.** Lifting the degeneration of the levels in the quadrupole deformation of the harmonic potential \(f = \alpha P_2 (\cos \theta)\); the explanations are in the text.

2. **Computer simulation for electroconductivity and optical transmission in granulated structures. Functional properties**

We discuss both some possible tunnel mechanisms being responsible for a high electroconductivity and the features of obtaining a hopping conductivity in such inhomogeneous thin film surface structures (with the thickness of up to 100 nm) when the charged particles are propagating along the boundary surface of granulated structures. The simulation results are shown in Fig.5. It is now of fundamental importance for us, that the movement of electrons in a nanostructured system under external voltage leads to electrical conductivity. Thus, the problem can be reduced to a dynamic model of the electrons motion, being nonlinear in a general case (e.g. with a hopping behavior) but may be discussed the problem in accordance with the path integrals over quantum trajectories (cf. [7]).

The calculated optical transmission for different conditions is shown in Fig.6.
Figure 5. Volt-Ampere Characteristics (vertically axis – current, horizontally axis – voltage) of thin films when both (a) the distance (S) between the conducting islands changes and (b) radius (R) of deposited particles changes. Different values of the parameters in arbitrary units are directly indicated in the pictures.

Figure 6. Modeling (layer by layer, arbitrary units) of both the surface images for thin films at different temperatures T (a) and the optical transmission spectra of deposited bimetallic films (b) for noble metal compositions. 1. Au: Ag (concentration 1:1), particle diameter (D) – 50 nm, one layer, the distance between the particles (gap) – 5 nm; 2. Au: Ag (1:1), D – 10 nm, five layers, a gap of 4 nm; 3. Au: Ag (1:1), D – 10 nm, five layers, a gap of 2 nm. The FDTD method has been used for the modeling.

In terms of discussion, first, it is obvious that electro-physics strongly depends on the topology of nanostructured films on the solid surface. Such nonlinear electrical transport properties are due to the quantum correlated states resulting in the tunnel and hopping electrical conductivity. There is a competition between the bulk and surface electrical conductivity contributions, controlled (to a great extent) by a deposited cluster topology/temperature [8].

Second, optical spectra of thin films (produced layer by layer) result in some new physical states of the system, particularly, in optical response (cf. [9]).

The optical transmittance dependence based on the Bouguer-Lambert approach. With the passage of optical radiation through an island nanofilm is its weakening due to reflection and absorption processes, therefore it is necessary to evaluate the transmittance values of $T_\lambda$ in depending on the film thickness. In the case of well-formed island films when the distance between the islands is much less of their medium size, we use the Bouguer – Lambert law for the case strong absorption in the absence of multiple internal reflections:

$$T_\lambda = (1-R)^2 \exp(-\alpha_\lambda d) \quad (27)$$

where $R = (n-1)^2/(n+1)^2$ – reflection coefficient from the surface; $n$ – refractive index of the island film material; $\alpha_\lambda$ – coefficient absorption; $d$ – film thickness. For the case of rarefied island films, a significant contribution to the transmittance contributes to the substrate on which it is formed such structure:
\[ T_s = (1-R_1)(1-R_2)(1-R_3)\exp(-\alpha \lambda d), \]
\[ R_1 = (n_{f}-1)^2/(n_{f}+1)^2, \quad R_2 = (n_{f}-n_{s})^2/(n_{f}+n_{s})^2, \quad R_3 = (n_{s}-1)^2/(n_{s}+1)^2, \] 

where \( n_f \) is the refractive index of the film material; \( n_s \) – indicator refraction of the substrate material [10].

Fig. 7 shows the dependence of the transmittance on film thickness when varying to 20 nm for the case well-formed film (27) and the case of sparse islands, taking into account glass substrates (28) \( n_s \) = 1.5 at wavelength of 1.06 μm, temperature 300 K, \( n = n_f = 6, \alpha = 0.5 \) [11].

\[ \text{Figure 7. The dependence of the transmittance on the film thickness} \]

Obviously, the transmittance value for the case of good formed film is higher than rare, which is explainable the influence of reflections and scattering at the interface of the film and the substrate.

The proposed model will allow in the first approximation to estimate optical properties of the films obtained [1], as well as the thickness formed structure by measured optical transmission characteristics.

Conclusions

The topology peculiarities of the granulated metallic film deposited on dielectric substrates have been analyzed by computer simulation approach for clustered metallic structures. The functional characteristics (optics+electrophysics) may be varied in necessary direction by modification of the 3D-cluster shape in laser experiment. The observed phenomena give us an opportunity to establish the basis of new physical principles to create the functional elements for topological photonics.

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