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Research Article

Evaluation of the Efficiency of Interparticle Interactions in Nanosystems

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Wide range of applications of nanoparticles causes the need to study their properties, and the influence of the interparticle interaction on the formation of the nanosystem properties is a well-known experimental phenomenon. The aim of this work is to study the influence of interparticle interactions on the properties of nanosystems theoretically. The influence of the interparticle interaction was simulated based on the near-field interaction potential and local field distribution. The local field distribution in the system was calculated using the Green function method and the concept of the effective susceptibility. The results show that interaction between nanoparticles can be neglected if the distance between them is bigger than the critical one. Expressions for evaluation of the efficiency of the interparticle coupling were proposed and compared with the existing experimental results. The results of the simulation are in good agreement with the measured values of the critical interparticle distance. The approach may be useful for simulation of interactions in the system of many nanoparticles and for engineering of nanostructures for different applications.

1. Introduction

Nowadays, nanoparticles and their interactions with other objects are actively used in various applications, e.g., films epitaxy [1], gas sensors [2-4], high-temperature superconductors [5-7], and luminescent materials development [8, 9]. But applications of nanoparticles in biomedicine are especially relevant, e.g., drug delivery [10], cancer therapy [11], enhanced tomography [12], and antiviral and antimicrobial therapy [13, 14]. The nanoparticles are usually in colloidal solutions; moreover, the task of making solutions of nanoparticles with their high concentration is of particular interest [15]. Obviously, in such solutions, there are interactions between nanoparticles that affect the properties of the entire system. Interparticle interactions manifest themselves in the formation of properties of nanosystems, and in some cases, this effect is undesirable. For example, high concentrations of nanoparticles on the plasmon surfaces cause a shift of the plasmon resonance peak [16]. This is due to the screening effect of one nanoparticle on others at a sufficiently close distance between them.

This is not unitary phenomenon. The influence of interparticle interaction on the formation of material

properties is widely observed for optical properties of materials, such as light absorption and excitation of surface plasmon resonance [16, 17], Raman scattering [18–20], and superconductive [21], rheological [22, 23], magnetic [24, 25] and catalytic [26] properties. All the papers [16–29] showed experimentally that the decrease in the distance between the nanoparticles changes the material properties.

Similar effects are also observed in biomedical applications of the nanoparticles. For example, the nanoparticles activity against viruses decrease at their high concentrations, which was observed for Au nanoparticles coated with the SiO₂ shell against adenovirus [30] and for Ag nanoparticles against Hepatitis B virus [31]. Basing on the assumption [30] about the physical mechanism of the antiviral action of nanoparticles, it can be assumed that, at high nanoparticles concentration, the compensation of their action due to interparticle interactions may occur. All of the mentioned above assumptions indicate the need to consider the interparticle interactions while designing the nanostructures. It may be conducted by evaluating the efficiency of the interparticle interactions.

Interparticle interaction is also an interesting phenomenon for simulation and theoretical study. However, in

systems of many nanoparticles, the number of interactions that should be taken into account arises with the increase in the number of the nanoparticles. Consequently, simulating the properties of their interaction becomes rather complicated. Evaluation of the interparticle interactions efficiency may allow simplifying theoretical consideration of systems of more than two nanoparticles.

Hence, in the work, we answer the following questions: is the action of one nanoparticle on another one significant just for rather small distance between them, and if so, how can this distance be easily evaluated?.

2. Materials and Methods

In order to achieve the goal of the work, we need to simulate the interaction between two or more nanoparticles. The interaction between nanoparticles at the absence of the external action is due to forces, which are similar to van der Waals forces [32]. There are three well-known types of van der Waals interactions which are due to orientation, induction, and dispersion interaction between the nanoparticles. The most common case of interaction is the dispersion interaction. The dispersion interaction exists between two nonpolarized nanoparticles at the absence of the external field [33]. Its mechanism can be easily explained for the case of two point-like nanoparticles. In average, the particles dipolar moments are equal to zero, but at the every time point, there are dipolar moment fluctuations. This nonzero dipolar moment is the source of the electric field acted on the second particle and generated the dipolar moment of the second particle. Hence, this leads to the electrostatic interaction between two dipoles, and the energy of this interaction is proportional to the square local electric field $U \sim E^2 \sim (1/r^6)$ [14]. The reason for such fluctuations may be due to the electron fluctuations or vacuum field fluctuations occurred in the ground state of the system. This is attractive interaction. However, there is also a repulsive interaction in such systems, which is the cause of formation of the well-known Lennard-Jones potential. Its formation may be caused by the rather high nonlinear polarizabilities of the nanoparticles [34] or by the mode softening [35]. The model of interaction between two nanoparticles proposed in [34] takes into account the nanoparticles shapes and sizes, their linear and nonlinear polarizabilities, and selfconsistence of the local field. It should be noted that results obtained in [34] need rather complicated calculations. As we want to analyze results of simulations, it would be better to obtain analytical results. Furthermore, taking into account the nonadditivity of van der Waals forces [36], a theoretical study of systems of many nanoparticles seems like too complicated using the model in [34]. Hence, for our calculations, we use the Green function method [37], as it allows obtaining analytical expression for the local field, the concept of the effective susceptibility [38], as it allows describing the nanoparticle response for the external field, and the quasi-point nanoparticles approximation [39], for simplification of the model in [34].

We study two types of systems. The simplest nanosized system is the system of two identical nanoparticles

(Figure 1). It should be noted that the quasi-point-like nanoparticles approximation [39] can be used for spherical nanoparticles. Hence, the first system under consideration is the system of two spherical nanoparticles with a shell located in the homogeneous medium. In contrast to the model in [34], we add the shells to the nanoparticles. It is caused by the fact that many nanoparticles in use are not homogeneous and usually there is a stabilizer, which composes the layer around the nanoparticles in solution.

It was mentioned above that dispersion interaction is in the ground state of the system; hence, we start from the condition of the system ground state. This means there must be the minimum of the system free energy. We apply the Taylor expansion of the nanoparticles energy as in [34], which gives the following expression for the system free energy:

$$F = \left\{ \frac{1}{2} \frac{1}{\varepsilon_0} \lambda_{ij}^a P_i^a \left(\mathbf{R}_a \right) P_j^a \left(\mathbf{R}_a \right) + \frac{1}{4} \frac{1}{\varepsilon_0} \beta_{ijrs}^a P_i^a \left(\mathbf{R}_a \right) P_j^a \left(\mathbf{R}_a \right) P_r^a \left(\mathbf{R}_a \right) P_s^a \left(\mathbf{R}_a \right)$$

$$- P_j^a \left(\mathbf{R}_a \right) E_j^a \left(P_i^a, P_i^b, \mathbf{R}_a \right) \right\} + \left\{ \frac{1}{2} \frac{1}{\varepsilon_0} \lambda_{ij}^b P_i^b \left(\mathbf{R}_b \right) P_j^b \left(\mathbf{R}_b \right)$$

$$+ \frac{1}{4} \frac{1}{\varepsilon_0} \beta_{ijrs}^b P_i^b \left(\mathbf{R}_b \right) P_j^b \left(\mathbf{R}_b \right) P_r^b \left(\mathbf{R}_b \right) P_s^b \left(\mathbf{R}_b \right)$$

$$- P_j^b \left(\mathbf{R}_b \right) E_j^b \left(P_i^a, P_i^b, \mathbf{R}_b \right) \right\} + \int_{\mathbb{R}^3} d\mathbf{R} \, \varepsilon \varepsilon_0 \frac{E_i^2 \left(P_i^a, P_i^b, \mathbf{R} \right)}{2},$$

$$(1)$$

where $P_i^a(\mathbf{R}_a)$ and $P_j^b(\mathbf{R}_b)$ are the dipole moments of the nanoparticle a or b, λ and β are tensors of the linear and nonlinear susceptibilities, and E is the local field. Here expressions in brakes describe the energy of each nanoparticle, and the last term is the energy of the field. However, it does not influence on the interaction, and it will be omitted during further considerations. The last term in each bracket is the energy of interaction. We are looking for the ground state using the following conditions:

$$\frac{\partial F}{\partial P_i^{(a)}} = 0,$$

$$\frac{\partial F}{\partial P_j^{(b)}} = 0,$$

$$A = \frac{\partial^2 F}{\partial P_i^{(a)} \partial P_j^{(a)}},$$

$$B = \frac{\partial^2 F}{\partial P_i^{(a)} \partial P_j^{(b)}},$$

$$C = \frac{\partial^2 F}{\partial P_i^{(b)} \partial P_j^{(b)}},$$

$$AC - B^2 > 0, \quad A > 0,$$
(2)

and for minimization via the dipole moments, we express the local field in the frame of the Green function method:

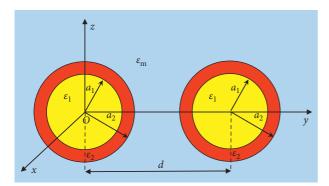


FIGURE 1: System of two nanoparticles.

$$E_{j}^{\alpha}(\mathbf{R}_{\alpha}) = E_{i}^{ex}(\mathbf{R}_{\alpha}) + \frac{k_{0}^{2}}{\varepsilon_{0}} \int_{V_{a}} d\mathbf{R}_{a}' G_{ji}(\mathbf{R}_{\alpha}, \mathbf{R}_{a}') P_{i}^{a}(\mathbf{R}_{a})$$
$$+ \frac{k_{0}^{2}}{\varepsilon_{0}} \int_{V_{b}} d\mathbf{R}_{b}' G_{ji}(\mathbf{R}_{\alpha}, \mathbf{R}_{b}') P_{i}^{b}(\mathbf{R}_{b}), \quad \alpha = a, b,$$
(3)

where the first term is the external field of vacuum fluctuations, and the two other terms describe the field scattering from the nanoparticles. The quasi-point-like nanoparticle approximation allows the following simplification:

$$E_{j}^{\alpha}(\mathbf{R}_{\alpha}) = E_{i}^{ex}(\mathbf{R}_{\alpha}) + \frac{k_{0}^{2}}{\varepsilon_{0}} P_{i}^{a}(\mathbf{R}_{a}) \int_{V_{a}} d\mathbf{R}_{a}' G_{ji}(\mathbf{R}_{\alpha}, \mathbf{R}_{a}')$$

$$+ \frac{k_{0}^{2}}{\varepsilon_{0}} P_{i}^{b}(\mathbf{R}_{b}) \int_{V_{b}} d\mathbf{R}_{b}' G_{ji}(\mathbf{R}_{\alpha}, \mathbf{R}_{b}'), \quad \alpha = a, b.$$

$$(4)$$

And as the nanoparticles are in the homogeneous medium, the Green function is the electrodynamic Green function for the free space [40]:

$$G_{ij}(\mathbf{R}, \mathbf{R}', \omega) = \frac{1}{4\pi k_0^2 R^3} \cdot \left[\delta_{ij} - 3e_i^{(R)} e_j^{(R)} \right],$$

$$R = \left| \mathbf{R} - \mathbf{R}' \right|, \quad e_i = \frac{\mathbf{R}_i - \mathbf{R}'_i}{R}.$$
(5)

Solutions of equation (2) were found by the method of successive approximations and their application to the interaction potential:

$$U(z) = W(P(z)) - W(P(z = \infty)), \tag{6}$$

where $W(P(z)) = -P \cdot E$ is the energy of interaction and gives us the following expression:

$$U(d) = \frac{M}{d^{12}} - \frac{N}{d^6}, \quad d = \left| \mathbf{R}_a - \mathbf{R}_b \right|, \tag{7}$$

where coefficients M and N are calculated in the frames of the proposed model (Supplementary Materials (available here)). Expression (7) allows us to determine the equilibrium distance in the system. It should be noted that the external field is the field of vacuum fluctuations and is defined by the field quantum commutators [33]. When the distance between the nanoparticles is known, we can calculate the local field distribution in the system in Figure 1 as follows [41]:

$$E_{i}(\mathbf{R}) = E_{i}^{0}(\mathbf{R}) + k_{0}^{2} \int_{V_{1}} d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}') X_{jk}^{(a)}(\mathbf{R}') E_{k}^{0}(\mathbf{R}')$$
$$+ k_{0}^{2} \int_{V_{2}} d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}') X_{jk}^{(b)}(\mathbf{R}') E_{k}^{0}(\mathbf{R}'),$$
(8)

where effective susceptibilities of the nanoparticles are

$$X_{ij}^{a,b}(\mathbf{R}) = \chi_{ik}^{a,b}(\mathbf{R}) \left[\delta_{kj} + \chi_{kj}^{a,b} \frac{(\mathbf{R})}{3} - k_0^2 \int_{V_{b,a}} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{b,a}(\mathbf{R}') \right]^{-1},$$
(9)

where χ_{lj} is the tensor of the dielectric susceptibility of the material.

However, nanoparticles are not of the same size, and there is some size range. This means that, in the solution, the small nanoparticle may be located beside the big one. This case may also be realized when using the nanoparticles for antiviral and antimicrobial therapy as the viruses and bacteria are much bigger then nanoparticles. Consequently, it is interesting to study the nonsymmetric system of three nanoparticles of different sizes (Figure 2). The distance between the small nanoparticle and the big one may be evaluated from (7). The positions of the big and one small nanoparticle are defined, whereas the position of the second small nanoparticle is defined by the angle α (Figure 2).

In this case, the local field can be found as

$$E_{i}(\mathbf{R}) = E_{i}^{0}(\mathbf{R}) + k_{0}^{2} \int_{V_{b}} d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}') X_{jk}^{(b)}(\mathbf{R}') E_{k}^{0}(\mathbf{R}')$$

$$+ k_{0}^{2} \int_{V_{p1}} d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}') X_{jk}^{(b)}(\mathbf{R}') E_{k}^{0}(\mathbf{R}')$$

$$+ k_{0}^{2} \int_{V_{p2}} d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}') X_{jk}^{(3)}(\mathbf{R}') E_{k}^{0}(\mathbf{R}'),$$
(10)

where indexes b, p1, and p2 correspond to the big nanoparticle and two small nanoparticles accordingly, with

$$X_{ij}^{b}(\mathbf{R}) = \chi_{ik}^{b}(\mathbf{R}) \left[\delta_{kj} + \chi_{kj}^{b} \frac{(\mathbf{R})}{3} - k_{0}^{2} \int_{V_{p1}} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{p1}(\mathbf{R}') - k_{0}^{2} \int_{V_{p2}} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{p2}(\mathbf{R}') \right]^{-1},$$

$$(11)$$

$$X_{ij}^{p1,p2}(\mathbf{R}) = \chi_{ik}^{p1,p2}(\mathbf{R}) \left[\delta_{kj} + \chi_{kj}^{p1,p2} \frac{(\mathbf{R})}{3} - k_0^2 \int_{V_b} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^b(\mathbf{R}') - k_0^2 \int_{V_{p2,p1}} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{p2,p1}(\mathbf{R}') \right]^{-1}.$$
(12)

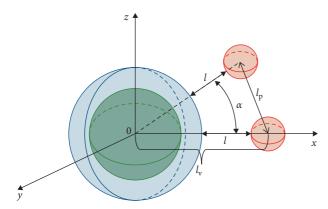


FIGURE 2: System of three nanoparticles.

Let us analyze (11) and (12). Two last terms in each bracket (the integrals) depend on the Green function of the system and on the nanoparticle volume and dielectric function. The Green function depends on the distance as $\sim (1/d^3)$. Consequently, for some system geometry, there may be the case when one of integrals is much smaller compared to another one. This may be at rather high values of angle α . Hence, if the distance is large enough and the interaction is weak, we may assume the following equation instead of (12):

$$X_{ij}^{p1,p2}(\mathbf{R}) = \chi_{ik}^{p1,p2}(\mathbf{R}) \left[\delta_{kj} + \chi_{kj}^{p1,p2} \frac{(\mathbf{R})}{3} - k_0^2 \int_{V_b} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^b(\mathbf{R}') \right]^{-1}.$$
 (13)

A compare of calculations using (12) and (13) allows us revealing the influence of the small nanoparticles interaction on the local field enhancement and understanding if the simplification may be applied.

3. Results and Discussion

We calculate the system in Figure 2 with the following parameters: the big nanoparticle is of 30 nm inner radius and has the shell of 10 nm thickness with an inner dielectric constant value $\varepsilon_1 = 1.2$ and the outer one $\varepsilon_1 = 1.8$ (which can be a model of the adenovirus) and the small Au nanoparticles of 5 nm inner radius with the 2.5 nm SiO₂ shell. The distance between the small and big nanoparticles was evaluated from (7) and is nearly 5 nm. The bright effect of the interparticle interaction can be observed for the case of $\alpha = 20^{\circ}$ (Figures 3(a) and 3(b)). The other results for this system demonstrate the applicability of the proposed simplification, whereas there is no difference between two results with and without taking into account the interparticle interaction (Figures 3(c)-3(f)). Calculations for the system of the same big nanoparticle with two homogeneous Au nanoparticles with a diameter of 5 nm give the similar result (Figure 4); namely, the results in Figures 3 and 4 demonstrate that interaction between small nanoparticles (which is rather intensive for

small distance between them) due to the self-consistence decreases the local field enhancement effect observed for one nanoparticle or two far located nanoparticles. The nature of this phenomenon is of particular interest; however, this is the task for further studies. In the frame of the present work, the mentioned observation gives us the possibility to make a proposal to neglect this interaction.

Let us obtain the expressions for both systems in Figures 1 and 2. The way for simplification of calculations was described above:

$$\int_{V_{p1}} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{p1}(\mathbf{R}') \ll \int_{V_b} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{b}(\mathbf{R}').$$
(14)

Taking into account the spherical symmetry of particles, in order to obtain an analytical expression, the integral can be approximately written as follows:

$$\int_{V_b} d\mathbf{R}' G_{kl}(\mathbf{R}, \mathbf{R}') \chi_{lj}^{b,p1}(\mathbf{R}')$$

$$\approx V_{b,p1} G_{b,p1}(\mathbf{R}_{p2}, \mathbf{R}_{b,p1}) \chi_{lj}^{b,p1}(\mathbf{R}_{b,p1}),$$
(15)

where \mathbf{R}_b is the big nanoparticle center. Using the Green function for free space (5) and the designation $l_p = 2l \cdot \sin{(\alpha/2)}$, we obtain the condition of weak interparticle interaction in the system in Figure 2:

$$l_p \gg l \frac{r_{p1}}{r_h} \left(\frac{\chi_{p1}}{\chi_h}\right)^{1/3},\tag{16}$$

where the sign \gg should be interpreted taking into account that expression (15) contains the distance in the third degree. Suppose that the left part of (14) is at least two orders of magnitude smaller than the right part of (14). Then, we may assume that the distance which is 6 times larger than the critical one is enough to neglect the interaction.

Consequently, the condition of weak interparticle interaction for system in Figure 2 is as follows:

$$l_{\text{pcrit}} \approx 6 \cdot l \frac{r_{p1}}{r_V} \left(\frac{\chi_{p1}}{\chi_V} \right)^{1/3}. \tag{17}$$

A similar condition can also be obtained for the case of a system of two nanoparticles in Figure 1. Simplification will occur when the value of the particle integral is much smaller than the other term. Then, expression (17) for this case is as follows:

$$l_{\text{crit}} \approx 6 \cdot r_{p1} \left(\frac{\chi_{p1}}{3 + \chi_{p2}} \right)^{1/3}. \tag{18}$$

It should be noted that the distance l_{crit} is the distance between the centers of nanoparticles, so their size must be taken into account for the real distance between their surfaces.

Let us apply the conditions, which were proposed earlier, to compare them with the experimentally detected phenomena, as shown in the papers [16–29]. We made calculations only for works where the distance was estimated

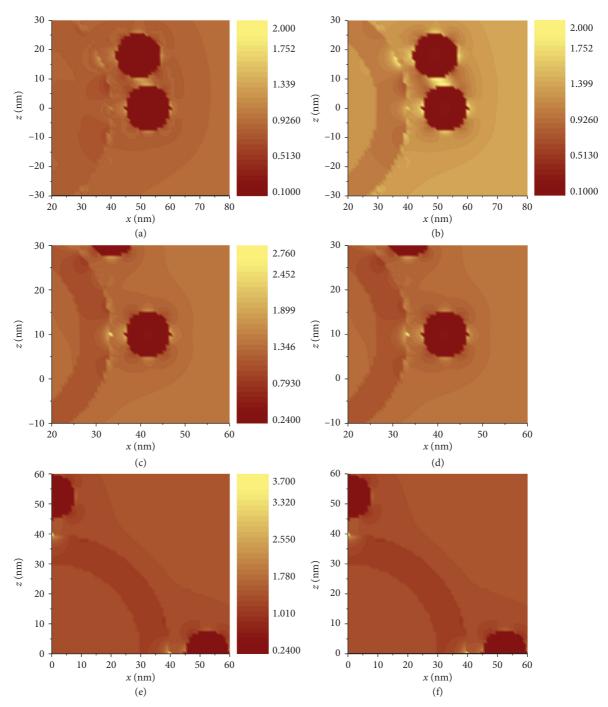


FIGURE 3: Local field distribution in the system of three nanoparticles in Figure 2. The big nanoparticle is of 30 nm inner radius and has the shell of 10 nm thickness with an inner dielectric constant value $\varepsilon_1 = 1.2$ and the outer one $\varepsilon_1 = 1.8$, and the small Au nanoparticles are of 5 nm inner radius with 2.5 nm SiO₂ shell for $\alpha = 20^{\circ}$ (a, b), $\alpha = 40^{\circ}$ (c, d), and $\alpha = 90^{\circ}$ (e, f). (a, c, e) Taking into account the nanoparticles interaction; (b, d, f) without taking into account the nanoparticles interaction.

experimentally in order to compare results. The calculation results and their comparison with the experimental results are given in Table 1. We should pay attention that the proposed condition can be used for approximate estimates. The difference from real results for the big nanoparticles is due to the fact that we used approximation of quasi-point particles; namely, we did not take into account the heterogeneity of the local field inside the nanoparticle.

Differences for small nanoparticles may be caused by the existence of significant nonlinear effects.

4. Conclusions

Hence, in this work, we have proposed the theoretical model explaining the influence of interparticle interaction on the materials properties. Experimental results and calculations

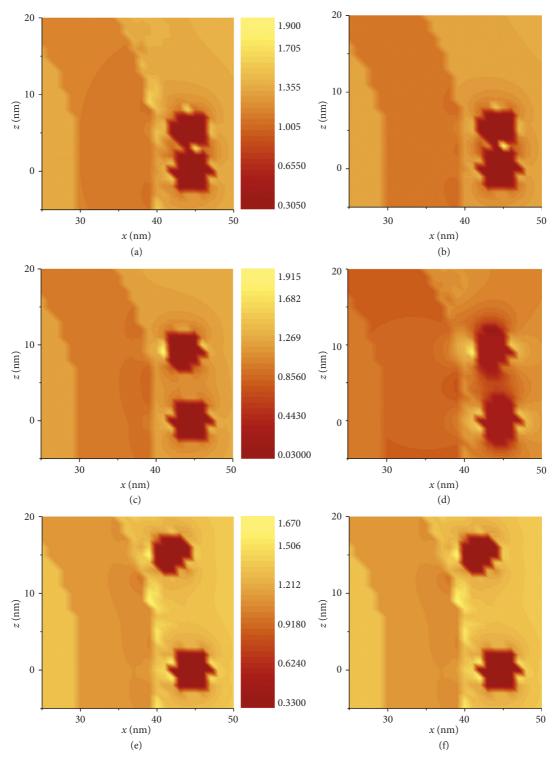


FIGURE 4: Local field distribution in the system of three nanoparticles in Figure 2. The big nanoparticle is of 30 nm inner radius and has the shell of 10 nm thickness with an inner dielectric constant value $\varepsilon_1 = 1.2$ and the outer one $\varepsilon_1 = 1.8$, and the small Au nanoparticles are of 5 nm radius without the shell for $\alpha = 7^{\circ}$ (a, b), $\alpha = 12^{\circ}$ (c, d), and $\alpha = 20^{\circ}$ (e, f). (a, c, e) Taking into account the nanoparticles interaction; (b, d, f) without taking into account the nanoparticles interaction.

using our model demonstrate that interaction between nanoparticles is negligible for the distance which can be estimated with proposed expressions (17) and (18). Consequently, theoretical consideration of the system of many nanoparticles can be simplified by neglecting the particleparticle interactions using the proposed conditions. The results of work may also be useful for engineers of nanoparticles and nanostructured surfaces. They may be

| Nanoparticles material and diameter (nm) | l _{cr} calculated (nm) | l _{cr} experimental (nm) | Reference |
|--|---------------------------------|-----------------------------------|-----------|
| Au, 72 | 250 | 258 | [16] |
| Au, 84 | 290 | 301 | [16] |
| Ag, 80 | 234 | 220 | [17] |
| Ag, 95 | 278 | 290 | [17] |
| Al, 6.8 | 15 | 13.5 | [21] |
| Ag, 10 | 29 | 30 | [26] |

TABLE 1: Critical distance between two nanoparticles.

applicable for the design of nanostructures for various applications, e.g., plasmonic surfaces, superconductors, biosensors, and antivirals. The nature of this phenomenon is the task for the further studies.

Data Availability

Previously reported experimental results of critical distance between the nanoparticles were used to support this study. These prior studies are cited at relevant places within the text as references [16–29].

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Supplementary Materials

Supplementary Materials contain the detailed description of the minimization procedure in formula (2) and formulae for calculation of the coefficients in expression for the adsorption potential (5). (Supplementary Materials)

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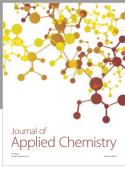
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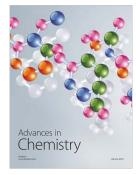


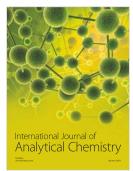














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