The Effective Susceptibility Concept in the Electrodynamics of Nano-Systems

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The effective susceptibility concept is developed for solution of electrodynamical problems of nano-systems. General principles of introducing effective susceptibility are discussed. The effective susceptibility for different nano-systems, namely, thin films, mesoparticles and quantum dots were calculated. Some examples of effective susceptibility uses were demonstrated. In part, the absorption profiles when inhomogeneous along the thickness thin film absorbs the light, was calculated. Discussed concept was used to show the influence of the shape of nano-particles covering the surface on dispersion of surface plasmon polaritons. It was shown that proposed approach can be useful for analysis of surface plasmon polariton resonance experiments. The effective susceptibility concept is useful for modeling of near-field images of linear and nonlinear scanning near-field optical microscopy. The presented method can be applied for modeling of near-field images of different objects. For example, there are pyramid-like quantum dots and semiconductor surface excited by strong focused Gauss-like beam of the light.

Keywords: Low-Dimensional System, Effective Susceptibility, Electrodynamics, Nano-Particles, Thin Film, Scanning Near-Field Optical Microscopy.

1. INTRODUCTION

The nano-technologies which nowadays demonstrate impetuous development has led to the necessity of consideration of electrodynamics of the systems, characterized by small linear dimension.1–3 In particular this problem became actual for consideration of electrodynamical properties of quantum dots4–5 and quantum dot arrays,6–7 development of scanning near-field optical microscopy,8–10 electrodynamics of nano-composite systems,11–13 etc. In any aspect of these one can point to some achievements in solving these problems. Namely, some ideas of the local field were used as base for numerous works in which the nonlocal electrodynamics of quantum well14–16 and quantum dot structures17–19 were studied. The main idea of the most of these works is finding the local field at arbitrary point of the system under consideration. The distillation of local-field method was introduction of local-field factor which connects the short-range local field with long-range external field (see, for example, the reviews20–23). The method is widely used and many results were obtained in the frame of the local-field method. The next step of nano-particle electrodynamics is reformulation of constitutive equations from connection between the local current and local field to connection between the local current and external (long-range) field. This connection lies in the base of effective susceptibility concept discussed in the present work. This approach has a clear physical argument—only external long-range field is controlled in experiment. A local-field having the short-range nature can not be measured directly in experiment. One should note that term “effective” here has any different mean from term “effective” in the effective medium theory.24–27 The effective medium theory is based on the idea of replacing the inhomogeneous (composite) medium by an equivalent homogeneous medium such that the fluctuations induced by restoring the heterogeneity average to zero. Then, roughly speaking, the idea of effective medium theory is replacing one (having complicated structure) medium by another one—homogeneous simple medium characterized by some effective permeability.25 Contrary to this, the idea of effective susceptibility concept consists in consideration an object (the nano-particle, thin film, etc.) which responds to external (probing) signal as a single whole. Then the linear response to the external field (effective susceptibility) is obviously the characteristic of the system. This characteristic is not trivial. Indeed, on the one hand the idea is to consider the system as a single whole but on the other hand the effective susceptibility connects the local characteristic of the system—the local current, which can strongly change from point to point inside the object—with the external long-range field acting on the system as a single whole. The
advantage of effective susceptibility introduction is obviously the possibility to calculate the most of electrodynamical characteristics of the system because the local current distribution which is the source of the field, can be calculated in the frame of discussed approach.

2. LINEAR RESPONSES TO THE LOCAL AND AN EXTERNAL FIELD

Contemporary electrodynamics dealt with small (nano) objects needs to distinguish local and external fields. Let the external long-range field acts on the bulk sample made from homogeneous material which electrodynamical properties are described by dielectric constant \( \varepsilon(\omega) \). It is well known, in any bulk homogeneous material the local field \( E_i(\mathbf{R}, \omega) \) differs from the external ones \( E_i^{(0)}(\mathbf{R}, \omega) \) by dielectric constant \( \varepsilon(\omega) \). Then, when the medium does not characterised by spatial dispersion, the local field can be defined via external field as

\[
E_i(\mathbf{R}, \omega) = \varepsilon(\omega) E_i^{(0)}(\mathbf{R}, \omega)
\]

In this case, when the external field, measured somewhere at infinity, depended on the coordinate as \( E_i^{(0)}(\mathbf{R}, \omega) = E_i^{(0)}(\omega) e^{i \mathbf{k} \cdot \mathbf{R}} \), the local field inside the bulk sample depends on coordinate of observation point only via phase factor

\[
E_i(\mathbf{R}, \omega) = E_i(\omega) e^{i \mathbf{k} \cdot \mathbf{R}}
\]

with \( \mathbf{k} = \sqrt{\varepsilon(\omega)} \mathbf{k} \) wave vector of the electromagnetic wave propagating inside a bulk. This local field is usually named the electric induction. The dielectric constant shows the field enhancement (weakening) by the local currents inside the bulk induced by external field. Differently other situation arises when one considers the acting long-range external field on the small particle. In this case, because near-surface transitional layer fills rather essential part of the volume of the particle, the local field will be strongly inhomogeneous. Then, the connection between the local and external fields can not be presented in a simple form of Eq. (1). To find this connection one should remember, that induced current and local field are connected by linear response to the local filed (or electrical susceptibility)

\[
j_i(\mathbf{R}, \omega) = -i \omega \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}, \omega)
\]

This relation is usually named as constitutive equation. The constitutive equation is not convenient for study of electrodynamics of small systems because the local field inside the particle can not be measured and controlled experimentally. Only external field can be controlled in experiment. Then, analogously to constitutive equation one can introduce the relation connecting the local current with external field.

\[
j_i(\mathbf{R}, \omega) = -i \omega \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}, \omega)
\]

One should note that the connection between the local current and external field [Eq. (4)] has an universal form. Indeed, in general case, because of nonlocal nature of the electrodynamic interactions this connection should be written in the form

\[
j_i(\mathbf{R}, \omega) = -i \omega \int_V d\mathbf{R'} \chi_{ij}(\mathbf{R} - \mathbf{R'}, \omega) E_j^{(0)}(\mathbf{R'}, \omega)
\]

with \( \chi_{ij}(\mathbf{R} - \mathbf{R'}, \omega) \) nonlocal tensor of linear response to external field. Taking into account that integration in Eq. (5) is over the particle volume and the external field in the most cases is the long-range and very weakly changes in the distance about linear dimension of the particle, one can remove the external field from integrand and obtain Eq. (5), where the effective susceptibility has a form

\[
\chi_{ij}(\omega) = \int_V d\mathbf{R'} \chi_{ij}(\mathbf{R} - \mathbf{R'}, \omega)
\]

Then, the linear response to a local field is the characteristic of the material and linear response to an external field is the characteristic of the object made of this material. As a result, the knowledge of induced currents inside the system under consideration allows us to find the local field at any point in the system, which means the solution of the electrodynamic problem. Then one can to insist that effective susceptibility characterizes the most electrodynamical properties of the nano-system.

3. LOW-DIMENSION SYSTEMS. ELECTRODYNAMICAL POINT OF VIEW

What is the reason of the difference in local field inside the bulk and inside the small particle? Obviously the differences are connected with contribution of interfaces. In the bulk materials the interfaces and transitional layers at interfaces fill only very small part of the volume of the object. On the contrary, the transitional layer fills the significant part of the volume of small particle (Fig. 1). Then, one can think that when the object is acted by long-range external field, the local field inside a small particle strongly changes from point to point, but the local field inside a bulk strongly changes only in small region near the interface. Then, the thickness of transition layer is the parameter of linear dimension with the help of which one should compare the linear dimension of the system under consideration. Then, when the characteristic linear dimension of the object at any direction is about of length of transition layer, the object can be considered as low-dimensional. From this point of view the thin film with thickness \( h \) about of transition layer length \( l \) should be supposed as
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4. SELF-CONSISTENT EQUATION AND EFFECTIVE SUSCEPTIBILITY

Let the system consisting of the object embedded in any medium is acted by external field \( E_i^{(0)}(\mathbf{R}, \omega) \). It is well known, the local field at any point in the system under consideration can be found with equation

\[
E_i(\mathbf{R}, \omega) = E_i^{(0)}(\mathbf{R}, \omega) - i\omega \mu_0 \int d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}', \omega) J_j(\mathbf{R}', \omega)
\]

(7)

where \( G_{ij}(\mathbf{R}, \mathbf{R}', \omega) \) is a photon propagator (Green function of Maxwell equations) describing the light propagation in the medium in which the object of volume \( V \) is situated. Because the local current and local field are connected by constitutive equation [Eq. (3)], one can rewrite Eq. (7) in the form

\[
E_i(\mathbf{R}, \omega) = E_i^{(0)}(\mathbf{R}, \omega) - (\omega/c)^2 \int d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}', \omega) \chi_{ij}(\omega) E_j(\mathbf{R}', \omega)
\]

(8)

with dimensionless electrical susceptibility (linear response to local field) \( \chi_{ij}(\omega) = (1/\varepsilon_0)\chi_{ij}'(\omega) \). The equation for self-consistent field is usually named as Lippmann-Schwinger equation. One should note, that the electrodynamical properties of the material of which the object is made, are described by linear response to local field \( \chi_{ij}'(\omega) \) and should be found either from microscopic calculations in the frame of quantum theory (see, for example, the problem N159 in textbook, and works) or from experimental measurements. Introducing the dimensionless effective susceptibility \( X_{ij}(\mathbf{R}, \omega) = (1/\varepsilon_0)X_{ij}'(\mathbf{R}, \omega) \), which connects the current inside the object and external long-range field (see, Eq. (5)), one can write the solution of Eq. (8) in the form

\[
E_i(\mathbf{R}, \omega) = E_i^{(0)}(\mathbf{R}, \omega) - (\omega/c)^2 \int d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}', \omega) \chi_{ij}(\omega) E_j(\mathbf{R}', \omega)
\]

(9)

From this point of view the effective susceptibility describes the electrodynamical properties of the system. Then, knowledge of effective susceptibility of the system allows us to calculate the local self-consistent field at any point of the system under consideration. It means that the problem of calculation of self-consistent field transforms to problem of effective susceptibility calculation. In the next sections we shall demonstrate some approaches for calculation of effective susceptibility.

5. EFFECTIVE SUSCEPTIBILITY OF SMALL PARTICLE

Let us consider the interaction of small particle with external electromagnetic field radiating the particle. The self-consistent electromagnetic field at any point inside the system “medium-particle” obeys to Eq. (8). To find the effective susceptibility one should to use Eqs. (3) and (5) and write connection between local and external fields, supposing that reciprocal matrix (\( X_{ij}(\mathbf{R}, \omega) \)) exists.

\[
E_j^{(0)}(\mathbf{R}, \omega) = (X_{ij}(\mathbf{R}, \omega))^{-1} \chi_{ik}(\omega) E_k(\mathbf{R}, \omega)
\]

(10)

Then, one can substitute this connection in Eq. (8) and obtain

\[
E_j(\mathbf{R}, \omega) = (X_{ij}(\mathbf{R}, \omega))^{-1} \chi_{ik}(\omega) E_k(\mathbf{R}, \omega) - k_j^2 \int d\mathbf{R}' G_{ij}(\mathbf{R}, \mathbf{R}', \omega) \chi_{ik}(\omega) E_k(\mathbf{R}', \omega)
\]

(11)
This equation is true for all points in the system, including all points inside the particle. Then, one can integrate the both parts of equation over the particle volume
\[
\int_V dR E_j(R, \omega) = \int_V dR (X_{ij}(R, \omega))^{-1} \chi_{ij}(\omega) E_i(R, \omega)
\]
\[
-k_0^2 \int_v dR' \int_V dR G_{ij}(R', R, \omega) \times \chi_{ji}(\omega) E_j(R, \omega)
\]
where replacing the integration variables in last term of right part was performed. Supposing that local field can be represented in the form
\[
E_j(R, \omega) = \sum_k E_j(k, \omega) e^{ikR}
\]
one obtains from Eq. (12)
\[
\sum_k \int_V dR \left[-(X_{ij}(R, \omega))^{-1} \chi_{ij}(\omega)ight.
\]
\[
+ \omega^2 \int_V dR' G_{ik}(R', R, \omega) \chi_{ki}(\omega) + \delta_{ij}
\]
\[
\left. \right] \times E_i(k, \omega) e^{ikR} = 0
\]
Because exponents are the complete set of orthonormal functions, to satisfy this equation one needs to put
\[
-(X_{ij}(R, \omega))^{-1} \chi_{ij}(\omega)
\]
\[
+ k_0^2 \int_V dR' G_{ik}(R', R, \omega) \chi_{ki}(\omega) + \delta_{ij} = 0
\]
Solution of this equation is
\[
X_{ij}(R, \omega)
\]
\[
= \chi_{ij}(\omega) \left[ \delta_{ij} + k_0^2 \int_V dR' G_{ik}(R', R, \omega) \chi_{ki}(\omega) \right]^{-1}
\]
One should to note that obtained expression for effective susceptibility [Eq. (16)] is true under condition \(\text{Im det}[\delta_{ij} + k_0^2 \int_v dR' G_{ik}(R', R, \omega) \chi_{ki}(\omega)] \neq 0\) which supplies with condition of nonzero determinant of matrix \([\delta_{ij} + k_0^2 \int_v dR' G_{ik}(R', R, \omega) \chi_{ki}(\omega)]\). Obviously that real nano-system has its linear response to the local field imaginary part and above mentioned condition is realized automatically.

6. EFFECTIVE SUSCEPTIBILITY OF QUANTUM DOT

Let us suppose that a quantum dot with characteristic linear dimension \(L\) (its volume \(V \sim L^3\)) has a set of electron quantum states \(|0\>, |1\>, \ldots |n\>\) with energies \(E_0, E_1, \ldots E_n\). To calculate the effective susceptibility of quantum dot one needs define the linear response to the local field which can be provided according to\(20, 38, 39\)
\[
\chi_{ij}(R, R', \omega) = \frac{1}{\mu_0 \omega^2} \sum_\alpha a_\alpha(\omega) j_\alpha^jo(R) j_\alpha^i(R')
\]
where \(j_\alpha^jo(R), j_\alpha^i(R')\) are the \(k\) and \(l\)-th components of the transition current densities from the \(\alpha\) level to the ground state \(|0\>\) and vice versa. Neglecting the small diamagnetic part, one has
\[
j_\alpha^ao = -\frac{e \hbar}{2 \imath m} (\psi_\alpha^o)^* \nabla \psi_\alpha^o - \psi_\alpha^o \nabla (\psi_\alpha^o)^*
\]
where \(\psi_\alpha^o\) and \(\psi_\alpha^o\) are the wave-functions of the ground and \(\alpha\) states, respectively. In Eq. (17) the abbreviation
\[
a_\alpha(\omega) = 2 \mu_0 \frac{E_\alpha - E_0}{\hbar^2 (\omega + i \nu)^2 - (E_\alpha - E_0)^2}
\]
is used, where \(E_\alpha\) and \(E_0\) are the energies of the \(\alpha\) and ground states, and \(\nu\) is a phenomenological relaxation frequency. One should note that in the case under consideration the nonlocal relation between the local current and the local field
\[
j_j(R, \omega) = -i \omega \int_V dR' \chi_{ij}(R, R', \omega) E_j(R', \omega)
\]
is used. Hereinafter we shall make after Ref. [20]. Substituting Eq. (17) into Eq. (20) and using Lippmann-Schwinger equation [Eq. (7)] one obtains the following equation for the \(i\)-th component of the self-consistent (local) field:
\[
E_i(R, \omega) = E_i^{(0)}(R, \omega) - \sum_\alpha a_{\alpha}(\omega) F_{ij}^{\alpha\alpha}(R, \omega) \times \int_V dR' j_\alpha^{i\alpha}(R') E_j(R', \omega)
\]
where
\[
F_{ij}^{\alpha\alpha}(R, \omega) = \frac{1}{\mu_0} \int_V dR' G_{ij}(R, R', \omega) j_\alpha^{i\alpha}(R')
\]
is proportional to the \(i\)-th component of the electric field which is induced by the electron transition \((\alpha \rightarrow 0)\) current density distribution at point \(R\). If one now acts with the integral operator \(\int_V dR' j_\alpha^{i\alpha}(R') \ldots\) on Eq. (21) and thereafter makes a summation over the Cartesian coordinates one obtains a system of the linear algebraic equations
\[
\sum_\alpha (\delta_{ij} + a_{\alpha}(\omega) N^{\alpha\alpha}(\omega)) \cdot \psi_\alpha^{i\alpha}(\omega) = \gamma_0^{i\alpha}(\omega)
\]
among the unknown quantities
\[ \gamma^{ab}(\omega) = \int_V dR \gamma^{ab,0}(R) E_i(R, \omega) \]  
(24)

In Eq. (23) the abbreviations
\[ N^{ab}(\omega) = \int_V dR \gamma^{ab,0}(R) E_i(0, \omega) \]

\[ = \frac{1}{\mu_0} \int_V dR dR' \gamma^{ab,0}(R) G_{ij}(R, R', \omega) j^{ai}(R') \]

and, taking into account that the incident field is slowly varying across the quantum dot,
\[ \gamma^{ab,0}(\omega) = \int_V dR \gamma^{ab,0}(R) E_i(0, \omega) = \gamma_i^{ab,0} E_i(0, \omega) \]

(26)

with
\[ \gamma_i^{ab} = \int_V dR i^{ab}(R) \]

(27)

and \( E_i(0, \omega) \) external field at quantum dot location were used. Formally, the solution of Eq. (23) can be written in the form
\[ \gamma^{ab}(\omega) = \frac{B^{ab}}{\Delta} \gamma_i^{ab,0} E_i(0, \omega) \]

(28)

where \( B^{ab} \) is the relevant algebraic complement and \( \Delta \) is the determinant of the matrix \( \{\delta_{ij} + a_{ij} N^{ab}\} \). Once the set of algebraic equations has been solved the relation between the local and external fields, i.e.,
\[ E_i(R) = \Lambda_{ij}(R, \omega) E_i(0, \omega) \]

with a local field factor
\[ \Lambda_{ij}(R, \omega) = \left\{ \delta_{ij} - \sum_{\alpha, \beta} a_{\alpha}(\omega) F_{ij}(R, \omega) \frac{B^{\alpha\beta}}{\Delta} \gamma^{\alpha\beta} \right\} \]

(30)

can be established. Then, one can obtain the “scattered” part of the field as
\[ E_i^{(\text{sc})}(R, \omega) = -\sum_{\alpha, \beta} a_{\alpha}(\omega) F_{ij}(R, \omega) \frac{B^{\alpha\beta}}{\Delta} \gamma^{\alpha\beta} E_i(0, \omega) \]

(31)

Let us suppose that the origin of the scattered field is the self-consistent current \( J(R) \) induced inside the quantum dot by an external field (see, Fig. 3), then the scattered field one can write in the form
\[ E_i^{(\text{sc})}(R, \omega) = -i \mu_0 \int G_{ij}(R, R', R) j_i(R') dR' \]

(32)

Compare Eqs. (31) and (32) with taking into account Eq. (22), one can write
\[ j_i(R) = \frac{1}{\mu_0} \sum_{\alpha, \beta} a_{\alpha}(\omega) \gamma^{\alpha\beta} E_i(0, \omega) \]

(33)

Then, the effective susceptibility of quantum dot connecting the local current and external field can be written in the form
\[ X_{ij}(R) = \frac{i}{\omega \mu_0} \sum_{\alpha, \beta} a_{\alpha}(\omega) \gamma^{\alpha\beta} \frac{B^{\alpha\beta}}{\Delta} \gamma_i^{\alpha\beta} \]

(34)

7. EFFECTIVE SUSCEPTIBILITY OF A SYSTEM OF QUANTUM DOTS

To calculate the electrodynamic properties of system of quantum dots one should to take into account the interactions between the particles inside the system under consideration. To clarify the problem of calculation of effective susceptibility of the system one should to start from consideration of system, consisting of two quantum dots. To calculate the effective susceptibilities of the particles in the system under consideration, one should write the equations for self-consisten fields inside of each of the particles. Introducing the vectors \( R_1 \) and \( R_2 \) of a point inside particle 1 and 2, respectively, one can write two equations relative the self-consistent fields inside the particles
\[ E_i(R_1, \omega) = E_i^{(0)}(R_1, \omega) - k_0^2 \int dR_i G_{ij}(R_1, R_2, \omega) \]

\[ \times \int dR' j_i(R') \frac{1}{\mu_0 \omega^2} \sum_{\alpha} a_{\alpha}(\omega) j^{\alpha}(R_1) \gamma_i^{\alpha\beta} \]

\[ \times (R') E_i(R', \omega) \]

\[ - k_0^2 \int dR_i G_{ij}(R_1, R_2, \omega) \]

\[ \times \int dR' j_i(R') \frac{1}{\mu_0 \omega^2} \sum_{\alpha} a_{\alpha}(\omega) j^{\alpha}(R_2) \gamma_i^{\alpha\beta} \]

\[ \times (R') E_i(R', \omega) \]

(35)

\[ E_i(R_2, \omega) = E_i^{(0)}(R_2, \omega) - k_0^2 \int dR_i G_{ij}(R_2, R_1, \omega) \]

\[ \times \int dR' j_i(R') \frac{1}{\mu_0 \omega^2} \sum_{\alpha} a_{\alpha}(\omega) j^{\alpha}(R_1) \gamma_i^{\alpha\beta} \]

\[ \times (R') E_i(R', \omega) \]

\[ - k_0^2 \int dR_i G_{ij}(R_2, R_1, \omega) \]

\[ \times \int dR' j_i(R') \frac{1}{\mu_0 \omega^2} \sum_{\alpha} a_{\alpha}(\omega) j^{\alpha}(R_2) \gamma_i^{\alpha\beta} \]

\[ \times (R') E_i(R', \omega) \]

\[ - k_0^2 \int dR_i G_{ij}(R_2, R_1, \omega) \]
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\[ \times \int_{V_1} dR_1 \frac{1}{\mu_0 c^2} \sum_a \hat{a}_a(\omega) f^{(0)}(R_1) f^{(0)}_\alpha \]
\[ \times (R_1^0) E(R_1^0, \omega) \]  

(36)

To solve these equations one can act on the both left and right parts of the Eqs. (35) and (36) with integral operators \[ \int_{V_1} dR_1^0 f^{(0)}(R_1^0) \ldots \int_{V_n} dR_n^0 f^{(0)}(R_n^0) \ldots \]  
respectively. As a result one obtains the next system of algebraic equations

\[ \left\{ \begin{array}{l}
y_1^\beta + \sum_a \tilde{a}_a(\omega) N_{11}^{ba} y_1^\alpha = \gamma_1^0 E_i(1, \omega) \\
y_2^\beta + \sum_a \tilde{a}_a(\omega) N_{21}^{ba} y_1^\alpha + \sum_a \tilde{a}_a(\omega) N_{22}^{ba} y_2^\alpha = \gamma_2^0 E_i(2, \omega)
\end{array} \right. \]

(37)

where the designations

\[ \gamma_\alpha^\beta = \int_{V_n} dR_n^0 f^{(0)}(R_n^0) E_i(R_n^0) \]  

(38)

\[ \gamma_\alpha^\beta = \int_{V_n} dR_n^0 f^{(0)}(R_n^0) E_i(1, \omega) \simeq \gamma_1^0 E_i(1, \omega) \]  

(39)

and \[ \tilde{a}_a(\omega) = a_a(\omega)/\mu_0 c^2 \]  
were used. One has taken into account that due to small dimension of the particles, the variations of the external field at the particles can be neglected. Solution of the system [Eq. (37)] can be written in the form

\[ \gamma_\alpha^\beta(\omega) = \frac{B^{ba}_1}{\Delta} \gamma_1^0 E_i(1, \omega) + \frac{B^{ba}_2}{\Delta} \gamma_2^0 E_i(2, \omega), \quad n = 1, 2 \]

(40)

The next designations

\[ N_{am}^{ba} = \int_{V_a} dR_a \int_{V_m} dR_m^0 f^{(0)}(R_a) G_{am}(R_a, R_m^0, \omega) f^{(0)}(R_m^0) \]  

(41)

were used above. Besides, \[ B^{ba}_m \]  
is the relevant algebraic complement and \[ \Delta \]  
is the determinant of the matrix

\[ \left[ \begin{array}{cc}
\delta^{ba} + \sum_a \tilde{a}_a(\omega) N_{11}^{ba} & \sum_a \tilde{a}_a(\omega) N_{12}^{ba} \\
\sum_a \tilde{a}_a(\omega) N_{21}^{ba} & \delta^{ba} + \sum_a \tilde{a}_a(\omega) N_{22}^{ba}
\end{array} \right] \]

(42)

In the case when the distance between quantum dots are much less than wavelength of an external field, the effective susceptibilities of the quantum dots are

\[ X_{ij}^{(0)}(R, \omega) = \frac{1}{\delta^{00}} \sum_{m, n} \tilde{a}_a(\omega) f^{(0)}(R) \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_i(1, \omega) \]

(43)

were \[ n \]  
and \[ m \]  
is a number of the particle. In the case when the distance between the particles is about wavelength and the external field has a form \[ E_i(1, \omega) = E_i^{(0)}(\omega) e^{ikR} \], one should write

\[ X_{ij}^{(1)}(R_1, \omega) = \frac{1}{k^2} \sum_a \tilde{a}_a f^{(0)}(R_1) \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_1(1, \omega) \]
\[ \times \left( \frac{B^{ba}_m}{\Delta} \gamma_1^0 + \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_i(R_1, \omega) \right) \]  

(44)

\[ X_{ij}^{(2)}(R_2, \omega) = \frac{1}{k^2} \sum_a \tilde{a}_a f^{(0)}(R_2) \]
\[ \times \left( \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_1(R_2, \omega) + \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_2(R_2, \omega) \right) \]

(45)

Similar expressions for effective susceptibilities of the particles one can write for the case of three quantum dots. Indeed, let one consider the system, consisting of three identical quantum dots, where any point inside of each of the particles is characterized by the vectors \[ R_1, R_2, \]  
and \[ R_3 \]  
respectively. To calculate the effective susceptibilities of the quantum dots one should to establish the self-consistent local fields inside of each of the particles. Then, instead of Eq. (37) one should to write

\[ \left( \delta^{ba} + \sum_a \tilde{a}_a(\omega) N_{11}^{ba} \right) \gamma^\alpha + \sum_a \tilde{a}_a(\omega) N_{12}^{ba} \gamma^\alpha + \sum_a \tilde{a}_a(\omega) N_{21}^{ba} \gamma^\alpha = \gamma_1^0 E_i(1, \omega) \]
\[ + \sum_a \tilde{a}_a(\omega) N_{22}^{ba} \gamma^\alpha + \left( \delta^{ba} + \sum_a \tilde{a}_a(\omega) N_{11}^{ba} \right) \gamma^\alpha = \gamma_1^0 E_i(2, \omega) \]
\[ + \sum_a \tilde{a}_a(\omega) N_{12}^{ba} \gamma^\alpha + \sum_a \tilde{a}_a(\omega) N_{22}^{ba} \gamma^\alpha + \left( \delta^{ba} + \sum_a \tilde{a}_a(\omega) N_{11}^{ba} \right) \gamma^\alpha = \gamma_1^0 E_i(3, \omega) \]

(46)

Solution of this system of algebraic equations one can write analogously to Eq. (35)

\[ \gamma_\alpha^\beta(\omega) = \frac{1}{\Delta} \sum_{i=1}^{3} \left( B^{ba}_1 \gamma_1^0 E_i(1, \omega) + B^{ba}_2 \gamma_1^0 E_i(2, \omega) + B^{ba}_3 \gamma_1^0 E_i(3, \omega) \right) \]

(47)

with \[ \Delta \]  
is determinant of the main matrix and \[ B^{ba}_m \]  
is relevant algebraic complements. Then, one can write for effective susceptibilities of the particles of the system

\[ X_{ij}^{(1)}(R_1, \omega) = \frac{1}{k^2} \sum_a \tilde{a}_a f^{(0)}(R_1) \frac{B^{ba}_m}{\Delta} \gamma_1^0 E_i(R_1, \omega) \]
\[ \times \left( B^{ba}_m + B^{ba}_m \gamma_1^0 E_1(R_2, \omega) + B^{ba}_m \gamma_1^0 E_2(R_2, \omega) \right) \]

(48)

It is clear that similar result one can obtain for the systems of quantum dots consisting of more than three particles. No doubt that the whole number of the particles in the system can be rather large. But for the systems, consisting of very large number of quantum dots one needs to use some different approaches. One of these approaches will be considered in this work below in Section 8. Because of equation \( \text{Re}\Delta = 0 \) determines the resonances inside the system, one can easily observe configuration resonances in the system. Indeed, due to \( N_{\text{num}} \) depends on the distance between particles \( n \) and \( m \), the resonance properties will be depended on relative position of the particles inside the system.

8. EFFECTIVE SUSCEPTIBILITY OF A FILM

Analogously to the case of nano-particle, one can calculate the effective susceptibility of a film. Discussed conception allows us to take into account the inhomogeneous along the thickness film, namely when dielectric constant is depended on \( z \)-coordinate. Then, one can write that linear response to a local field is depended on \( z \)

\[
\chi_{ij}(z, \omega) = e_{ij}(z, \omega) - \delta_{ij}
\]

(51)

Because the film is homogeneous in its plane (XOY plane in Cartesian coordinate system), one can perform the partial Fourier transformation in the film plane, then one can transform to the \( k-z \) representation. In this representation Eq. (8) has a form

\[
E_i(k, z, \omega) = E_i^{(0)}(k, z, \omega) - k_0^2 \int \frac{dz'}{\Delta} G_{ij}(k, z', \omega) \chi_{ji}(z', \omega) E_j(k, z', \omega)
\]

(52)

To calculate the effective susceptibility of the thin film one needs to differ at least two different cases. First, the film can be very thin—the case of ultra-thin film. It means that one can not say that light propagates across the film thickness. From this point of view, the film and light acting the film should be considered as a single whole. Namely one should consider the single excited system consisting of the film and electromagnetic field localized at the film. Then, when one would like to calculate the effective susceptibility of the ultra-thin film, one should consider the film as secondary source of the “scattered” field, and make the calculation similar to demonstrated in Section 5 of this work. As a result, one can obtain for effective susceptibility of the ultra-thin film

\[
X_{ij}^{(u)}(k, z, \omega) = \chi_{ij}(z, \omega) \left[ \delta_{ij} + k_0^2 \int_0^h dz' G_{ij}(k, z', \omega) \chi_{ji}(z, \omega) \right]^{-1}
\]

(53)

where the integration is over the film thickness.

Definitely other situation arises when the thickness of the film is rather large, when the phase shift can plays the role. In this case, one should take into account a possibility of light propagation across film thickness. Other words, one should to solve Eq. (52) explicitly with taken into account processes of light propagation along the thickness of the film. The correct procedure of solution this problem was proposed in Ref. [42] when homogeneous isotropic material of film (\( e_{ij}(\omega) = e_2(\omega) \delta_{ij} \)). Unfortunately we cannot point out to the method of finding effective susceptibility of the inhomogeneous film where the phase shifts can play the role. From other hand it is clear that only when thickness of the film is more (about) than \( \lambda/2 \) (\( \lambda \) is the wavelength of probing light) the phase shifts can contribute to optical properties of the film. Then, one has dealings with the thick film when its thickness \( h \geq \lambda/2 \).

It means that one can use standard approach to calculate the field inside the film, taking into account the border conditions and reflection and transmission coefficients. To obtain the effective susceptibility of the homogeneous film with taken into account the wave propagation along the thickness of the film, one should use two equations of self-consistent fields

\[
E_i(k, z, \omega) = E_i^{(0)}(k, z, \omega) - (e_2(\omega) - e_1) k_0^2
\]

(54)

\[
\times \int \frac{d \omega}{\Delta} G_{ij}^{(1)}(k, z, \omega) E_j(k, z', \omega)
\]

\[
E_i(k, z, \omega) = (e_2(\omega) - e_1) k_0^2 \int \frac{dz'}{\Delta} G_{ij}^{(2)}(k, z', \omega) E_j(k, z', \omega)
\]

(55)

where \( k_0 = \omega/c \) and \( \int d \omega' = \int_0^\omega dz' + \int_0^\omega dz' \ldots \) because the integral is over all space outside the film. Designations \( G_{ij}^{(1,2)}(k, z, \omega) \) mean the photon propagators of medium (1) where the thin film made from material of medium 2 is situated. Performing solution of Eqs. (54) and (55) according to Ref. [42], one obtains the connection between the current and external field. In the case
of s-polarized incident field \( E_i^{(0)}(k, z, \omega) = E_i^{(0)}(k, \omega) e^{i\eta_1 z} \)

this connection has a form

\[
J_i(k, z, \omega) = (e_2(\omega) - e_1) [A^+ e^{i(\eta_1 - \eta_2) z} + A^- e^{-i(\eta_2 + \eta_1) z}] E_i^{(0)}(k, \omega) e^{i\eta_2 z} 
\]  

(56)

where \( \eta_{1,2} = k_0 \sqrt{\varepsilon_{1,2} - \sin^2 \vartheta} \), \( A^\pm \) are functions of incident angle \( \vartheta \) and frequency \( \omega \), depending on film thickness \( h \) and dielectric constants of environment \( \varepsilon_1 \) and film material \( \varepsilon_2 \) (see, Appendix).

Taking into account that

\[
\bar{X}_y(k, z, \omega) = \frac{V_M}{\chi_{ij}}(\omega),
\]

with \( \bar{X}_y(\omega) = V_M \chi_{ij}(\omega) \), and \( \chi_{ij}(\omega) \) the tensor of linear response of the ellipsoidal particle. Averaging over positions of the particles performed according to

\[
\sum_{\alpha=1}^{N} G_{ij}(\mathbf{r} - \mathbf{r}_\alpha, z, z_\alpha, \omega) \bar{X}_{ij}(\omega) E_i(\mathbf{r}_\alpha, z_\alpha, \omega)
\]

\[
= \frac{1}{2^{N-1}} \int d\mathbf{r}_1 d\mathbf{r}_2 \ldots d\mathbf{r}_N
\]

\[
\times \sum_{\alpha=1}^{N} \frac{d\mathbf{k}}{(2\pi)^2} e^{-i \mathbf{k} \cdot (\mathbf{r}_\alpha - \mathbf{r}_\beta)} G_{ij}(\mathbf{k}, z, z_\alpha, \omega) \times \bar{X}_{ij}(\omega)
\]

\[
\times \int \frac{d\mathbf{k}'}{(2\pi)^2} e^{-i \mathbf{k}' \cdot \mathbf{r}} E_i(\mathbf{k}', z, z_\alpha, \omega)
\]

(60)

where \( N \) is concentration of the particles at the surface of a substrate, \( \mathbf{r} \) is the coordinate in the layer plane, \( \mathbf{r}_\alpha \) is the position of \( \alpha \)-th particle in the layer plane. Then, the equation of self-consistent field written in the \( \mathbf{k}, z \) — representation has a form

\[
E_i(\mathbf{k}, z, z_\alpha, \omega) = E_i^{(0)}(\mathbf{k}, z_\alpha, \omega) - n k_0^2 G_{ij}(\mathbf{k}, z, z_\alpha, \omega)
\]

\[
\times \bar{X}_{ij}(\omega) E_j(\mathbf{k}, z_\alpha, \omega)
\]

(62)

Taken into account that \( \bar{X}_{ij}(\omega) \) is a response to a local field, one can write

\[
E_i(\mathbf{k}, z, z_\alpha, \omega) = (\bar{X}_{ij}(\omega))^{-1} P_j(\mathbf{k}, z_\alpha, \omega)
\]

(63)

where \( P_j(\mathbf{k}, z_\alpha, \omega) \) is the polarization of the submonolayer film in \( \mathbf{k} - z \) representation. Substitution of Eq. (63) into Eq. (62) gives us the equation connecting the layer polarization and an external field

\[
(\bar{X}_{ij}(\omega))^{-1} P_j(\mathbf{k}, z_\alpha, \omega)
\]

\[
= E_i^{(0)}(\mathbf{k}, z_\alpha, \omega) - n k_0^2 G_{ij}(\mathbf{k}, z, z_\alpha, \omega) P_j(\mathbf{k}, z_\alpha, \omega)
\]

(64)

It should be noted that one supposes in Eqs. (62)–(64) that all particles are situated at the same distance \( z_\alpha \) from the surface. The solution of Eq. (64) has a form

\[
P_j(\mathbf{k}, z, z_\alpha, \omega) = X_{ij}(\mathbf{k}, z, z_\alpha, \omega) E_i^{(0)}(\mathbf{k}, z_\alpha, \omega)
\]

(65)

where the effective susceptibility

\[
X_{ij}(\mathbf{k}, z, z_\alpha, \omega) = [(\bar{X}_{ij}(\omega))^{-1} + n k_0^2 G_{ij}(\mathbf{k}, z, z_\alpha, \omega)]^{-1}
\]

(66)
One should to note that obtained expression for effective susceptibility of the submonolayer cover is remarkable because of as concentration as shape (and dimension) of the particles define the susceptibility. This, for example allows develop the methods of modeling of submonolayer covers optical properties which can be useful for determination of concentration, shape, and dimensions of the particles by simple optical measurements.44

The optical response of the quasi-pointness dipole two-dimensional lattice was considered in Ref. [48]. The effective susceptibility of the dipole layer was calculated too.

10. CALCULATION OF SELF-ACTION FIELD INSIDE OF NANO-OBJECT

The self-action field (local-field reaction) is the important characteristic of the nano-particle. It defines, in part, the Lamb shifts of the energy levels of quantum dot, and, generally, the linear response to the external field. Moreover, the need of calculation of the local field inside the source of the field arises when the electrodynamics properties of nano-systems is calculated. Let us suppose that one needs to calculate the field, which is caused by the currents induced in the particle, acted at any point inside the particle. Then one needs to calculate the field scattered by the particle inside the particle. Other words, one should to calculate the field which in the frame of effective susceptibility concept has a form

\[ E_i^{(as)}(\mathbf{R}, \omega) = -k_0^2 \int_V d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) \times \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}', \omega), \quad \mathbf{R} \in V \]  

(67)

Because Green function has a singularity at \( \mathbf{R} = \mathbf{R}' \), the problem of calculation of self-action field, when \( \mathbf{R} \in V \) exists. The method of calculation of integrals similar to

\[ \int_V d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}', \omega) \]

when \( \mathbf{R} \in V \) was discussed in Refs. [49, 50]. The method proposed in these references named as inclusion volume method is widely used, especially when numerical calculations in near-field.32,51,52 Calculation of self-action field in the frame of effective susceptibility concept has some specific features, which we demonstrate here. To calculate the self-action field let one choose the small region at point \( \mathbf{R} \) (see, Fig. 3), and rewrite right part of Eq. (67) in the form

\[ E_i^{(as)}(\mathbf{R}, \omega) = -k_0^2 \int_{V - \delta} d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}', \omega) \]

\[ -k_0^2 \int_{\delta} d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) \chi_{ij}(\omega) E_j^{(0)}(\mathbf{R}', \omega) \]  

(69)

In the near-field limit one can suppose that

\[ \lim_{\mathbf{R} \to \mathbf{R}} G_{ij}(\mathbf{R} - \mathbf{R}', \omega) = \frac{1}{k_0^2} \frac{g_{ij}}{(\mathbf{R} - \mathbf{R}')^3} \]  

(70)

Taking into account this circumstance, one can write for effective susceptibility

\[ \begin{align*}
\lim_{\mathbf{R} \to \mathbf{R}} X_{ij}(\mathbf{R}) &= \left( \chi_{ij}(\omega) \right)^{-1} + \int_V d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} \\
&= \left( \chi_{ij}(\omega) \right)^{-1} + \int_V d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} + \int_{\delta} d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} \\
&= \left( \chi_{ij}(\omega) \right)^{-1} + \int_{V - \delta} d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} + \int_{\delta} d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} \\
\end{align*} \]  

(71)

Because

\[ \begin{align*}
\left( \chi_{ij}(\omega) \right)^{-1} + \int_V d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} &\simeq \int_{\delta} d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} \\
\left( \chi_{ij}(\omega) \right)^{-1} + \int_{V - \delta} d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} &\simeq \left[ \int_{\delta} d\mathbf{R}' \frac{1}{(\mathbf{R}' - \mathbf{R})^3} \right]^{-1} g_{ij}^{(3)} \\
\end{align*} \]  

(72)

one can see, that two first terms in the right part of (72) is not singular but the third term is singular. Then, one can evaluate

\[ \left( \chi_{ij}(\omega) \right)^{-1} + \int_V d\mathbf{R}' \frac{g_{ij}}{(\mathbf{R}' - \mathbf{R})^3} \simeq \left[ \int_{\delta} d\mathbf{R}' \frac{1}{(\mathbf{R}' - \mathbf{R})^3} \right]^{-1} g_{ij}^{(3)} \]  

(73)

From this equation one immediately obtains

\[ \lim_{\mathbf{R} \to \mathbf{R}} X_{ij}(\mathbf{R}) = \left[ \int_{\delta} d\mathbf{R}' \frac{1}{(\mathbf{R}' - \mathbf{R})^3} \right]^{-1} g_{ij}^{(3)} \]  

(74)

As a result last term in (69) can be rewritten in the form

\[ \int_{\delta} d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) X_{ij}(\mathbf{R}', \omega) E_j^{(0)}(\mathbf{R}', \omega) \]

\[ \simeq \int_{\delta} d\mathbf{R}' \frac{g_{ij} g_{ij}^{(3)}}{(\mathbf{R} - \mathbf{R}')^3} \left[ \int_{\delta} d\mathbf{R}' \frac{1}{(\mathbf{R}' - \mathbf{R})^3} \right]^{-1} E_j^{(0)}(\mathbf{R}) \]

\[ = E_j^{(0)}(\mathbf{R}) \]  

(75)

It means that self-action field in the frame of effective susceptibility concept will be reduced to calculation of non-singular terms

\[ E_j^{(as)}(\mathbf{R}, \omega) = -k_0^2 \int_{V - \delta} d\mathbf{R}' G_{ij}(\mathbf{R} - \mathbf{R}', \omega) X_{ij}(\mathbf{R}', \omega) E_j^{(0)}(\mathbf{R}', \omega) \]

\[ -E_j^{(0)}(\mathbf{R}, \omega), \quad \mathbf{R} \in V \]  

(76)

Because of effective susceptibility can be calculated in many instances analytically, Eq. (76) can be useful for numerical calculations.

11. APPLICATIONS OF THE EFFECTIVE SUSCEPTIBILITY CONCEPT

To demonstrate the use of discussed concept we will calculate different electrodynamic properties of nano-systems. First, one shall calculate the absorption profiles when inhomogeneous thin nano-composite films with different distributions of inclusions along the film absorbs the incident light beam, dispersion of electromagnetic waves localized at the submonolayer film of nanoparticles situated on the surface and of near- (far-) field images of scanning optical microscopy of nano-objects at the surface.
11. Light Absorption by Thin Nano-Composite Film with Inhomogeneous Distribution of Inclusions

To calculate absorption profiles one should to find the energy of external field which is absorbed by the film per unit of time. The energy of monochromatic radiation of wave vector \( \mathbf{k} \) absorbed by the film can be presented as Joule heat and calculated according to

\[
Q(k, \omega) = \frac{1}{4} \left( |J_1(k, \omega)|^2 + |J_2(k, \omega)|^2 + |E_1(k, \omega)|^2 + |E_2(k, \omega)|^2 \right)
\]

(77)

with local current \( J_i(k, \omega) \) and local field \( E_i(k, \omega) \), which are induced inside the sample by external radiation of the field \( E_i^{(0)}(k, \omega) \). The next designation

\[
\langle \ldots \rangle = \lim_{T \to \infty} \frac{1}{T} \int_{-T/2}^{T/2} dt \langle \ldots \rangle
\]

(78)

which means the averaging over long-time period, is used in Eq. (18), and

\[
\langle \ldots \rangle = \frac{1}{h} \int_{-h/2}^{h/2} dz \langle \ldots \rangle
\]

(79)

means the averaging over film thickness.

Then, making these averagings and taking into account that Eq. (52) can be rewritten in the form

\[
\chi^{(ef)}_{ij}(k, \omega, \omega') = \chi_{ij}(\omega) \Omega_{ij}(k, \omega, \omega')
\]

(80)

with

\[
\Omega_{ij}(k, \omega, \omega') = \left[ \delta_{ij} + k_i^2 \int_{0}^{h} dz' \Delta(k, z', \omega)\chi_{ij}(\omega) \right]^{-1}
\]

(81)

local-field correction factor, one obtains

\[
Q(\omega) = -\frac{i}{4} \omega \left( \chi^{(ef)}_{ij}(k, \omega) \Omega_{ij}(k, \omega, \omega') \Omega_{\omega'}(k, \omega) \right)^* \chi^{(0)}_{ij}(k, \omega, \omega') E^{(0)}(k, \omega, \omega')^* \Omega_{ij}(k, \omega, \omega') \chi^{(0)}_{ij}(k, \omega, \omega')^*
\]

(82)

One should take into consideration that due to in-plane isotropy of the film, the linear response to a local field tensor has the structure

\[
\chi_{ij}(z, \omega) = \begin{pmatrix}
\chi_{11}(z, \omega) & 0 & 0 \\
0 & \chi_{11}(z, \omega) & 0 \\
0 & 0 & \chi_{11}(z, \omega)
\end{pmatrix}
\]

(83)

When absorption of normally incident light (XOZ is the incidence plane), the absorption profile can be written in a form

\[
I_0(\omega) = \frac{1}{2h} \omega \int_{0}^{h} dz \text{Im} \chi_{11}(z, \omega) \Omega_{00}(k, \omega, \omega)|^2,
\]

(84)

where \( I(\omega) = Q(\omega)/I_0(\omega) \), \( I_0(\omega) = |E^{(0)}(\omega)|^2 \) is the intensity of incident beam. As it can be easily seen from Eq. (84), the absorption of external field energy is determined by imagine part of linear response to the local field tensor. The local-field effects, for one's turn, are described by local-field correction factor \( \Omega_{ij}(k, z, \omega) \). To demonstrate the developed approach, we shall calculate the absorption profiles for the thin film with different distributions of inclusions along the film thickness. For example, we consider homogeneous (Fig. 4(a)), Gaussian-like (Fig. 4(b)), and key pattern (Fig. 4(c)) distributions of inclusions inside the film. It was supposed that films of thickness 98 nm consists of Teflon matrix in which the gold spherical inclusions are embedded. The volume part of inclusion was supposed 0.1. The half-width of Gaussian-like distribution and width of the step of key pattern were supposed 0.4 h. The results of calculations of absorption profiles are presented in Figure 5. All curves are normalized on the particle number. We can see that different distributions of inclusions lead to different spectral properties of the nano-composite film. As a result, one can assert that effective susceptibility concept is adequate approach to describe the optical properties of nano-composite thin films with different distribution of the inclusions along the thickness. Because of proposed approach is based on the using of initial susceptibility which can be calculated in the frame of effective medium theory, one easily can calculate the absorption spectra of thin nano-composite films with shell particles or three-component nano-composite thin films.

11.2. Dispersion of Electromagnetic Waves Localized at the Submonolayer Film of Nanoparticles at the Solid Surface

The concept of effective susceptibility can be easily used for description of resonant properties of the nano-systems.

![Sketch of the films with different distribution of inclusions.](image)
Indeed, as it can be seen from Eq. (66), the equation
\[
\text{det}[(\alpha_{\mu}(\omega))^{-1} + n k_0^2 G_{ij}(k, z_\alpha, \omega)] = 0 \tag{85}
\]
describes the condition of existence of the eigenmode in the system under consideration. Here \(\alpha_{\mu}(\omega)\) is the electric susceptibility of a single particle at the surface. This eigenmode is the electromagnetic wave localized at the submonolayer film situated at the surface. Let us suppose that the surface is covered by the particles of ellipsoidal shape. The particles are oriented of its main axis normally to the surface (Fig. 6). In this case the initial susceptibility tensor (linear response of a single particle at the surface) has a form
\[
\alpha_{ij} = \begin{pmatrix}
\alpha_\parallel & 0 & 0 \\
0 & \alpha_\parallel & 0 \\
0 & 0 & \alpha_\perp
\end{pmatrix} \tag{86}
\]

The normal \((\alpha_\perp)\) and lateral \((\alpha_\parallel)\) components of the tensor were calculated in Ref. [53]
\[
\alpha_{\parallel, \perp} = \varepsilon_i \varepsilon_f \frac{(\varepsilon_p - \varepsilon_r)}{(\varepsilon_p + (\varepsilon_p - \varepsilon_r) m_{\parallel, \perp}} L_{\parallel, \perp} \tag{87}
\]

with
\[
L_{\parallel, \perp} = \left[ 1 + \frac{(\varepsilon_p - \varepsilon_r)(\varepsilon_p - \varepsilon_r)}{3(\varepsilon_p + \varepsilon_r)(\varepsilon_p + (\varepsilon_p - \varepsilon_r) m_{\parallel, \perp}} U_{\parallel, \perp} \right]^{-1}
\]
\[
U_{\parallel, \perp} = \vartheta, 2 \vartheta, \quad \vartheta = h_\perp h_\perp^2 / (2 \varepsilon_r)^3 \tag{88}
\]

and \(m_{\parallel, \perp}\) is the depolarization factor, which describes the influence of the particle shape on its electrodynamical properties.\(^{54}\) In the case of the particle shape as a prolate ellipsoid \((h_\perp > h_\parallel)\) the components of depolarization factor have a form\(^{44}\)
\[
m_{\parallel} = \frac{1}{2} (1 - m_{\perp})
\]
\[
m_{\perp} = \frac{h_\perp^2 / h_\parallel^2 - (1 - h_\perp / h_\parallel)}{(1 - h_\perp / h_\parallel)^{3/2}} \times \left( \frac{1}{2} \ln \frac{1 + \sqrt{1 - h_\perp^2 / h_\parallel^2}}{1 - \sqrt{1 - h_\perp^2 / h_\parallel^2}} \right) \tag{89}
\]

Using Cartesian coordinate system in which the XOY plane coincide with the surface of a substrate and wave vector of “surface” wave is directed along OX axes, Eq. (85) reduces to two simple dispersion relations. One of them describes the s-polarized wave
\[
[\alpha_\parallel(\omega)]^{-1} + n k_0^2 G_{\parallel i}(k, \omega) = 0 \tag{90}
\]

Another equation describes a dispersion relations for p-polarized wave
\[
[\alpha_{\perp}(\omega)]^{-1} + n k_0^2 G_{\perp i}(k, \omega) = 0 \tag{91}
\]

This electromagnetic waves are an evanescent waves, localized at the covered by the particles surface. The cover of nano-particles strongly influences on the dispersion properties of surface plasmon polariton. In particular, the additional branches of p-polarized waves arise. There are four branches of dispersion curves when ellipsoid particles cover the surface.\(^{35}\) At that the shift of the dispersion curve of surface plasmon polariton can be observed. In Figure 7 the influence of the surface cover by the particles having shape as prolate ellipsoids \((h_\perp = 2\text{ nm}, h_\parallel = 1\text{ nm})\) on dispersion curve of surface plasmon polariton is shown. Concentration of the particles were chosen \(n = 10^{12}\) part/cm\(^2\). Figure 7(b) demonstrates that small shift of the dispersion curve due to covering layer can lead to rather appreciable shift of the curves of surface plasmon polariton resonance (SPPR), which can be observed experimentally.\(^{44}\) Because the shift of SPPR curve is strongly depended on the shape of the particle, there is a possibility to define the concentration and shape of the particles with measurement of dispersion curves of surface plasmon polariton and SPPR. In particularly, one can think that the experiments of this kind can give some information about albumen state at the surface.
11.3. Modeling of Scanning Optical and Near-Field Optical Microscopy

Scanning Optical Near-Field Optical Microscopy (SOM and SNOM, respectively) turned from an exotic experimental technique into ordinary method for study the micro and nano-systems.21,61,62 The main idea of SOM and SNOM consists in transforming of evanescent field localized at inhomogeneity of the system under consideration to radiation which can be detected and analyzed.62 In accordance with this, some modes of SNOM microscopes are used.62 Let us consider one of these modes more detail. For example let it would be so-called illumination mode of SNOM. The sketch of the SNOM of illumination modes is shown in Figure 8(a). The probe (P) which is a source (S) of illuminating field is scanned along the scanning plane. The field, emitted by the probe, induces the currents inside the object (O). These currents are the repeated sources of the field which is radiated to the far zone and detected by the detector (D). Then, as we shall see below, the dependence of the light intensity at the detector on the probe coordinate characterizes the local-field distribution at the region where the object is situated. The three-dimension pattern of dependence of light intensity on the probe coordinates is usually named as near-field image. There exists other type of the illumination mode of scanning optical microscopy (SOM). In this type of SOM the strong focused Gaussian-like beam plays a role of the source of the illumination light (Fig. 8(b)). As it is clear, the far-field image in this case is the three-dimension pattern of dependence of the light intensity at the detector on the coordinate of the center of the spot of illuminating beam. Then, the main purpose of analyze of SNOM or SOM experimentally obtained near/far-field images is to connect the local-field distribution at the object region with the light intensity detected in the far zone. Because of the field at the detector is defined by currents inside the object

\[ E_i(R, \omega) = -i \omega \mu_0 \int dR' G_{ij}(R, R', \omega) J_j(R', \omega) \]  

The obtaining currents distribution caused by local field is the main purpose of near-field image modeling. Then, the self-consistent local field should be calculated in the frame of effective susceptibility concept. Let us consider the experimental setup of illumination mode SNOM which mainly will be considered here. The probe, which is the source of external field, induces the currents inside...
the object. The currents in ones turn induce the field at the detector in the far zone (See, Fig. 9)

\[ E_i(R_d, \omega) = \int_V d\mathbf{R}' G_{ij}(R_d, \mathbf{R}', \omega) X_j(R', \omega) E^{(0)}(\mathbf{R}', \omega) \]  
(93)

Because a distance between the system probe + object is much longer than linear dimensions \( L \) the system, one can rewrite Eq. (93) in the form

\[ E_i(R_d, \omega) \approx G_{ij}(R_d, R_0, \omega) \int_V d\mathbf{R}' X_j(\mathbf{R}', \omega) E^{(0)}(\mathbf{R}', \omega) \]  
(94)

with \( R_d \) coordinate of detector and \( R_0 \) coordinate of the center of the system probe + object. Obtaining Eq. (94) one taken into account that inequality \( L \ll |R_d - R_0| \) allows us to assume that the diagonal components of photon propagator give the main contribution in the integrand in Eq. (93). Then, the pre-integral factor \( G_{ij}(R_d, R_0, \omega) \) depends only on characteristic of microscope and can be supposed as instrument function and putted as the constant for the experiment. As a result one can write the near-field image in the form

\[ N_{(ij)}(R_{\text{probe}}) = \frac{I(R_{\text{probe}})}{I_0} = \text{const} \left| \int_V d\mathbf{R} X_j(R, \omega; R_{\text{probe}}) \right|^2 \]  
(95)

which emphasizes that to calculate the near-field image, one needs to calculate the effective susceptibility of the object under consideration. Value \( N_{(ij)} \) has a clear physical meaning, namely, it is proportional to the intensity of \( i \)-component of the local field with the illumination field polarized along \( l \)-axis, and is measured in SNOM experiments. Calculating effective susceptibility can be performed analogously demonstrated above. The explicit form of the effective susceptibility in the SNOM is

\[ X_{ij}(\mathbf{R}, \omega; R_{\text{probe}}) = \chi_{il}(\omega) \left[ \delta_{il} + k_0^2 \int_{V_{\text{probe}}} d\mathbf{R}' \Re \delta_{ij}(\mathbf{R}, \mathbf{R}', \omega; R_{\text{probe}}) \chi_{ik}(\omega) \right]^{-1} \]  
(96)

where the generalized photon propagator, which includes both the direct and indirect (via the probe) scattering channels,

\[ \Re \delta_{ij}(\mathbf{R}, \mathbf{R}', \omega; R_{\text{probe}}) = G_{ij}(\mathbf{R}, \mathbf{R}', \omega) - \int_{V_{\text{probe}}} d\mathbf{R}'' G_{ik}(\mathbf{R}, \mathbf{R}'', \omega) G_{jk}(\mathbf{R}_{\text{probe}}, \mathbf{R}', \omega) \]  
(97)

was used. In the framework of developed approach the near-field images of the objects of different shapes can be easily calculated. The advantage of proposed approach consists in analytical solution of self-consistent Lippmann-Schwinger equation via effective susceptibility. It means that numerical calculations are reduced in this case to a simple numerical tabulation. As an example the near-field image of triangular pyramid-like InAs quantum dot on the GaAS substrate is shown in Figure 10 (see, more detail Ref. [64]). It should be noted that calculation of near-field images by direct numerical calculation of Lippmann-Schwinger equation for the object of complicated shape is rather difficult problem connected with a necessity divide the object on the number subvolumes and solution of the system of algebraic equations of high dimension. Proposed approach can be easily used for modeling the nonlinear SNOM, particularly, SNOM at second harmonics. The main idea which is the base of the second harmonic (SH) SNOM consists in establish the structure of the self-consistent current at second harmonic frequency. Namely, the effective SH current inside the object consists of two parts Linear and nonlinear. The linear contribution \( J^{(L)}(\mathbf{R}, 2\omega) \) to the current is driven by self-consistent...

![Fig. 9. Setup of the experiment of illumination mode SNOM.](image)

![Fig. 10. The near-field image of triangular pyramid InAs quantum dot at the GaAs surface.](image)
SH field. The nonlinear contribution $J^{(NL)}(\mathbf{R}, \omega)$ is generated (via SHG) by the self-consistent field at fundamental frequency. Taking into account these circumstances, one should calculate the self-consistent SH field obeyed the equation similar to Lippmann-Schwinger equation, in which $\omega \rightarrow 2\omega$ and instead of external field $E_i^{(0)}$, one should use the incoming field at SH

$$E_i^{(inc)}(\mathbf{R}, 2\omega) = -i2\omega \mu_0 \int d\mathbf{R}' n_i(\mathbf{R}, \mathbf{R}', 2\omega) \chi^{(2)}(\omega, \omega; 2\omega) \times E_i(\mathbf{R}', \omega) E_i(\mathbf{R}', \omega)$$

(98)

where $\chi^{(2)}$ is the local nonlinear (second-order) susceptibility of the object material and $E_i$ is the self-consistent field given as solution of Lippmann-Schwinger equation with photon propagator given by Eq. (97). Proposed approach allows calculate the near- and far-field images at SH (see, for example, Refs. [59, 60, 63]). On the base of developed concept of effective susceptibility a method for calculation of near-field images of semiconductor surface with inhomogeneous electron/exciton distribution can be proposed. In the frame of the concept of effective susceptibility the studies of fast dynamical processes such as the relaxation, transport, and recombination of carriers in subsurface domains of the semiconductor can be provided. Because this requires both as high a spatial resolution as a temporal resolution the ultrafast SNOM (UF-SNOM) should be used. For near-field imaging of semiconductor surface with an inhomogeneous electron/exciton distribution and simulation of the configuration we consider the so-called far-field-pump and near-field-probe geometry. Here the probe pulse is sent through the fiber while the pump is focused down externally on the sample by a far-field lens. Thus the semiconductor surface is illuminated by the strong focused femtosecond Gauss-like pulses. After absorption of pump pulses in the semiconductor, the nonequilibrium carrier distributions of both electrons and holes are generated under the semiconductor surface. In the case of coherent polarizations being neglected, that is, the time resolution of the experiment is less than the dephasing time of the material excitation, the modulating detection in the combined SNOM allows the measured local-field distribution to be considered as a static one. It means that proposed approach of near-field imaging based on concept of effective susceptibility can be used. The UF-SNOM allows study time dependence of local field pattern in strong inhomogeneous structure under semiconductor surface. Let the Gaussian-like light beam pulse acting on the surface generates exciton cloud under surface of Si at vicinity of some inhomogeneity—Si$_1$-$x$Ge$_x$ domain (Fig. 11). Let Ge is distributed under Si surface with the low $u(\mathbf{R}) = u_0 e^{-(x^2/l_x^2) - (y^2/l_y^2) - (z^2/l_z^2)}$

(99)

with $u_0 \in [0, 1]$, $x, y, z$ the linear dimensions of SiGe domain. The presence of Ge leads to varying of energy gap width and dielectric constant $\varepsilon(\mathbf{R}) = 11.7 + 4.5u(\mathbf{R})$

(101)

Exciton cloud generated near the subsurface SiGe domain interacts with the domain and widens. Interaction between the exciton cloud and subsurface domain leads to retraction of the cloud at the region of SiGe localization. As a result, under surface the inhomogeneous distribution of electron density $n(\mathbf{R}, t)$ varying with time is formed. The detail consideration gives the exciton cloud linear response to the local field in the form $\chi^{ex}(\mathbf{R}, \omega, t) = \frac{\pi e^2(\mathbf{R}) \omega_{LT}}{a_0(\mathbf{R}) - \omega - i\gamma} a_b^3 n(\mathbf{R}, t)$

(102)

with $\omega$ frequency of tested light, $a_b$ Bohr radius of exciton, $\gamma$ damping constant, $\omega_{LT}$ exciton transverse-longitudinal splitting, and

$$\omega_b(\mathbf{R}) = E_b(\mathbf{R})/\hbar - E_{ex}$$

(103)

exciton resonance frequency. Here $E_{ex} = e^2/(2\pi a_b)$ is exciton energy. Then, the initial susceptibility of the system “SiGe domain—exciton cloud” can be written as

$$\chi(\mathbf{R}, t) = (\varepsilon(\mathbf{R}) - \varepsilon(\infty))/4\pi + \chi^{ex}(\mathbf{R}, t)$$

(104)

Finally the problem of calculation of near-field pattern at different times reduces to trivial calculation of effective susceptibility of the system with initial susceptibility of Eq. (104). The results of calculations of near-field
The effective susceptibility concept in the electrodynamics of nano-systems

The near-field images were calculated at different time after pumping beam excited the exciton cloud action, namely, at $t = 0, 1.3, 1.7, \text{and } 2.5 \text{ ns}$. The two cases of mutual polarizations of illuminating light field and field detected in the far zone were calculated. The case when the both illuminating and detected fields are polarized parallel (polarization vector is oriented along OX axes) is shown in the left panels of Figure 12. The case of cross-polarization of illuminating and detected fields is shown in the right panels of Figure 12. The intensities are given in the arbitrary units. The dark areas correspond to low field intensity and bright areas correspond to high intensity of the field. One can see, that the evolution of near-field images is strongly connected with the evolution of exciton cloud which is involved in the area of SiGe example on surface diffusion.

**Fig. 12.** The time evolution near-field images of exciton cloud under Si surface near the SiGe domain. The case of parallel polarization of illuminating and detected fields is shown in the left panels. The case of cross-polarization of illuminating and detected fields is shown in the right panels. The time is given in ns.
coefficient, the measurement of near-field images and compare them domain. Because of the evolution is depended on the surface parameters of semiconductor, for with the same calculated in the frame of the present model can give us a possibility to determine the diffusion coefficient and its dependence on coordinate in the surface.

12. CONCLUSION

In this paper we described the general concept of effective susceptibility (or response to the external field) which can be used when the electrodynamic properties of nano-systems are studied. The concept gives some universal approach to study of optical properties different systems, which from electrodynamical point of view can be considered as low-dimension systems. The main idea underlying of the concept is the generalizing of connection between the local field and local current inside the object for the connection between the local current and external field. This generalization allows to directly write the solution of Lippmann-Schwinger equation, which means to solve the electrodynamical problem. In the lot of cases the effective susceptibility can be calculated in an analytical form. Then, the solution of Lippmann-Schwinger equation can be written in the analytical form too. This means that only simple numerical tabulation of the analytically obtained expressions for self-consistent local field calculation is needed. Moreover, in the case when the analytical obtaining of effective susceptibility is impossible, the solution of Lippmann-Schwinger equation can be written formally in the analytical form. Then to calculate the electrodynamical characteristics of a system under consideration one should no used the numerical methods for finding solution of Lippmann-Schwinger equation. As in the previous case, the solution will be reduced to tabulation of (may be very complicating) analytical formulae, because solution is written in the analytical form. Particularly, this circumstance allows to calculate the nearfar-field images of SNOM at second harmonics for different objects.

One should to note that developed concept has one annoying feature which is connected with determination of eigenmodes in the nano-systems. Namely, the effective susceptibility of a mesoparticle is written in the form of Eq. (16). One can easily see that the pole part of the effective susceptibility depends on coordinate \( \mathbf{R} \). It means that the equation \( \text{Re} \{ \gamma_0 k^2 + \int V \mu \mu^* \mathbf{R} \} = 0 \) does not define the eigenmodes in the system under consideration. The attempt of determination of eigenmodes in the nano-system using the effective susceptibility of the form of Eq. (16) was made earlier, when the absorbed energy of external field by nano-system was analyzed.\(^{33}\) But some questions remain open and additional study is needed to clarify this problem.

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APPENDIX

Functions \( A^\pm \) defining the effective susceptibility of the film have the next form

\[
A^+ = \frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 - \eta_1)} - \frac{(\varepsilon_2 - \varepsilon_1)\gamma}{2\Delta \eta_2} \times \left[ \frac{1}{\eta_2 - \eta_1} \left\{ \gamma(b^+b^- - a^+a^-) - a^- \right\} \right. \\
\left. - \frac{b^-e^{i(\eta_2 + \eta_1)b\hbar}}{(\eta_2 + \eta_1)} \right]
\]

\[
A^- = -\frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 - \eta_1)} e^{i(\eta_2 + \eta_1)b\hbar} \\
- \frac{(\varepsilon_2 - \varepsilon_1)\gamma}{2\Delta \eta_2} \times \left[ \frac{e^{i(\eta_2 + \eta_1)b\hbar}}{(\eta_2 + \eta_1)} \left( \beta(b^+b^- + a^+a^-) + a^- \right) + \frac{b^-e^{i(\eta_2 + \eta_1)b\hbar}}{(\eta_2 + \eta_1)} \right]
\]

where \( \Delta = (1 + a^+b)(1 + a^-b) + b^+b^-\beta\gamma \),

\[
a^+ = \frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 + \eta_1)} \left[ e^{i(\eta_2 + \eta_1)b\hbar} - 1 \right], \quad \beta = \frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 + \eta_1)} \\
b^+ = \frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 + \eta_1)} \left[ e^{i(\eta_2 + \eta_1)b\hbar} - 1 \right]
\]

\[
\gamma = \frac{(\varepsilon_2 - \varepsilon_1)k_0^2}{2\eta_2(\eta_2 + \eta_1)} e^{i(\eta_2 + \eta_1)b\hbar}
\]

References

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