The thickness dependence of dielectric permittivity in thin films

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Abstract. It is well known that the physical properties of thin films depend on their thickness. For a description of such dependences, it is proposed to use a classical model taking into account the presence of film interfaces. A dielectric ball near the half-space was chosen to adopt the approach. The dependence of the effective permittivity of the ball on geometrical and physical parameters of the system is analyzed. It is demonstrated that the dielectric constant of a film can be presented as a sum of the constant of a bulk material and the interface term.

1. Introduction

The classical theory of the dielectric constant (Lorentz-Lorenz, Weiss and others [1]) are based on the expressions for the dipole moment of the sphere (ball) or spheroid placed in a uniform electric field. Obviously, as the sphere is in an infinite space, the effect of boundaries in these models is not considered. In order to account for this issue, it is necessary to examine another model problem – a dielectric ball near the flat surface. This case was repeatedly discussed in the literature (see, e.g., [2]). However, it is possible to obtain an explicit solution only for a particular scenario of the metal sphere [3]. In general, the problem is reduced to a system of difference equations whose solution is given by an infinite product [2]. Due to the complexity of numerical calculations according to the above scheme, it seems convenient to find another solution of the problem. In this study, the approach adopted for this purpose is based on the method of inversion [1].

The method of inversion allows to consistently determine the electric field of a ball or cylinder near the surface. Note that spheroids reproduce the inclusions shape more precisely. Nonetheless, an explicit analytical solution is unknown even for prolate spheroids. In view of this, we consider two bodies simultaneously, as both a ball and cylinder are the extremes of the spheroid. If the dependence of the dielectric constant on the film thickness is known for the both types of inclusions, then for the inclusions of an arbitrary shape this dependence can be found by using the shape factor interpolation, as will be explained below.

2. The statement of the problem

Consider the problem of finding the electrostatic potential of the electric field $u$ from the dielectric sphere located near a semi-infinite dielectric medium in an external electric field $u_{ext} = -E_x x - E_y y$ (see, figure 1). Here $E_{x,y}$ are components of an external electric field. We are interested in the dipole moment $p$ of the described ball, or rather, in the impact of a flat surface on this moment. The potential of the electric field $u$ satisfies the Laplace equation, boundary conditions of continuity of the normal components of the electric displacement at the media interfaces, and tends to $u_{ext}$ for an increasing distance from the origin. The geometry of the problem is greatly simplified if we perform an inversion
of the coordinate system [1] with respect to a specially selected point on the symmetry axis and a coordinate system shift. In this way, the eccentric sphere transforms into concentric one, figure 1. Similarly, the eccentric cylinder becomes concentric. Under this transformation the equation and boundary conditions describing the field do not change, and the external uniform field becomes a dipole field. As far as the dipole field may be simply expressed through the field of a point source (charge), the initial problem is reduced to finding a field of a point charge for three-layer media, where the boundaries between the balls are concentric spheres, figure 1 (bottom). Unfortunately, the potential is changed in the case of sphere [1]. After the inversion the Laplace equation is not fulfilled for the potential in the stretched variables, but for a new function that is different from the potential by the factor $F(\rho, \theta) = (\rho^2 + \rho_0^2 + 2\rho\rho_0 \cos \theta)^{1/2}$. Here we introduced a spherical coordinate system $(\rho, \theta, \phi)$ whose origin is located in the centre of the concentric spheres or cylinders, while $\rho_0$ is the distance to the point at which, after inversion, transforms infinitely distant point. The potential function $U$ is sought as a series in Legendre polynomials

$$U(\rho, \theta, \phi) = \sum_{m=0}^{\infty} P^m_n(\cos \theta) \frac{U^m_n}{\rho^{m+1}} + V^m_n(\rho^n) \delta(\phi).$$

In the above expression we have $m=0$, $\delta(\phi)=1$ for the field parallel to the $z$-axis and $m=1$, $\delta(\phi)=\sin \phi$ for the perpendicular one. The series (1) satisfies the Laplace equation with arbitrary coefficients $U^m_n$. That is, the problem is reduced to the determination of three pairs of sets (for the three regions under examination) for the coefficients $U^m_n$ and $V^m_n$. In the outer space $\rho > r_2$, the coefficients $V^m_n$ are known and defined by the external field. In addition, the coefficients $U^m_n$ must be equal to zero in the inner sphere $\rho < r_1$ because of the limitation of the potential at the centre of the coordinate system. There exist four boundary conditions to find the rest four sets of unknown functions. The equality of the potentials at the interfaces $\rho = r_1$ and $\rho = r_2$ makes possible to express two sets of coefficients through the remaining ones. Difficulties arise when considering the second pair of boundary conditions containing derivatives of $\rho$. Due to the presence of the factor $F(\rho, \theta)$, the boundary conditions become dependent on $\cos \theta$. This leads to the need to solve the system of difference equations. These equations can be solved by performing the Fourier transform. As a result, it manages to obtain explicit equations for the desired potential.

**Figure 1.** The geometry of the direct (top) and inverse (bottom) problems.

**Figure 2.** The dependence of the normalized dipole moment on the distance from the interface for the cylinder and sphere. The calculations were performed for a series of $\varepsilon_{\text{ext}}$ at $r_0=60$ nm, $\varepsilon_1=1$, $\varepsilon_s=4$. The solid and dotted lines reflect the electric field perpendicular and parallel to the $x$ axis.
3. Results and discussion

The derived expressions enable us to calculate the required dipole moment of the ball located near the interface, and indicate correction terms to the dielectric constant caused by the presence of the plane. Apparently, the dipole moment $p$ is dependent on the distance from the ball to the plane and the direction of the electric field. Such behaviour is illustrated in figure 2. The dielectric constant of the film $\varepsilon$ is completely determined by the dipole moment, which depends on the distance from the film interface, i.e., on the coordinate $p=p(x)$. As a consequence, the film permittivity also should be dependent on the distance to the surface $\varepsilon=\varepsilon(x)$. At large distances from the film interface, the permittivity must tend to a constant value $\varepsilon(\infty)=\varepsilon_{\text{bulk}}$, where $\varepsilon_{\text{bulk}}$ is the dielectric constant of a bulk material. Moreover, because of the corresponding behaviour of the dipole moment, the dielectric constant becomes a tensor. There appear the dielectric permittivities $\varepsilon_\parallel$ and $\varepsilon_\perp$ for the electric field perpendicular and parallel to the $x$ axis. The calculation results are presented in figure 3. The dipole moment of the ball varies largely at short distances from the ball to the dielectric surface. Its maximum value is achieved at the contact of the interface with the ball. The situation is similar for cylinder, but the change of the dielectric constant is more drastic than that of the sphere. This finding is due to the greater area of the interaction between the cylinder and interface.

Such an approach is not always convenient to describe the electrical properties of the film. Therefore, we use the theory of surface phenomena and introduce the Gibbs surface permittivity [4]

$$\varepsilon_{\text{surface}} = \int_0^\infty (\varepsilon(x) - \varepsilon_{\text{bulk}}) \, dx.$$  \hspace{1cm} (2)

Hence, the dielectric constant of the film can be replaced by the constant $\varepsilon_{\text{ef}}$ given by

$$\varepsilon_{\text{ef}} = \frac{\varepsilon_{\text{surface}} \cdot S}{V} + \varepsilon_{\text{bulk}} = \frac{k \cdot \varepsilon_{\text{surface}}}{R} + \varepsilon_{\text{bulk}},$$  \hspace{1cm} (3)

where $k=S\cdot R/V$ is a dimensionless parameter characterizing the shape of an object (shape factor), $R$ typical size of an object (distance from the surface to the most remote interior point of the body). A film of the thickness $2R$, cylinder or sphere of radius $R$ possesses a shape factor 1, 2, and 3.
respectively. Thus, the dielectric constant of the film depends on its thickness as $\varepsilon_{\text{ef}} = \varepsilon_{\text{bulk}} + \varepsilon_{\text{surface}} k/R$. Naturally, this ratio may be employed for $R > r_0$. The applicability of (3) requires the smallness of the ball size compared to the dimensions of the object. For example, the size of the modelling sphere in the case of crystalline films must be the order of the lattice constant, i.e., several angstroms. Consequently, the film thickness must exceed 2-4nm. Since the shape of inclusions is, as a rule, different from a regular (spherical and cylindrical), it is possible to use an interpolation approach via the shape factor $2 < k \leq 3$ for simulation of inclusions of arbitrary shape. The dependence of $k$ on the axis ratio for a prolate spheroid is shown in figure 4. The consideration of the shape factor allows to model various types of inclusions. The inclusions are strictly spherical for $k = 3$. With the reduction of $k$, their shape becomes more elongated, and with $k = 2$ inclusions transform into cylinders. It is worth to mention that the proposed strategy can be generalized to multilayer cylindrical and spherical inclusions by the matrix homogenization method [5]. Furthermore, the results obtained can be used as a generalization of the Weiss model for ferroelectric thin films [6].

4. Conclusion
The dielectric constant of the film depends on its thickness in a simple way predicted by the developed model. The approach considers polar dielectric regions by inclusions of different shape. The dielectric permittivity varies most strongly (~5%) for cylindrical inclusions. In turn, the most significant variation of the dipole moment is demonstrated by the sphere in contact with the interface. Accounting for the finite film thickness leads to the anisotropy of the dielectric constant. Even if the film is made of a homogeneous material, its dielectric constant depends on the direction of the electric field. For a film thickness of 10nm, the difference of dielectric permittivities $\varepsilon_{\perp,\text{ef}}$ and $\varepsilon_{\parallel,\text{ef}}$ is about 1%. The presented equations can be used to find the dielectric constant of the mixture and allow to describe the properties of polycrystalline films as a function of the size and shape of the grains. The more exact expressions must take into consideration the influence of the elastic field and the impact of the ball’s field on the interface dielectric permittivity.

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