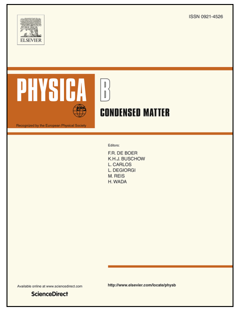
Accepted Manuscript



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PII: S0921-4526(18)30441-1

DOI: 10.1016/j.physb.2018.06.041

Reference: PHYSB 310949

To appear in: *Physica B: Physics of Condensed Matter*

Received Date: 16 May 2018

Accepted Date: 29 June 2018

Please cite this article as: V.Z. Lozovski, M.A. Razumova, Optical response of thin nanocomposite films with transverse inhomogeneity, *Physica B: Physics of Condensed Matter* (2018), doi: 10.1016/j.physb.2018.06.041.

Optical response of thin nanocomposite films with transverse inhomogeneity

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Abstract:

In-plane homogenization, taking into account inhomogeneity in distribution of nanoinclusions across the film thickness, was performed within the framework of self-consistent approach based on the concept of effective susceptibility and the Green function method. We apply this approach to obtain the optical response of films consisting of metal nanoparticles in a dielectric matrix that exhibits plasmonic absorption. We have calculated the absorption spectra of nanocomposite films with gold spheroidal inclusions in Teflon matrix at a silicon substrate for several different transversal distributions of inclusions, maintaining a constant inclusion volume fraction. The absorption dependence on the light polarization and incident angle, as well as on the shape of inclusions and their orientation is also under study. From the absorption maps we have extracted dispersion curves for plasmon-polariton resonances in the nanocomposite films.

Keywords: Nanocomposite; effective susceptibility; thin film; local field; absorption; dispersion relations

PACS number(s)**:** 78.66.Sq; 78.67.Sc; 78.20.-e; 78.66.-w

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1. Introduction

Nanocomposite thin films find extensive applications in sensorics [1, 2], solar cells [3-5], and highly absorptive films for solar harvesting [6]. The influence of morphology of nanocomposite thin films on their optical properties is studied in numerous works (see, e.g., [7-11]). The optical properties of nanocomposite thin films are commonly modeled by effective medium theories (EMT), which approximate a complex electromagnetic medium (nanocomposite) with an effective homogeneous medium described by some effective dielectric function, using as an input the dielectric functions of the constituents and their respective volume fractions.

However, in many studies it has been found that a direct use of EMT for analysis of optical properties of thin nanocomposite films encounters difficulties [8-10, 12, 13]. For instance, in Ref. [8] Au/TiO_2 nanocomposite thin films have been studied as a part of anti-reflective coating in silicon solar cells, and it was demonstrated that extinction and reflectance spectra of silicon coated by the plasmonic films cannot be explained by the Maxwell Garnett effective medium theory. In Ref. [9], single-layer mixture coatings of ZrO_2–SiO_2 and Nb_2O_5–SiO_2 produced by the ion beam sputtering deposition technique were prepared and investigated. The Bruggeman, Maxwell Garnett and Lorentz-Lorenz EMTs were applied for characterization of elemental composition of these films, and the corresponding estimates exhibited controversial results. In Ref. [10], the absorption spectra of nanocomposite films of Au/Teflon, obtained by thermal vacuum deposition, were obtained and analyzed taking into account the local field interaction: it was found that such a semi-phenomenological approach, in contrast to the standard EMT treatment, gives a possibility to draw conclusions about the concentration, shapes and shape distributions of the inclusion particles. In Ref. [11] Ag/SiO_2 nanocomposite thin films obtained by sputtering were studied by means of the spectroscopic ellipsometry, and the results were found to be consistent with those obtained from optical absorbance data. The Bruggeman effective medium theory was used to explain the experimental results. While the authors of Ref. [7] successfully used EMT for a description of ellipsometric measurements for a film consisting of silver nanoparticles embedded in a dielectric matrix, other authors [12], conversely, reported that a direct use of EMT to describe optical properties of thin films does not give correct results. Moreover, the same authors that claimed a successful application of EMT in Ref. [7] in other works [13] conclude that EMT is not valid for nanocomposite systems.

These discrepancies, in our view, stem from the effects related to contributions of out-of-plane (normal to the film plane) interactions (between nanoparticles inside the matrix as well as between nanoparticles and interfaces). If the nanocomposite is arranged so that these contributions play a significant role, the application of the EMT fails. Indeed, a general prerequisite for the applicability of the EMT is that the linear dimensions of inhomogeneities or inclusions and characteristic distances between them are much smaller than the thickness of the film, so that the following scale hierarchy is satisfied:

$$d_p < l << L << d \sim D \leq \lambda \,, \tag{1}$$

where d_p is a characteristic linear dimension of the inclusions, l is a characteristic distance between particles of the composite (mean distance between neighboring particles), d is the thickness of the film, D is a characteristic length of the