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Anisotropic Electromagnetic Properties of RF Sputtered Ni–Al$_2$O$_3$ Composite Thin Films

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Abstract

Experimental data of the permittivity of RF sputtered Ni-Al$_2$O$_3$ thin films were studied in the wavelength region from 300 to 2500 nanometers [1]. The data were analyzed in the framework of the Bergman-Milton theory and it was shown that they lie inside the Hashin-Shtrikman bounds, but outside the Bergman-Milton bounds. The latter indicates a strong electromagnetic anisotropy of the composite thin film. For a more detailed theoretical study, we performed calculations using the Maxwell Garnett, Bruggeman and Incremental Maxwell Garnett homogenization formalisms for uniaxial dielectric composites [2–4]. A comparison between the calculated and measured permittivities was made on the basis of a least-square fit. In this paper, we interpret the results and suggest models for the microstructures of the Ni-Al$_2$O$_3$ thin films.

1. Introduction

The dielectric properties of composite thin films made of a mixture of two particulate materials are of great practical interest with potential applications in many disciplines of science and technology. Composite thin films can be fabricated by a variety of vacuum techniques. In order to obtain the desired optical properties, the film microstructure has to be optimized in terms of concentration, size and shape of the inclusions. If the nonhomogeneities of the composite thin film are electrically small, homogenization theory can be used to obtain estimates of its effective permittivity, provided the permittivities of the component materials and some details about its microstructure are known ("direct problem"). Conversely, if the effective permittivity of the composite thin film has been determined experimentally, homogenization formalisms can be applied for microstructural characterization ("inverse problem").

The Bergman-Milton theory [5, 6] provides rigorous bounds for the effective permittivity in the complex plane [7] and therefore is an important tool for a preliminary analysis of experimental data [8]. Homogenization formalisms, such as the Maxwell Garnett and Bruggeman formula and their extensions, can be used for more detailed studies.

Due to the axial symmetry of the thin film geometry, we expect its effective permittivitiy dyadic $\underline{\underline{\epsilon}}^{\text{eff}}$ to be uniaxial

$$\underline{\underline{\epsilon}}^{\text{eff}} = \epsilon_0 \left[\epsilon_u^{\text{eff}} \underline{u}\,\underline{u} + \left(\underline{\underline{I}} - \underline{u}\,\underline{u} \right) \epsilon_t^{\text{eff}} \right], \tag{1}$$

with the free-space permittivity ϵ_0, the unit dyadic $\underline{\underline{I}}$, and the unit vector $\underline{u}$ perpendicular to the surface of the thin film; ϵ_u^{eff} and ϵ_t^{eff} are the axial and transverse components of the relative

effective permittivity dyadic, respectively. The electromagnetic anisotropy of the film may be caused by an anisotropic growth of the film on the substrate, by the non-spherical shape of the particulates the film is made of, and by an anisotropic spatial arrangement of these particulates. Homogenization formalisms that are able to take the anisotropy into account have become available only recently; see [9] for a review. The main intention of this paper is to interpret measured permittivities obtained for Ni-Al_2O_3 composite thin films in the framework of these formalisms.

2. Theory

The Al_2O_3 host material of the thin film is taken to be uniaxial dielectric with a permittivity dyadic $\underline{\underline{\epsilon}}^a$ of the form

$$\underline{\underline{\epsilon}}^a = \epsilon_0\epsilon^a \left[\gamma\underline{u}\underline{u} + \left(\underline{\underline{I}} - \underline{u}\underline{u}\right)\right],\tag{2}$$

where the parameter γ specifies the degree of anisotropy. The permittivity dyadic $\underline{\underline{\epsilon}}^b$ of the inclusions (Ni particulates in our case) is isotropic:

$$\underline{\underline{\epsilon}}^b = \epsilon_0\epsilon^b\underline{\underline{I}}.\tag{3}$$

For the reasons discussed before, the effective permittivity dyadic $\underline{\underline{\epsilon}}^{\text{eff}}$ of the composite thin film is assumed to be uniaxial, having the form specified in (1).

Let us first assume that the host material is isotropic ($\gamma = 1$ in equation (2)). The Bergman-Milton theory is then applicable. Accordingly, the scalar relative permittivities ϵ_t^{eff} and ϵ_u^{eff} must lie inside a region of the complex permittivity plane which is bounded by the so-called Hashin-Shtrikman (HS) bounds. Failure of measured permittivities to fulfill this constraint indicates serious inconsistencies of the data. If the microstructure of the composite medium is isotropic, the effective permittivity will be isotropic too ($\epsilon_u^{\text{eff}} = \epsilon_t^{\text{eff}}$). The value of the effective permittivity can then be further restricted by the so-called Bergman-Milton (BM) bounds. If the measured effective permittivities lie outside the BM bounds, the thin film must be anisotropic.

Homogenization formalisms can be used to get a more detailed description of the experimental data. For dilute composites in which one component exists as inclusions embedded in the other one, the Maxwell Garnett (MG) formalism is expected to produce good results. For non-dilute, mutually isolated inclusions, the Incremental Maxwell Garnett (IMG) formalism (or, alternatively, the Differential Maxwell Garnett formalism) is an appropriate choice [4], whereas for composites with percolated inclusions the Bruggeman (Br) formalism is recommendable. In these homogenization formalisms, anisotropy can be implemented in two ways: by using uniaxial permittivity dyadics for the component materials ($\gamma \neq 0$ in eq. (2)), and by assigning the host and inclusion particulates a spheroidal effective shape with a certain aspect ratio $a : b$, the latter quantitiy being > 1 for prolate and < 1 for oblate spheroids (and 1 for spheres). The effective shape is a parametrization of both shape and relative spatial arrangment of the particulates.

3. Experiment

The Ni-Al_2O_3 composite films were deposited in a planar magnetron assisted RF sputtering system. The fill factor f of Ni was found to be 0.21, 0.42 and 0.61 in the examples studied here. Detailed reports on the experimental set-up and characterization techniques have been given elsewhere [10]. The optical constants n and k of the composite films in the wavelength range from 300 to 2500 nm were calculated from the measured near-normal incidence reflectance (R), transmittance (T) and thickness (t) values as described elsewhere [1]. Because of the near-normal incidence technique used, the electromagnetic field is approximately parallel to the surface of the film; therefore only the transverse component of the relative permittivity ϵ_t^{eff} can be deduced from this experiment. The relation between n, k, and ϵ_t^{eff} is $\epsilon_t^{\text{eff}} = (n + ik)^2$.

4. Results and Discussion

For all fill factors and wavelengths, the measured permittivities were compared with the HS and BM bounds. All experimental data lie within the HS bounds, but outside the BM bounds. The latter provides evidence that the composite is anisotropic. Typical results for various fill factors f are shown in Figure 1, where we used the optical constants for Ni and Al_2O_3 at wavelength $\lambda = 500$ nm. Assuming isotropy ($\gamma=1$, $a{:}b=1$), we computed the MG, Br, IMG estimates, which turned out to lie far off the experimental results (Figure 1). Allowing for an oblate spheroidal effective shape of the particulates ($a : b < 1$), the agreement between theory and experiment is greatly improved. This is also shown in Figure 1 in the case of the IMG estimate.

A further comparison between the calculated and measured permittivites of the thin film was made on the basis of a least-square fit, minimizing the quantity

$$\chi^2 = \sum_i \left| \epsilon_t^{\text{eff}}(\lambda_i) - \epsilon_t^{exp}(\lambda_i) \right|^2, \tag{4}$$

where the sum extends over all measured wavelengths λ_i, ϵ_t^{exp} is the measured and ϵ_t^{eff} the computed value of the transverse component of the permittitivity. Assuming that the Al_2O_3 host material is anisotropic ($\gamma \neq 0$ in equation (2)) does not lead to a significant improvement of the fit between experiment and calculations. The effective shape of the particulates, however, significantly influences the quality of the fit.

For $f = 0.21$ and $f = 0.42$ the IMG formalism performs best while for $f = 0.61$ the Br formalism leads to the smallest χ^2-values (Figure 2). The best fit is obtained, when the *effective* shape of the particulates is oblate spheroidal with aspect ratios in the range from 0.1 to 0.6, depending on the fill factor. This strong dependence on the fill factor cannot be explained by assuming a non-spherical shape of the particulates itself. Rather it must be due to their anisotropic arrangement in the film. A possible interpretation of these findings is that the Ni inclusions form planar structures parallel to the surface of the thin film. For larger fill factors ($f = 0.62$) percolation sets in and therefore the Br estimate is in better agreement with experiment.

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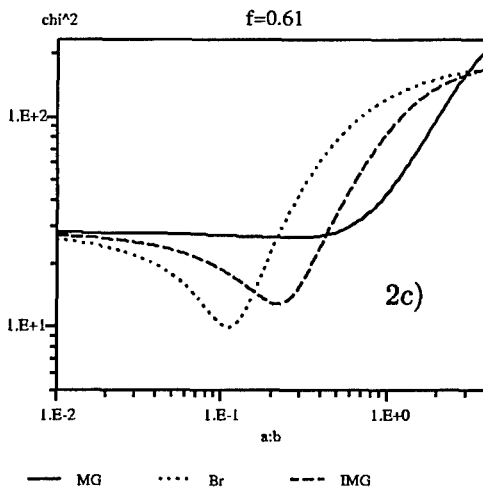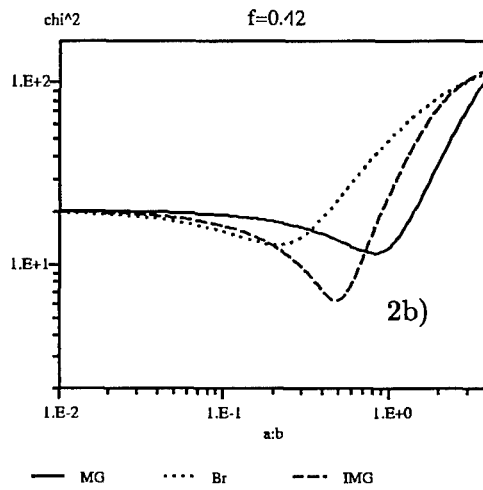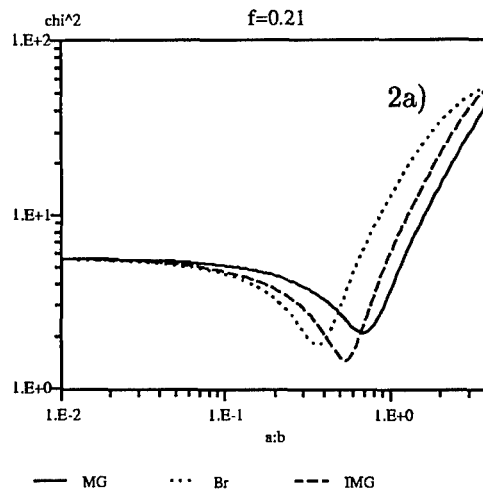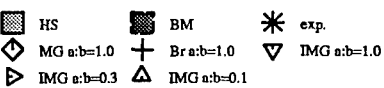
Img[eps] lambda=500 nm, f=0.21

1a)

Re[eps]

HS	BM	exp.
MG a:b=1.0	Br a:b=1.0	IMG a:b=1.0
IMG a:b=0.5	IMG a:b=0.1	

Img[eps] lambda=500 nm, f=0.42

1b)

Re[eps]

HS	BM	exp.
MG a:b=1.0	Br a:b=1.0	IMG a:b=1.0
IMG a:b=0.4	IMG a:b=0.1	

Img[eps] lambda=500 nm, f=0.61

1c)

Re[eps]

HS	BM	exp.
MG a:b=1.0	Br a:b=1.0	IMG a:b=1.0
IMG a:b=0.3	IMG a:b=0.1	

chi^2 f=0.21

2a)

a:b

—— MG   ··· Br   - - - IMG

chi^2 f=0.42

2b)

a:b

—— MG   ···· Br   - - - IMG
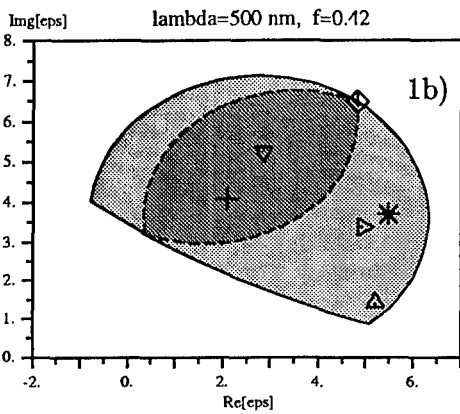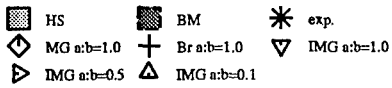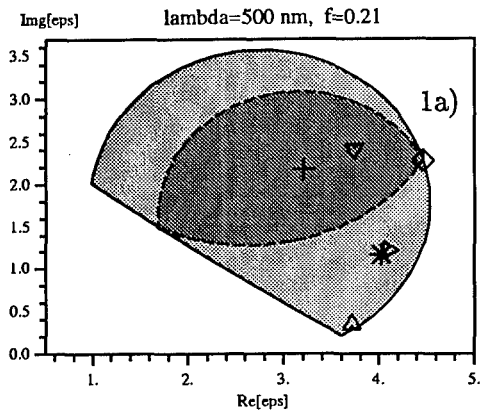
chi^2 f=0.61

2c)

a:b

—— MG   ···· Br   - - - IMG
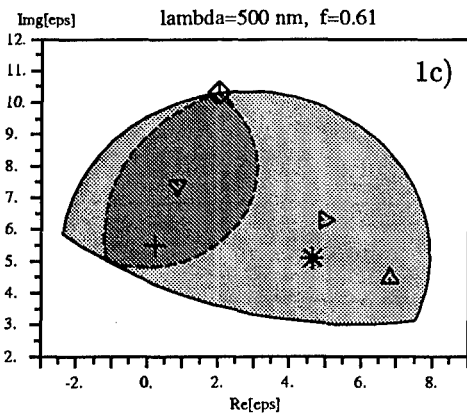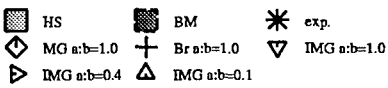
Figure 1: Comparison of experimental data (exp.), HS and BM bounds, and various homogenization estimates calculated for different aspect ratios $a : b$.
a) $f = 0.21$, b) $f = 0.42$, c) $f = 0.61$.

Figure 2: χ^2 as function of the aspect ratio $a : b$ for the MG, Br, and IMG homogenization formalisms.
a) $f = 0.21$, b) $f = 0.42$, c) $f = 0.61$.