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The effective medium and the average field approximations vis-à-vis the Hashin–Shtrikman bounds. II. The generalized self-consistent scheme in matrix-based composites

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ABSTRACT

The present Part II of this two-part study is concerned with the average field approximation (AFA), and the effective medium approximation (EMA) in two-phase matrix-based dielectric composites through the use of an auxiliary configuration in which a particle of the inclusion phase is first surrounded by some matrix material, and then embedded in the effective medium. Those models will be referred as the generalized self-consistent scheme-average field approximation (GSCS-AFA), and the generalized self-consistent scheme-effective medium approximation (GSCS-EMA). We show that there are four types of the GSCS-AFA and a single type of the GSCS-EMA. In this paper the application of those models to dielectric composites with isotropic constituents and an inclusion phase that consists of randomly oriented ellipsoidal particles will be studied. The analytical solution of the auxiliary problem, which consists of an ellipsoidal particle confocally surrounded by a matrix shell and embedded in the effective medium, is achieved by means of ellipsoidal harmonics. Our results show that the effective property predictions of the GSCS-EMA and GSCS-AFA for the considered systems differ from each other, and more importantly, out of the four GSCS-AFA models, three of them violate the Hashin-Shtrikman bounds. The predictions of the GSCS-EMA obey the bounds. It is then shown that the version of the GSCS-AFA which obeys the Hashin-Shtrikman bounds for an inclusion phase with randomly oriented ellipsoids will violate them in the case of a particle shape which is not simply connected. Moreover, it turns out that the SCS-AFA studied in Part I also violates the Hashin-Shtrikman bounds in that case; the EMA, as expected, owing to its realizability property, continues to obey the bounds. Among the AFA and EMA in matrix-based composites, the GSCS-EMA therefore stands out as the method to be recommended.

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1. Introduction

As indicated in Part I of this two-part study, the application of the average field approximation (AFA) in two-phase matrix-based composites can be done through two different embedding configurations. The first configuration consists of a particle of the inclusion phase embedded in the effective medium. That assembly is then subjected to a uniform field at

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infinity and the average field induced in the embedded particle is used to approximate the actual average field in the inclusion phase of a composite sample that is under the same uniform field. This leads to an implicit equation for the sought effective property. In Part I, this type of AFA was called the self-consistent scheme-average field approximation (SCS-AFA). The second configuration, which is more appropriate to matrix-based composites with well-separated inclusions, consists of a grain of the particulate phase first surrounded by some matrix material and then embedded in the effective medium. The use of that configuration in the application of self-consistent models in the setting of different physical phenomena (viscous dispersions, conductivity, and elasticity) are encountered, for example, in Fröhlich and Sack (1946), Mackenzie (1950), Kerner (1956a,b), Van der Poel (1958), Smith (1974, 1975), Hashin (1968, 1990), Christensen and Lo (1979, 1990), Miloh and Benveniste (1988), and Benveniste (1985, 2008) where additional references on the subject can be found.

In reference to the self-consistent models which use the coated particle embedding configuration, Hashin (1968) introduced the term 'the generalized self-consistent scheme' (GSCS), whereas Christensen and Lo (1979) used the term 'the three-phase model'. In the present study we clarify that this matrix-coated particle embedding configuration can be implemented either in the spirit of the average field approximation or in the spirit of the effective medium approximation. The affiliated models will be called here the GSCS-AFA and the GSCS-EMA, respectively. The GSCS-AFA aims at estimating the average fields in the phases in the actual composite sample through the above described embedding configuration. The GSCS-EMA on the other hand demands the vanishing of the dominant far-field part of the disturbance caused to the uniform field, which was present in the effective medium before the embedding with the coated particle was made. The studies of Mackenzie (1950), Van der Poel (1958), and Smith (1974, 1975) invoke certain considerations, introduced by Fröhlich and Sack (1946) about the far-field behaviour of the fields in the coated particle configuration, which are akin to the assumptions of the GSCS-EMA. Christensen and Lo (1979), on the other hand, apply the assumption that the internal elastic energy in the effective medium does not change as a result of the insertion of the coated particle in it. It turns out that this demand results in the same condition used by Smith (1974, 1979) about the far-field behaviour; thus all those studies could be safely classified under the category of GSCS-EMA. On the other hand, the articles by Hashin (1968, 1990), Miloh and Benveniste (1988), and Benveniste (1985, 2008), among others, use the GSCS-AFA. As shown by Benveniste (1985) and Hashin (1990), in the case of matrix based composites with spherical particles or parallel cylindrical fibers of a circular cross-section, the GSCS-AFA gives results that coincide with those provided by the approach of Christensen and Lo (1979), which is essentially equivalent to that of the GSCS-EMA. Yet, being based on different assumptions, both methods could be expected to give different predictions for other particle shapes. We finally mention here that a configuration akin to the that used in the GSCS has been used by Hori and Nemat-Nasser (1993) in their double-inclusion model whose relation with other micromechanics models was discussed by Hu and Weng (2000).

It should be noted that the embedding configuration used in the GSCS-AFA allows one to compute the average electric fields *both* in the core and the coating of the embedded particle. Possible identification of those values with the corresponding actual average fields in the inclusion and matrix phases in a sample of the composite leads to four different versions of the model. It was recently shown by Benveniste (2008) that those four versions of the GSCS-AFA give identical results if the inclusion phase consists of spherical particles, provided that the ratio of the volume of the core to that of the coated embedded entity is chosen to be equal to the volume fraction of the inclusion phase in the actual composite sample. We will show in this paper that for the case of randomly oriented ellipsoidal particles, the predictions of the above versions of GSCS-AFA will differ from each other even if the embedding is done in the above described manner. It will furthermore be shown in that some of those versions will violate the Hashin–Shtrikman (1962) bounds, to be denoted here as the HS-bounds.

In contrast to the above described behaviour of the GSCS-AFA, and we point out that there exists a single type of the effective medium approximation (GSCS-EMA) which, as expected, will always obey the HS-bounds since it has been shown to be realizable (Milton, 1984, 1985; Avellaneda, 1987). In the latter reference, the application of the EMA in matrix-based composites through a coated particle configuration was indicated, and implemented in the derivation of differential coated sphere and cylinder models. On the other hand, the implementation of the EMA to granular aggregates with randomly oriented ellipsoidal grains has been given by Berryman (1980). To the best of our knowledge, our present study is the first implementation in the literature of the EMA to matrix-based composites containing randomly oriented ellipsoidal particles, through the auxiliary configuration consisting of a particle which is surrounded by some matrix material and embedded in the effective medium.

In Section 2 of the present part, the formulation of the GSCS-AFA and GSCS-EMA is given in two-phase matrix-based dielectric composites with isotropic constituents, while the inclusion phase is of arbitrary simply-connected shape. In Section 3 the GSCS models are applied to the case in which the inclusion phase consists of randomly oriented ellipsoidal particles. Since the electric potential satisfies Laplace's equation, the solution of the auxiliary problem of the embedded coated ellipsoid is carried out conveniently in the setting of ellipsoidal coordinates and through the use of the Lamé functions (Hobson, 1955). In Section 3 and in Appendix A, following the analysis existing in Miloh and Benveniste (1988), we give a solution of that auxiliary problem that produces the average electric fields in the core and coating. Numerical results for the effective dielectric constant are given for the case of randomly oriented prolate or oblate spheroidal particles. It is observed that, as expected, the prediction of the GSCS-EMA obeys the bounds. On the other hand, three of the four versions of the GSCS-AFA violate the HS-bounds (the version used by Miloh and Benveniste (1988) is the one that

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