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2001 J. Phys.: Condens. Matter 13 8217

(http://iopscience.iop.org/0953-8984/13/35/326)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **13** (2001) 8217–8234 PII: S0953-8984(01)24213-1

Effective dielectric response of a shape-distributed particle system

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Received 24 April 2001, in final form 6 July 2001
Published 16 August 2001
Online at stacks.iop.org/JPhysCM/13/8217

Abstract

To calculate the effective dielectric response of a dilute composite, a generalization of the Maxwell Garnett theory for small nonspherical particles distributed in shape is proposed. Various types of distribution function are analysed and the applicability of the simplest (steplike) distribution is discussed. It is shown that the use of the steplike distribution is more valid for particles having a higher imaginary part of the permittivity in the actual region. Besides, an alternative approach to the problem based on the spectral representation is also considered. As an illustration, the effective dielectric response of a system of semiconductor (SiC) and metal (Al) ellipsoidal particles is calculated.

1. Introduction

The dielectric response is one of the most important quantities describing the electrical and optical properties of materials. It is of particular interest when studying composite systems. They are the mainstay of advanced engineering structures and their importance continues to grow.

It is well known that most theories on the dielectric response of composite systems use spherical models. Up to now such theories as the Maxwell Garnett (MG) and Bruggeman ones, in spite of their limitations, have been widely used in practical calculations (see, e.g., [1–18]). Their application, however, is under scrutiny rather frequently. Below the MG theory will be considered that is usually applied to dilute suspensions of uniform spheres.

Since the first appearance of the MG theory [19], various corrections to its initial form have been proposed. It is known that this theory completely neglects fluctuations of the local field. Many works take these fluctuations into account [20]. It should be noted that if these fluctuations are strong, then mean-field theories collapse and other approaches to description of the optical properties of composites are to be applied (see, e.g., [21] and [22]). A theory of linear and nonlinear dielectric responses of the Maxwell Garnett composites based on a spectral

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representation is developed in [23]. Mention may be also made of MG theory generalizations for small particles with consideration of their multipole interaction [24–27] and for an ensemble of semiconducting nanoparticles [28], as well as for chiral composites [29–31].

As was mentioned above, both MG and Bruggeman models employ the spherical shape concept. At the same time in actual systems the particles (inclusions) are usually not spheres. It is clear that for the most part the ellipsoidal shape depicts the actual inclusion shape more adequately than the spherical one. Besides, in the actual particle systems the inclusions are usually shape distributed. However, as far as we know, in the literature only the generalizations of the MG equation for ellipsoids of a fixed shape are considered [32–35]. (There are also generalizations of the Bruggeman equation for ellipsoids, see, e.g., [2, 15] and [36–38]. This problem, however, is beyond the scope of our present work.) Galeener [32] has proposed a generalization of the MG equation considering a spherical cavity surrounding the ellipsoid. However, Cohen *et al* [33] have pointed out that Galeener's equation does not reproduce correct results in extreme cases when prolate spheroids approach long cylinders (needles) and oblate spheroids become wide dishes (discs). To account for these extreme geometries, the authors have proposed using an ellipsoidal cavity of the same shape as the inclusions instead of the spherical cavity. Later Fu and Resca [35] considered the above problem in more detail and showed that both results, of Galeener and Cohen *et al*, correspond merely to different choices of pair distributions. These results, however, concern the system of aligned ellipsoids with one of their axes parallel to the applied field. Some problems arising when the ellipsoids are not aligned are considered by Landauer [34]. Another important development in this field belongs to Hayashi *et al* [4], who have considered the case of randomly oriented ellipsoids.

What if the ellipsoids are shape distributed? Earlier we found the effective absorption cross section [39], as well as the effective scattering cross section [40, 41], for a system of small non-interacting ellipsoids and spheroids distributed in shape. An approach by Bohren and Huffman [42] enables one to calculate the effective dielectric response. However, as far as we know, many important problems have not been considered earlier. In particular, one can enumerate some problems that are of theoretical, as well as practical, interest, namely the following.

(1) What are the ways to extend the MG theory for shape-distributed particle systems?
(2) When one can use the simplest (steplike) approximation for the distribution function of ellipsoidal shapes?
(3) What are the properties that the distribution function in natural particle composites has to possess?
(4) Can the distribution function parameters be determined from experimental data?

Here we consider such complex composites in more detail and try to answer these questions. In particular, we consider the case of the uniform (steplike) distribution function, as well as the case of the generalized distribution functions. Particular emphasis has been placed on the case of small nonsphericity that is of practical interest. At the same time, our consideration uses quasi-static (long-wavelength) approximation, i.e. finite-size effects are neglected.

The structure of our paper is as follows. In section 2, the formalism used for description of shape-distributed particle systems is developed. First we give the well known results for systems of ellipsoids of a fixed shape. Then we present results obtained for ellipsoids distributed in shape with the simplest (steplike) distribution. The generalizations of the MG theory for nonspherical particles that are based on the spectral (Bergman) representation are to be further considered. Consideration of generalized distribution functions for ellipsoidal shapes completes section 2. In section 3, we present the results of numerical calculations performed for the effective dielectric response of a dilute system of silicon carbide and aluminum ellipsoids having complex permittivities and distributed in shape with different distributions. In section 4, the main results of our work are discussed. Some concluding remarks are presented in section 5.

2. System of ellipsoids

2.1. System of ellipsoids of a fixed shape

The effective dielectric response $\langle\varepsilon\rangle$ of a composite system may be defined as

$$\langle\varepsilon\rangle = \frac{\int_V D\,\mathrm{d}V}{\int_V E\,\mathrm{d}V} = \frac{f_m\langle D_m\rangle + f_i\langle D_i\rangle}{f_m\langle E_m\rangle + f_i\langle E_i\rangle}. \tag{1}$$

Here the subscript m refers to the matrix, the subscript i refers to the inclusions, $f_i(f_m)$ is the filling factor of the inclusions (matrix), $f_i + f_m = 1$ and $\langle D\rangle$ and $\langle E\rangle$ are the spatial averages of the displacement and electric field, respectively. It should be noted that such an approach implies that here only one-particle correlations are taken into account. We deal with a system of ellipsoidal particles (inclusions) whose (scalar) permittivity is ε_i. The inclusions are embedded in a matrix whose permittivity is ε_m. Then the expression (1) may be rewritten as

$$\langle\varepsilon\rangle = \frac{f_m\varepsilon_m\langle E_m\rangle + f_i\varepsilon_i\langle E_i\rangle}{f_m\langle E_m\rangle + f_i\langle E_i\rangle}. \tag{2}$$

Let us now consider a single ellipsoid made of a material with the permittivity ε_e embedded in a medium with the permittivity ε_s and subjected to an applied field E_0. Then the internal ellipsoid field E_e and the applied field may be related by the linear equation [42, 43]

$$\vec{E}_e = \overleftrightarrow{\lambda}\,\vec{E}_0 \tag{3}$$

where the diagonal components λ_j of the tensor $\overleftrightarrow{\lambda}$ are

$$\lambda_j = \frac{\varepsilon_s}{\varepsilon_s + L_j(\varepsilon_e - \varepsilon_s)} \tag{4}$$

and L_j is the jth depolarization factor of the ellipsoid. Note that a normalization condition, $\sum_j L_j = 1$, holds for any ellipsoid. Besides, if the ellipsoids are randomly distributed, one can take

$$\langle E_e\rangle = \langle\lambda\rangle E_0 \tag{5}$$

where $\langle\lambda\rangle$ is

$$\langle\lambda\rangle = \frac{1}{3}\sum_{j=1}^{3}\lambda_j. \tag{6}$$

It should be noted that we assume that the applied electric field $E_0 = \langle E\rangle$, i.e., it is constant and homogeneous and may result from averaging the microscopic field over a volume containing many inclusions. Besides, we suppose that the matrix (the space between inclusion particles) can be filled by (matrix) ellipsoids. To find the function $\langle\varepsilon\rangle$, we have to specify the relationships between $\langle E_m\rangle$, $\langle E_i\rangle$ and $\langle E\rangle$. The relationships $\varepsilon_i = \varepsilon_e$ and $\langle E_i\rangle = \langle E_e\rangle$ are obvious. It seems to be reasonable also to suggest that for diluted particle systems $\varepsilon_m = \varepsilon_s$ (Bruggeman's concept suggests $\varepsilon_m = \langle\varepsilon\rangle$). If so, then $\langle E_m\rangle$ does not depend on L_j. Such a choice yields the trivial relation $\langle E_m\rangle = \langle E\rangle$. This results in the equation for $\langle\varepsilon\rangle$

$$\langle\varepsilon\rangle = \frac{f_m\varepsilon_m + f_i\varepsilon_i\langle\lambda\rangle}{1 - f_i + \langle\lambda\rangle}. \tag{7}$$

Setting $L_i = \langle L\rangle = 1/3$ in equation (7), one obtains the classical MG equation for spherical inclusions.

2.2. Uniform distribution function

Let us now assume that the ellipsoids are distributed in shape. Besides, we operate on the premise that all orientations are equiprobable and there is no correlation between the volume of ellipsoids and their shape and orientation, as well as between the shape and orientation. In this case one can use the following generalized equation for the effective dielectric response [42]:

$$\langle\varepsilon\rangle = \varepsilon_m \frac{f_m + f_i \varepsilon_i \beta}{f_m + f_i \varepsilon_m \beta} \tag{8}$$

where

$$\beta = \frac{1}{3} \iint P(L_1, L_2) \sum_j [\varepsilon_m + L_j(\varepsilon_i - \varepsilon_m)]^{-1} \mathrm{d}L_1 \, \mathrm{d}L_2 \tag{9}$$

and $P(L_1, L_2)$ is the distribution function for the depolarization factors of ellipsoids. It is apparent that this function is to be normalized to unity:

$$\iint P(L_1, L_2) \, \mathrm{d}L_1 \, \mathrm{d}L_2 = 1. \tag{10}$$

We note that equation (9) can be rewritten as $\beta = \langle\alpha\rangle/(\varepsilon_i - \varepsilon_m)$ where

$$\langle\alpha\rangle = \frac{1}{3} \iint P(L_1, L_2) \left[\sum_{j=1}^{2} (\gamma + L_j)^{-1} + (\gamma + 1 - L_1 - L_2)^{-1} \right] \mathrm{d}L_1 \, \mathrm{d}L_2 \tag{11}$$

is the effective polarizability of an ensemble of non-interacting ellipsoids [39] and $\gamma = \varepsilon_m/(\varepsilon_i - \varepsilon_m)$. Then equation (8) takes the form

$$\langle\varepsilon\rangle = \varepsilon_m \left(1 - \frac{f_i \langle\alpha\rangle}{1 - f_i + f_i \gamma \langle\alpha\rangle} \right). \tag{12}$$

Thus, to find the effective dielectric response $\langle\varepsilon\rangle$, one suffices to calculate the effective polarizability $\langle\alpha\rangle$. It should be noted that if $f_i \ll 1$, then

$$\langle\varepsilon\rangle \cong \varepsilon_m (1 - f_i \langle\alpha\rangle). \tag{13}$$

Hence the behaviour of $\langle\varepsilon\rangle$ is qualitatively similar to that of the effective polarizability. A general scheme allowing calculating $\langle\varepsilon\rangle$ is given in the appendix.

Let us suppose that the shape of particles slightly differs from spherical. Then one can assume that the shape variance (nonsphericity) of the particles Δ is rather small. The simplest form of the distribution function in this case is the uniform (steplike) one [39, 41]

$$P_S(L_1, L_2) = \frac{2}{\Delta^2} \chi(L_1 - \Delta/3) \chi(L_2 - \Delta/3) \chi(1 - L_1 - L_2 - \Delta/3) \tag{14}$$

where χ is the Heaviside unit function. For this case the effective polarizability was obtained earlier [39]:

$$\langle\alpha\rangle_S = \frac{2}{\Delta^2} \left[(\gamma + 1/3 + 2\Delta/3) \ln \frac{\gamma + 1/3 + 2\Delta/3}{\gamma + 1/3 - \Delta/3} - \Delta \right]. \tag{15}$$

Taking into account that $\Delta \ll 1$, one may expand equation (15) into a Taylor series. This yields

$$\langle\alpha\rangle_S \cong \langle\alpha\rangle_0 \left(1 + \frac{1}{18} \langle\alpha\rangle_0^2 \Delta^2 \right). \tag{16}$$

By substituting equation (15) into equation (12) one obtains a generalized MG equation for $\langle\varepsilon\rangle_S$. On the other hand, substitution of equation (16) into equation (12) gives an approximate equation

$$\langle\varepsilon\rangle_S \cong \langle\varepsilon\rangle_0 + \frac{1}{18} \left[\gamma (1 - \langle\varepsilon\rangle_0) \left(1 - \frac{\langle\varepsilon\rangle_0}{\varepsilon_m} \right) + \langle\varepsilon\rangle_0 - \varepsilon_m \right] \langle\alpha\rangle_0 \Delta^2 \tag{17}$$

where $\langle\varepsilon\rangle_0$ satisfies the classical MG equation (at $\Delta = 0$).

It is of interest to find a condition when Im $\langle\varepsilon\rangle$ peaks. It is well known that for the classical MG equation this condition is Re $\langle\alpha\rangle_0 = 3/f_i$. If $\Delta \neq 0$, then one can suppose that such a condition is of the form

$$\mathrm{Re}\,\langle\alpha\rangle_0 \cong \frac{3}{f_i}(1+\delta) \tag{18}$$

where $\delta \ll 1$. Substituting equation (15) into equation (8) and setting the denominator equal to zero, one can estimate the value of δ:

$$\delta \cong \frac{f_m}{f_i}\frac{3\Delta^2}{6\Delta^2 - 9\Delta^2/f_i - 6f_i}. \tag{19}$$

It is easy to verify that $\delta < 0$ for any f and Δ. Equation (18) can be rewritten in the form

$$\mathrm{Re}\,\varepsilon_i \cong \varepsilon_m \frac{1 + 2(1/f_i + \delta/3)}{1 - (1/f_i + \delta/3)}. \tag{20}$$

One can see that the distribution in shape shifts the peak of Im $\langle\varepsilon\rangle$ to lower frequencies. This is an evident consequence of equation (20) when $\delta < 0$. A similar phenomenon takes place also for the effective absorption cross section [39].

Questions arise concerning the effect of the distribution function form on the effective dielectric response. Earlier Hansen and Travis showed [44] that scattering properties of size-distributed spherical particles depend primarily on the effective variance of the distribution, while its actual form has a minor influence. Later Mishchenko and Travis [45] generalized this result to randomly oriented nonspherical particles.

2.3. *Generalizations based on the spectral representation*

Generally speaking, there is some arbitrariness when choosing the distribution function $P(L_1, L_2)$. Indeed, the spectral representation [46–48] enables one to write for $\langle\varepsilon\rangle$ of any composite

$$\langle\varepsilon\rangle = \varepsilon_m\left[1 + f_i\int_0^1 G(x)(\gamma + x)^{-1}\mathrm{d}x\right] \tag{21}$$

where $G(x)$ is the spectral density function that specifies the composite topology. Comparing equations (13), (11) and (21), one obtains the following equation linking the function $P(L_1, L_2)$ with $G(x)$:

$$\frac{1}{3}\int_0^1\int_0^{1-L_1} P(L_1, L_2)\sum_{j=1}^{3}(\gamma + L_j)^{-1}\mathrm{d}L_1\,\mathrm{d}L_2 = \int_0^1 \frac{G(x)}{\gamma + x}\mathrm{d}x. \tag{22}$$

This means that the representation (13) is as general as the representation (21). In particular, it is suitable not only for the composites with matrix topology where inclusions (particles) are embedded in a continuous matrix (host), but for any composites as well.

The spectral density function $G(x)$ corresponding to the classical MG equation is [47]

$$G(x) = \delta(x - f_m/3) \tag{23}$$

where δ is the Dirac delta-function. One can try to extend the MG theory by generalizing the above equation. For example, one can use the approach proposed earlier [49] for superlattices. Similar to this, in the case of slightly nonspherical particles the spectral density function can be specified in the form of the step

$$G(x) = \begin{cases} \Delta^{-1} & \text{if } f_m(1-\Delta)/3 \leqslant x \leqslant f_m(1-\Delta)/3 + \Delta \\ 0 & \text{if not.} \end{cases} \tag{24}$$

Here the nonsphericity parameter $0 \leqslant \Delta \leqslant 1$ and the left limit of the step $f_m(1-\Delta)/3 \to 0$, while the right limit $f_m(1-\Delta)/3 + \Delta \to 1$ as $\Delta \to 1$. Substituting equation (24) into equation (21), one gets

$$\langle\varepsilon\rangle = \varepsilon_m\left[1 + \frac{f_i}{\Delta}\ln\frac{\gamma + f_i(1-\Delta)/3 + \Delta}{\gamma + f_i(1-\Delta)/3}\right]$$
$$= \varepsilon_m\left\{1 + \frac{f_i}{\Delta}\ln\left[1 + \Delta\frac{\varepsilon_i - \varepsilon_m}{\varepsilon_m + f_i(\varepsilon_i - \varepsilon_m)(1-\Delta)/3}\right]\right\}. \tag{25}$$

If $\Delta \ll 1$, then

$$\langle\varepsilon\rangle \cong \varepsilon_m\left[1 + \frac{f_i}{\gamma + f_m/3} - \Delta f_i\frac{1+2f_i}{6(\gamma + f_m/3)^2}\right] = \langle\varepsilon\rangle_0 - \frac{1}{6}\Delta f_i\varepsilon_m\frac{1+2f_i}{(\gamma + f_m)^2}. \tag{26}$$

What is the generalization specified by equation (25)? Close inspection of equation (21) shows that the second factor in the integrand is (with some constant factor) nothing but the dipole polarizability of a spheroid whose depolarization factor is x. In other words, the representation (21) means that a diagonal component of the effective dielectric response of any two-phase composite is the same as for a system of equally oriented non-interacting spheroids (evidently, the electric field orientation along the spheroid rotation axis corresponds to the above component). Thus we have derived the generalization of the classical MG equation for spheroids distributed in shape and aligned along an axis. In a sense, equation (25) gives only a diagonal component of the effective dielectric tensor of such an ensemble. If spheroids are randomly oriented, then the integrand in equation (21) has to contain the spheroid polarizability averaged over three main axes. This yields

$$\langle\varepsilon\rangle = \varepsilon_m\left\{1 + \frac{f_i}{\Delta}\left[\ln\left(1 + \frac{\Delta}{\gamma + f_m(1-\Delta)/3}\right) - 4\ln\left(1 + \frac{\Delta}{f_m(1-\Delta)/3 - 2\gamma - 1}\right)\right]\right\}. \tag{27}$$

If $\Delta \ll 1$, then

$$\langle\varepsilon\rangle \cong \varepsilon_m\left\{1 + \frac{f_i}{\gamma + f_m/3} + 4\frac{f_i}{2\gamma + 1 - f_m/3} - \frac{1}{6}\Delta f_i\left[\frac{1}{(\gamma + f_m/3)^2} - \frac{4}{(2\gamma + 1 - f_m/3)^2}\right]\right\}$$
$$= \langle\varepsilon\rangle_0 - \frac{1}{6}\Delta f_i\varepsilon_m\left[\frac{1}{(\gamma + 1/3)^2} - \frac{4}{(2\gamma + 1 - f_m/3)^2}\right]. \tag{28}$$

How to specify the spectral density function for a system of shape-distributed ellipsoids? To answer this question, one can combine equations (12) and (21) and then solve the equation for $G(x)$ (see, e.g., [47]). If, for example, one uses for the effective polarizability the approximation (16), then such an approach gives for the function $G(x)$

$$G(x) = \tfrac{1}{3}[\delta(x - x_0) + \delta(x - x_1) + \delta(x - x_2)] \tag{29}$$

where $x_{0,1,2}$ are the solutions of the equation $1 + \Delta^2(1/3 - x)^{-2}/18 = (1/3x - 1)f_m/f_i$. It is easy to see that if $\Delta \to 0$ then $x_0 = x_1 = x_2 \cong f_m/3$. It should be noted that this result (equation (29)) is in agreement with that obtained earlier [50] for the two-parameter spectral density function.

Above we have noted that the spectral density function $G(x)$ specifies the distribution of the depolarization factors of an ensemble of spheroids with the same effective dielectric response as that of the real particle composite. It is not fully clear how to construct the corresponding spectral density. At the same time, the distribution function $P(L_1, L_2)$ can be considered as the distribution in shape of *real* particles forming the composite. Because of this one might expect to construct a generalized distribution function for *real* particle composites from some general considerations.

2.4. Generalized distribution function

Here we restrict our analysis to the case of composites with matrix topology, namely, the composites consisting of small ellipsoids distributed in shape. Unfortunately, even in this case it seems to be highly conjectural to construct a microscopic theory for the distribution function $P(L_1, L_2)$. As is known [45], the data on the distribution of particle shapes in natural ensembles are very limited. However, it is reasonable to suggest that the function $P(L_1, L_2)$ has to possess some properties for most systems that are realized in practice. On such a basis phenomenological models for this function can be constructed. So we assume that the distribution function $P(L_1, L_2)$ has to have the following properties that appear reasonable for many natural particle ensembles: (i) $P(L_1, L_2) = P(L_2, L_1)$; (ii) if $L_1(L_2) \to 0$ or $L_1(L_2) \to 1$, then $P(L_1, L_2) \to 0$, i.e., both strongly prolate (needle-like) and strongly oblate (disclike) particles are highly improbable; (iii) $P(L_1, L_2)$ peaks at $L_1 = L_2 = 1/3$, i.e., the spherical shape of the particles is most probable; (iv) the function $P(L_1, L_2)$ is centred at $L_1 = L_2 = 1/3$, namely, for any parameter $0 < \sigma < 1/3$

$$\int_{1/3-\sigma}^{1/3} \mathrm{d}L_1 \int_{1/3-\sigma}^{L_1} P(L_1, L_2)\, \mathrm{d}L_2 = \int_{1/3}^{1/3+2\sigma} \mathrm{d}L_1 \int_{1/3-\sigma}^{(1-L_1)/2} P(L_1, L_2)\, \mathrm{d}L_2 \tag{30}$$

(here integration is performed over right triangles). Equation (30) expresses a specific symmetry of the system under consideration; it means that deviations of particle shape from spherical to flat and extended ellipsoidal are equiprobable [39, 42].

Let us now consider a two-parameter distribution function of a rather general form:

$$\begin{aligned} P(L_1, L_2) = P_1(L_1, L_2) = C_1 &\left\{ \frac{\pi}{2} + \operatorname{arctg}\left[a_1 \frac{-L_1 + 1/3 - \Delta_1/3}{L_1(L_1 - 1)} \right] \right\} \\ &\times \left\{ \frac{\pi}{2} + \operatorname{arctg}\left[a_1 \frac{-L_2 + 1/3 - \Delta_1/3}{L_2(L_2 - 1)} \right] \right\} \\ &\times \left\{ \frac{\pi}{2} + \operatorname{arctg}\left[a_1 \frac{-L_1 - L_2 + 2/3 + \Delta_1/3}{(1 - L_1 - L_2)(L_1 + L_2)} \right] \right\} \end{aligned} \tag{31}$$

where the constant C_1 can be determined from the normalization condition (10). Here the passages to limits take place, namely, if $a_1 \to \infty$, then $P(L_1, L_2) \to P_S(L_1, L_2)$ and if $a_1 \to 0$, then $P(L_1, L_2) \to P_S(\Delta = 1) = 2$. This two-parameter function appears as a natural generalization of the one-parameter distribution function (14) and we will use it in what follows.

One can consider also another distribution function:

$$\begin{aligned} P(L_1, L_2) = P_2(L_1, L_2) = C_2 &\left\{ 1 + \exp\left[-a_2\left(L_1 + \frac{\Delta_2}{3} - \frac{1}{3} \right) \right] \right\}^{-1} \\ &\times \left\{ 1 + \exp\left[-a_2\left(L_2 + \frac{\Delta_2}{3} - \frac{1}{3} \right) \right] \right\}^{-1} \\ &\times \left\{ 1 + \exp\left[-a_2\left(-L_1 - L_2 + \frac{\Delta_2}{3} + \frac{2}{3} \right) \right] \right\}^{-1}. \end{aligned} \tag{32}$$

It is easy to check that the function $P_2(L_1, L_2)$ obeys conditions (i) and (iii) exactly, as well as the above limits, if one substitutes a_2 for a_1. The conditions (ii) and (iv) are satisfied approximately. We note also that the variances of the distributions (31) and (32) (cross-sectional area on half of the height) are approximately $\Delta_1^2/2$ and $\Delta_2^2/2$, respectively; this claim breaks down at small a_1 (a_2) values only. The general view of the distribution functions $P_1(L_1, L_2)$ and $P_2(L_1, L_2)$ is shown in figure 1.

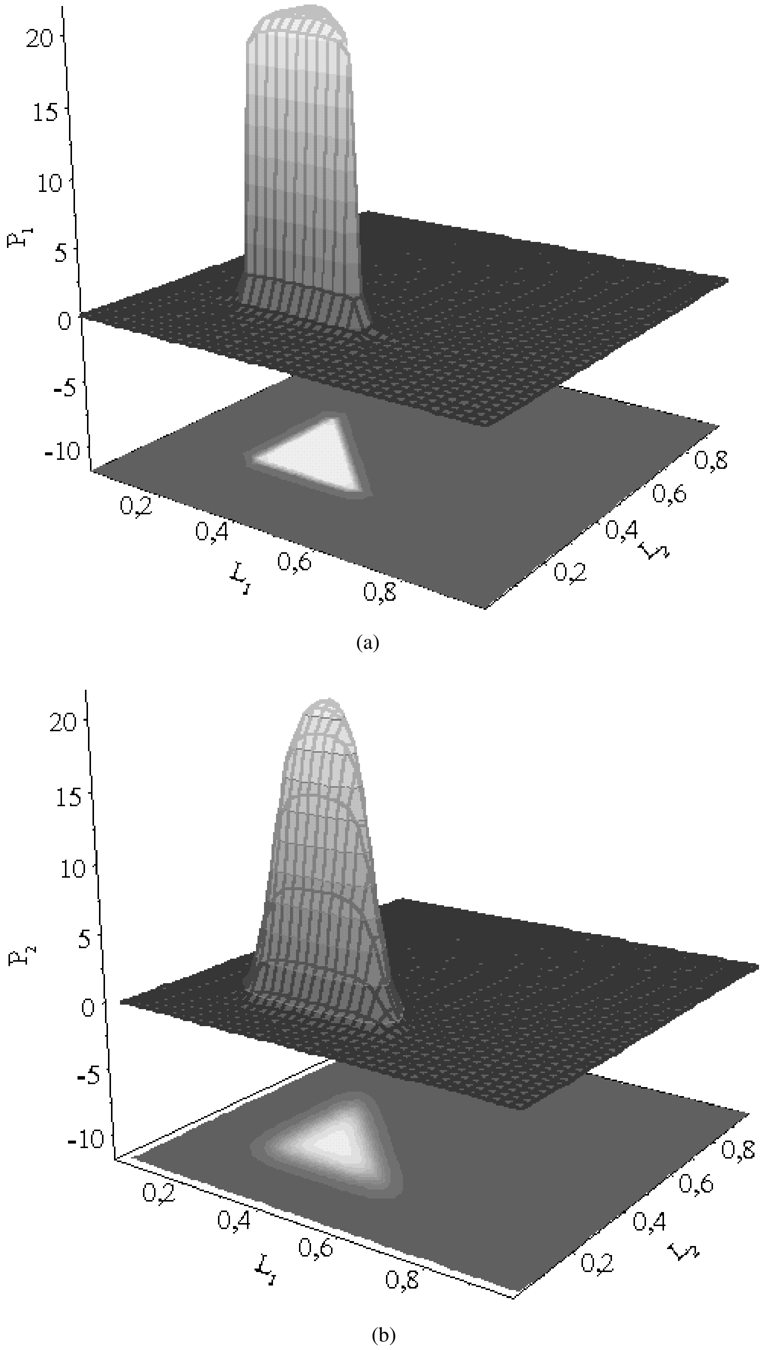

Figure 1. The general view of the distribution functions (31) (a) and (32) (b) at the values of parameters $\Delta_1 = \Delta_2 = 0.3$ and $a_1 = a_2 = 50$. The projections of the distribution functions on the (L_1, L_2) plane are shown at the bottom.

Unfortunately, the use of the distributions (31) and (32) does not enable us to perform the analytical integration in equation (11). So our further analysis will be numerical. Substituting equation (31) (or equation (32)) into the general equation for polarizability (11), one can calculate an averaged polarizability and then (from equation (12)) the effective dielectric response of the system of ellipsoids distributed in shape with the generalized distributions.

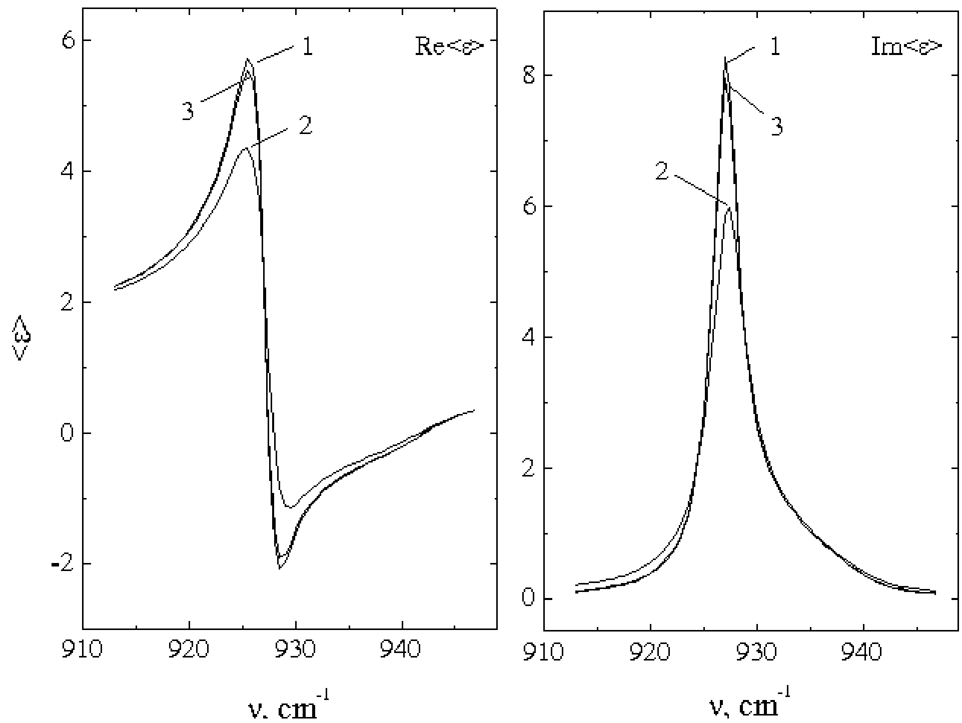

Figure 2. The effective dielectric response of a system of semiconductor ellipsoids. Curve 1 corresponds to the steplike distribution; curves 2 ($a_1 = 10^2$) and 3 ($a_1 = 10^3$) correspond to equation (31) with $\Delta_1 = 0.1$.

3. Some numerical examples

To gain greater insight into how the shape distribution influences the effective dielectric response, we consider particles of two types having essentially different bulk dielectric properties: cubical SiC and Al. In [39] the behaviour of $\langle\alpha\rangle$ as a function of parameter Δ was considered. Earlier we noted that the behaviour of the function $\langle\varepsilon\rangle(\Delta)$ is qualitatively similar to that of the function $\langle\alpha\rangle(\Delta)$ (see equation (13)). Hence, here we focus our attention on the effect of the parameter a_1 (a_2) (specifying the distribution function spreading) on $\langle\varepsilon\rangle$.

Cubic silicon carbide (β-SiC) is a wide-gap semiconductor that can be considered in the IR as a typical insulator whose permittivity is well described by the one-oscillator Lorentz model. The effective dielectric response function for silicon carbide particles calculated with the use of distributions (31) and (32) at different values of parameter a_1 (a_2) is shown in figures 2 and 3, respectively. We should like to note the following peculiarities of this behaviour:

(i) the frequency position of the Im $\langle\varepsilon\rangle(\nu)$ peak practically does not depend on a_1 (a_2);
(ii) the value of Im $\langle\varepsilon\rangle(\nu)$ at its peak depends on the parameters a_1 and a_2 only slightly (however, at small a_1 (a_2) this dependence becomes more pronounced);

(iii) when a_1 (a_2) becomes small, then an asymmetry of the $\in \langle\varepsilon\rangle(\nu)$ curve occurs.

Light absorption here is caused by surface phonon modes which manifest themselves in the relatively narrow band (*reststrahlen* range) lying in the IR.

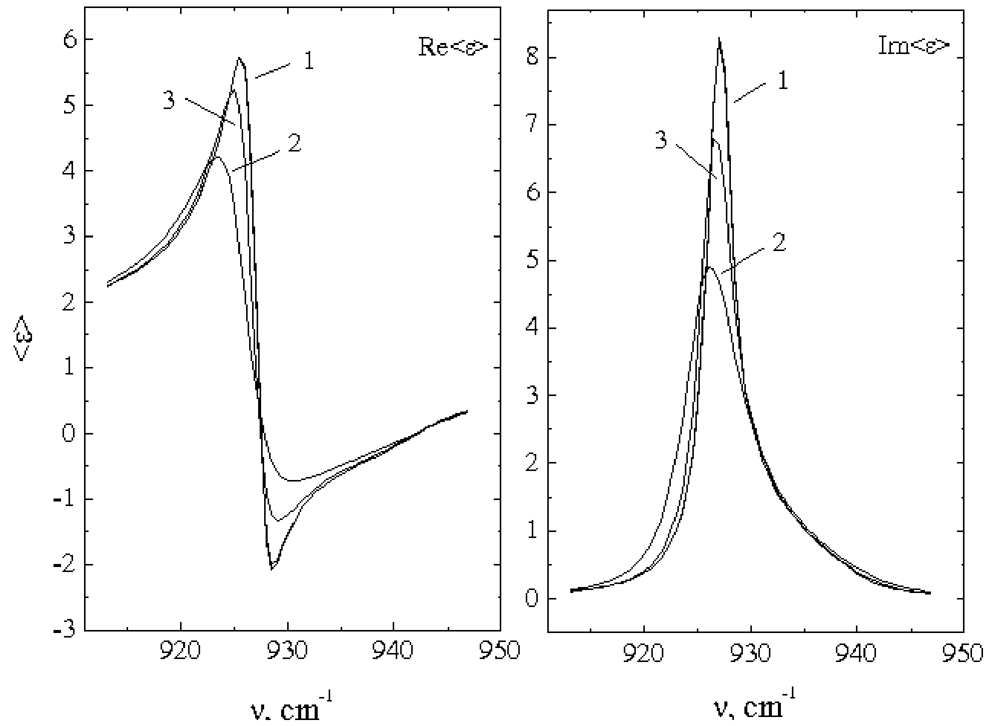

Figure 3. The effective dielectric response of a system of semiconductor ellipsoids. Curve 1 corresponds to the steplike distribution; curves 2 ($a_2 = 10^2$) and 3 ($a_2 = 5 \times 10^3$) correspond to equation (32) with $\Delta_2 = 0.1$.

Similar peculiarities take place also for metal particles. For example, in figures 4 and 5 the effective dielectric response function for shape-distributed aluminum particles is presented. One can note, however, that for metal particles, in contrast to dielectric ones, there is a wide band of heavy absorption caused by surface plasmons which can excite below the plasma frequency. The reason is that in metals there is a wide frequency band where their permittivity is negative (this condition is necessary to satisfy equation (18)); the band can cover a part of the UV, visible and IR.

It is of interest to analyse the dependence $F = \text{Im}\,\langle\varepsilon\rangle(\nu_{max})/\text{Im}\,\langle\varepsilon\rangle_S(\nu_{max})$ on a_1 (a_2) where ν_{max} is the frequency position of the Im $\langle\varepsilon\rangle$ peak. Examples of such dependences calculated for silicon carbide and aluminum particles are given in figure 6. We see that with the use of the distribution (32) the dependences rapidly approach unity; with the use of the distribution (31) they approach unity more slowly. Obviously, the reason is that the distribution (32) converges to $P_S(L_1, L_2)$ with a_2 faster than the distribution (31) converges to $P_S(L_1, L_2)$ with a_1.

4. Discussion

In the actual particle systems absorption spectra differ significantly from theoretical ones calculated for balls [42, 51]. There are two main reasons for this. The first of these is the particle

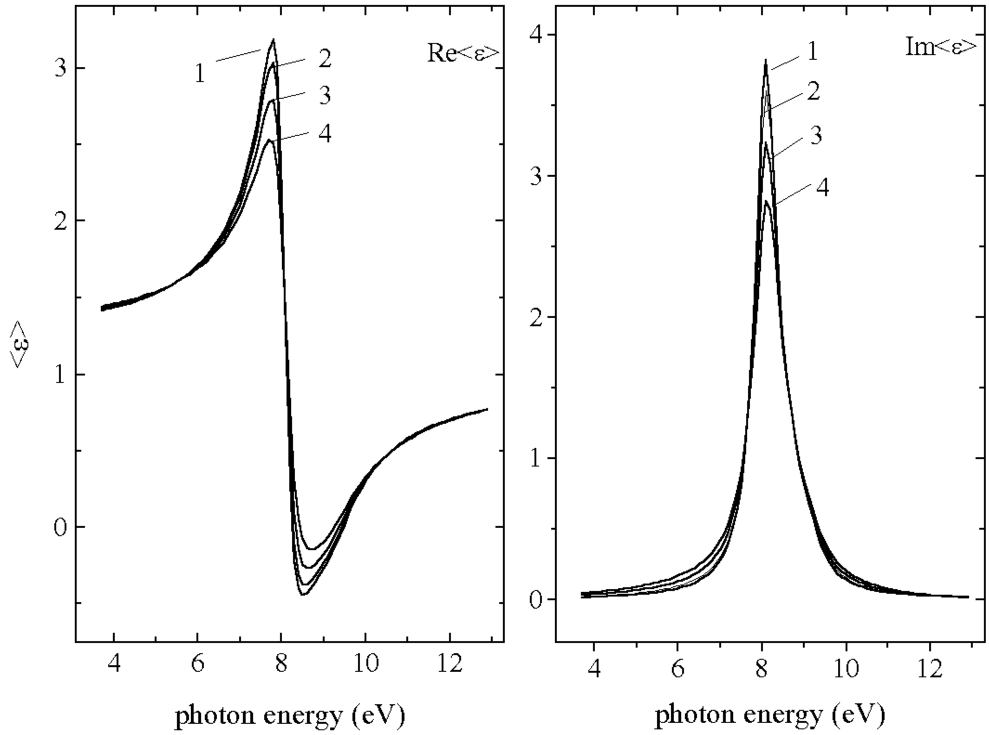

Figure 4. The effective dielectric response of a system of metal ellipsoids. Curve 1 corresponds to the steplike distribution; curves 2 ($a_1 = 10^3$), 3 ($a_1 = 10^2$) and 4 ($a_1 = 50$) correspond to equation (31) with $\Delta_1 = 0.1$.

aggregation. The second one is the nonsphericity and shape distribution. Both phenomena lead to a lowering of the absorption at its peak and a broadening of the absorption band. In this work the second phenomenon has been the subject of our studies. The problem lies in the fact that the shape distribution function is usually unknown. How much is this problem of real concern? When is the use of the steplike distribution justified?

To answer these questions, here we discuss in greater detail the effect of the shape distribution form on the function $\langle\varepsilon\rangle(\nu)$. To do this, let us now turn our attention to figure 6. We see that if $a_1 = 200$, then $F = 0.83$ for SiC particles and $F = 0.92$ for Al particles. At the same time, if $a_2 = 200$, then $F = 0.84$ for SiC particles and $F = 0.93$ for Al particles. If $a_1 = a_2 = 1000$, then the distributions (31) and (32) are markedly nearer to the distribution (14) than in the above example. In this case $F = 0.96$ for SiC particles and $F = 0.975$ for Al particles (with the use of the distribution (31)) and $F = 0.99$ (with the use of the distribution (32)). Thus, at $a_1 = a_2 = 1000$ both distributions, (31) and (32), give a result close to that obtained with the use of the steplike distribution (14) (the deviation is less than 5%).

Why does the $F(a_{1(2)})$ dependence for Al particles converge to unity faster than that for SiC particles? The reason seems to be that the $\mathrm{Im}\,\varepsilon_i$ value for SiC is less than that for Al in the actual frequency region. Indeed, for both cases $\mathrm{Re}\,\varepsilon_i(\nu_{max}) \cong -2$ (this is evident from equation (20) when $f_i \ll 1$). At the same time our estimation for $\mathrm{Im}\,\varepsilon_i(\nu_{max})$ gives 0.11 for SiC and 2.06 for Al. To check our assumption, we have calculated the $F(a_1)$ dependence for SiC particles using the lower (curve 5) and higher (curve 6) TO-phonon damping constant. Clearly

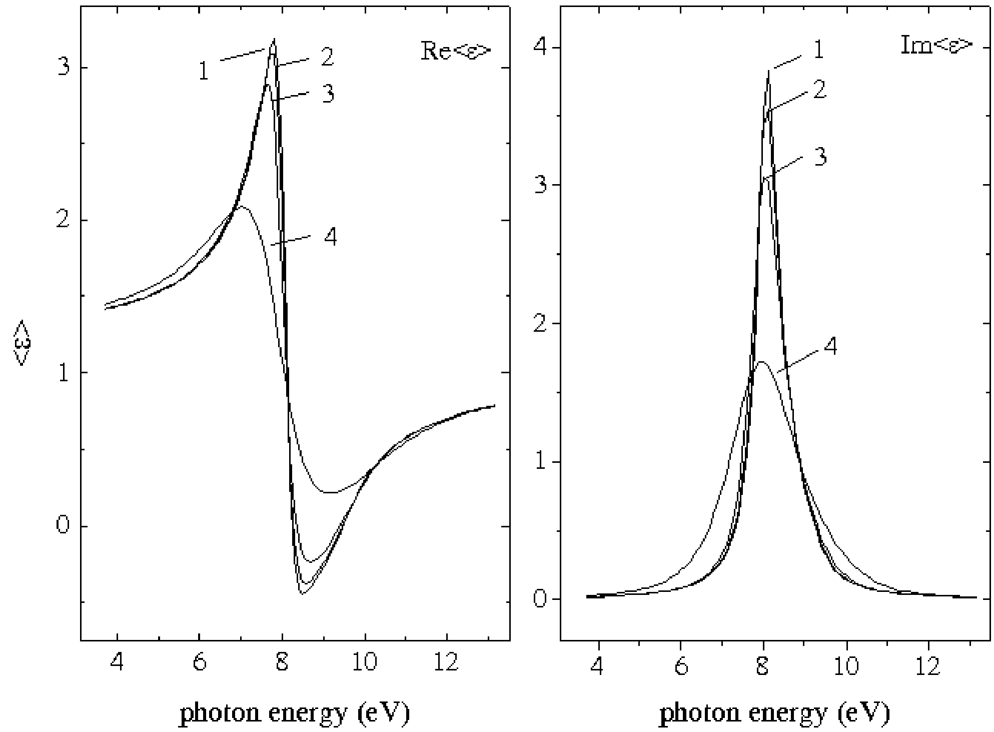

Figure 5. The effective dielectric response of a system of metal ellipsoids. Curve 1 corresponds to the steplike distribution; curves 2 ($a_1 = 10^3$), 3 ($a_1 = 10^2$) and 4 ($a_1 = 50$) correspond to equation (32) with $\Delta_1 = 0.1$.

such a change of TO-phonon damping constant leads to a respective decrease or increase of the imaginary part of the particle permittivity. We see that the function $F(a_1)$ rises with $\mathrm{Im}\,\varepsilon_i$. One can show that the same takes place for the function $F(a_2)$ too.

One can assume that a general statement holds, namely

$$\frac{\partial F}{\partial \mathrm{Im}\,\varepsilon_i} > 0 \tag{33}$$

where $F = \mathrm{Im} \iint P(L_1, L_2)\Phi_{max}(L_1, L_2)\,\mathrm{d}L_1\,\mathrm{d}L_2/\mathrm{Im} \iint P_S(L_1, L_2)\Phi_{max}(L_1, L_2)\,\mathrm{d}L_1\,\mathrm{d}L_2$, $\Phi_{max} = \sum_{i=1}^{3}[\gamma(\nu_{max}) + L_i]^{-1}$ and $P(L_1, L_2)$ is a one-mode distribution function obeying the above conditions (i)–(iv). Rigorous proof of this statement is not trivial and we hope to give it elsewhere.

Let us now consider the accuracy of the steplike distribution in some frequency range, i.e., the function

$$\delta^* = \frac{1}{\nu_2 - \nu_1}\int_{\nu_1}^{\nu_2}\left|\frac{\langle\varepsilon\rangle(\nu) - \langle\varepsilon\rangle_S(\nu)}{\langle\varepsilon\rangle_S(\nu)}\right|\mathrm{d}\nu. \tag{34}$$

For this purpose we first calculate the function $\langle\varepsilon\rangle(\nu)$ using the distributions (31) and (32) with fixed values of $\Delta_1 = \Delta_2 = 0.1$. Then we fit the obtained functions $\langle\varepsilon\rangle_1(\nu)$ and $\langle\varepsilon\rangle_2(\nu)$ by the function $\langle\varepsilon\rangle_S(\nu)$ varying the parameter Δ. We name the value of Δ obtained as a result of such fitting procedure 'optimal' and denote it Δ_{opt}. Our calculations for the functions $\delta^*(a_{1(2)})$ and $\Delta_{opt}(a_{1(2)})$ for SiC particles in the frequency range 750–1000 cm^{-1} (that includes the reststrahlen range) are presented in figures 7 and 8, respectively.

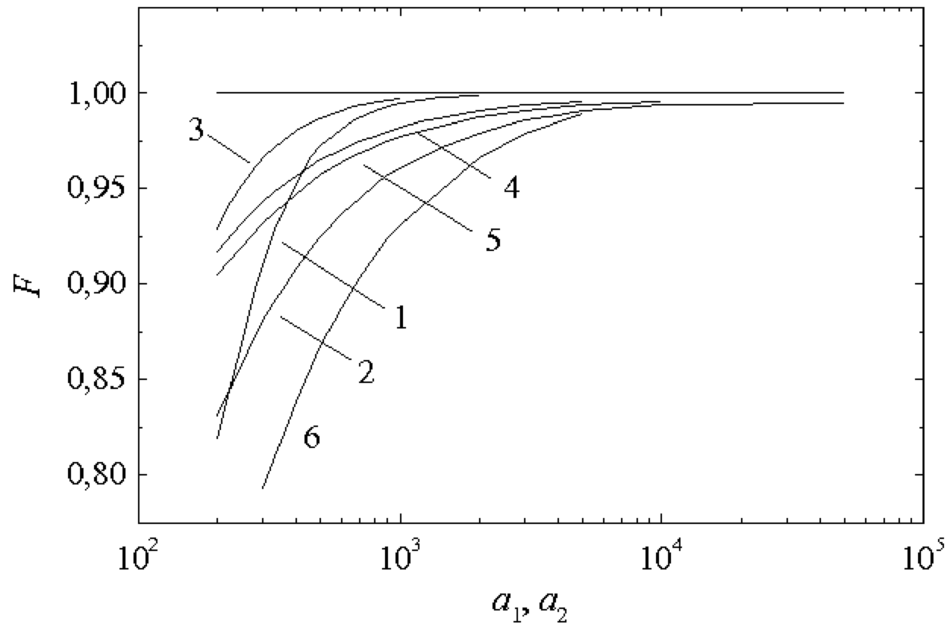

Figure 6. Dependence of the function $F = \text{Im}\,\langle\varepsilon\rangle(\nu_{max})/\text{Im}\,\langle\varepsilon\rangle_S(\nu_{max})$ calculated according to equations (31) and (32) on parameters a_1 and a_2, respectively. The effective dielectric response is calculated with distribution as equation (32) for semiconductor inclusions (curve 1); with distribution as equation (31) for semiconductor inclusions (curve 2); with distribution as equation (32) for metal inclusions (curve 3); with distribution as equation (31) for metal inclusions (curve 4). Curve 5—the same as curve 2 but with imaginary part of inclusion permittivity ten times as much; curve 6—the same as curve 2 but with imaginary part of inclusion permittivity one-tenth as much.

We see that the steplike distribution (14) may not always be successfully used to describe the function $\langle\varepsilon\rangle$ for actual particle systems. In our example an average error can reach 10%. At the same time, when $a_{1(2)}$ is sufficiently large (over 10^2), then the average error is less than 2%. Besides, we see that an increase of Im ε_i improves the applicability of the steplike distributions.

In practice the use of the distributions (31) and (32) seems to be more valid. Can one determine parameters of these distributions from experimental data, for example, from transmission spectra? To put it differently, can the inverse spectroscopic problem be solved? If the particle system possesses the needed properties allowing one to use the generalized MG equation (12), then our answer is 'yes'. Such a conclusion follows from the fact that the parameters $a_{1(2)}$ and $\Delta_{1(2)}$ affect differently the form of the Im $\langle\varepsilon\rangle(\nu)$ dependence. Indeed, the nonsphericity parameter $\Delta_{1(2)}$ essentially influences both the peak frequency position and the Im $\langle\varepsilon\rangle(\nu_{max})$ value [39]. At the same time the spreading parameter $a_{1(2)}$ affects practically the Im $\langle\varepsilon\rangle(\nu_{max})$ value only. We predict also that the accuracy of determination of the parameters $a_{1(2)}$ cannot be high when the shape-distribution function is close to the steplike one, i.e., when $a_{1(2)}$ is large.

One can also consider a more general inverse problem, namely, extracting the function $P(L_1, L_2)$ from experimental measurements. It needs to be noted that similar problems fall in the category of ill posed (or incorrect) ones: their solutions are unstable to weak changes of input data. Such problems and methods of their solution are considered, in particular, in [52, 53]. A closely related example is presented in [54], where a method for extracting the spectral density function from experimental data is developed.

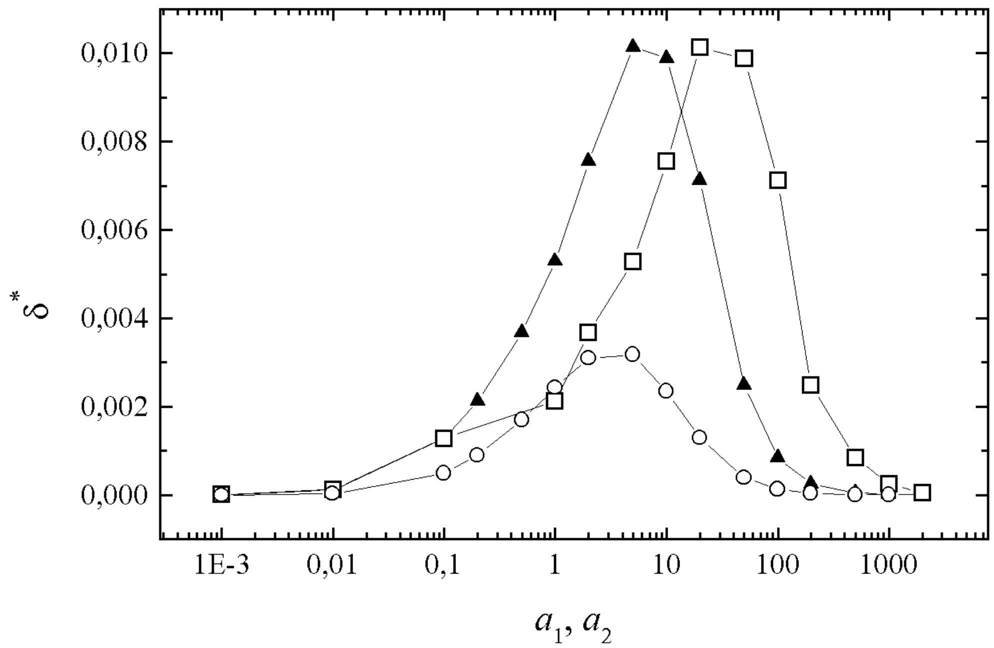

Figure 7. Function δ^* for SiC particles in the frequency range 750–1000 cm^{-1} calculated according to equation (34) with the use of equation (31) (triangles), equation (32) (squares) and with the use of equation (31) and an enlarged imaginary part of particle permittivity (circles).

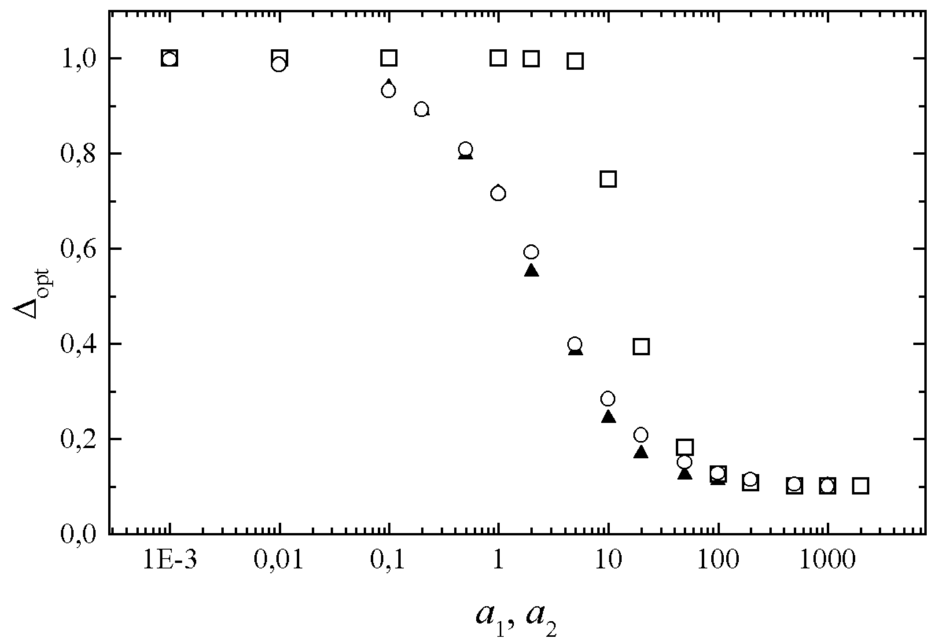

Figure 8. Function Δ_{opt} for SiC particles in the frequency range 750–1000 cm^{-1} calculated according to equation (34) with the use of equation (31) (triangles), equation (32) (squares) and with the use of equation (31) and an enlarged imaginary part of particle permittivity (circles).

5. Conclusion

We have considered the generalized Maxwell Garnett equations for small ellipsoids and spheroids taking into account their shape distribution. The generalizations are based on the spectral representation, as well as on the distribution function for depolarization factors of ellipsoids. Three possible distribution functions are examined. The simplest distribution function is the steplike one. It is a one-parameter function, and the only parameter describes the measure of nonsphericity. Two other distributions are two-parameter ones. The second parameter describes the spreading of the distribution function. Considering an example of SiC particles in the *reststrahlen* range, we conclude that the steplike distribution is not always applicable in practice. However, its applicability is improved when the imaginary part of the particle permittivity rises.

In conclusion, it should be noted that one of conditions that restricts essentially the use of our approach in practice is the necessity to take into account the particle aggregation. This problem is rather complex and calls for a special consideration. Some approaches to its solution are outlined, in particular, in [55–57]. One can assume that the discrimination between the effects caused by the particle shapes and those due to a strong interaction between particles could be extremely difficult. However, we think that even in this case the problem of the effect of the shapes of both particles forming the aggregate (cluster) and the particle clusters on the effective polarizability (dielectric response) also deserves attention.

Appendix

Let us assume that $P(L_1, L_2)$ is a narrow bell-shaped distribution function peaking at L_1, $L_2 = 1/3$ and that when its variance $\Delta \to 0$, the function $P(L_1, L_2)$ becomes infinite. This means that in this case the distribution function is defined on the P-null sets. Such function can be expanded in a series as an orthonormal set of δ-function and its derivatives [58]

$$P(L_1, L_2) = \sum_{n,m=0}^{\infty} C_{nm}\delta^{(n)}(L_1 - 1/3)\delta^{(m)}(L_2 - 1/3). \tag{A1}$$

It must be emphasized that, of course, equation (A1) should not be taken too literally. It is valid for integral relations only. From the normalization condition (10) it follows that

$$\sum_{n,m=0}^{\infty} C_{nm} \int_0^1 \mathrm{d}L_1\delta^{(n)}(L_1 - 1/3) \int_0^1 \mathrm{d}L_2\delta^{(m)}(L_2 - 1/3) = 1. \tag{A2}$$

The last equation enables us to find the coefficient C_{00}. Indeed, using the fact that the relations hold for the δ-function [59]

$$\int_a^b \mathrm{d}\xi f(\xi)\delta^{(n)}(\xi - x) = \begin{cases} 0, x \notin [a, b] \\ \frac{1}{2}(-1)^n f^{(n)}(x+0), x = a \\ \frac{1}{2}(-1)^n f^{(n)}(x-0), x = b \\ \frac{1}{2}(-1)^n [f^{(n)}(x+0) + f^{(n)}(x-0)], x \in [a, b] \end{cases} \tag{A3}$$

and taking here $f(\xi) = 1$, one obtains $C_{00} = 1$. Thus, the distribution function can be represented as

$$P(L_1, L_2) = \delta(L_1 - 1/3)\delta(L_2 - 1/3) + \sum_{n,m=1}^{\infty} C_{nm}\delta^{(n)}(L_1 - 1/3)\delta^{(m)}(L_2 - 1/3). \tag{A4}$$

Substituting this expression into equation (11), one gets

$$\langle\alpha\rangle = \langle\alpha\rangle_0 + \frac{1}{3}\int_0^1 \mathrm{d}L_1 \int_0^{-L_1+1} \mathrm{d}L_2 \sum_{n,m=1}^{\infty} C_{nm}\delta^{(n)}(L_1 - 1/3)\delta^{(m)}(L_2 - 1/3)\Phi(L_1, L_2) \tag{A5}$$

with

$$\langle\alpha\rangle_0 = (\gamma + 1/3)^{-1} \tag{A6}$$

that specifies the polarizability of spherical inclusions and

$$\Phi(L_1, L_2) = \sum_{j=1}^{2}(\gamma + L_j)^{-1} + (\gamma + 1 - L_1 - L_2)^{-1}. \tag{A7}$$

The infinite sum

$$\tilde{P}(L_1, L_2) = \sum_{n,m=1}^{\infty} C_{nm}\delta^{(n)}(L_1 - 1/3)\delta^{(m)}(L_2 - 1/3) \tag{A8}$$

after its integration with $\Phi(L_1, L_2)$ yields

$$\begin{aligned} I &= \frac{1}{3}\int_0^1 \mathrm{d}L_1 \int_0^{-L_1+1} \mathrm{d}L_2 \tilde{P}(L_1, L_2)\Phi(L_1, L_2) \\ &\cong \sum_{m,n=1}^{\infty} (-1)^{m+n} C_{mn} \frac{\partial^{m+n}}{\partial^m L_1 \partial^n L_2}\Phi(L_1, L_2)|_{L_1,L_2=1/3}. \end{aligned} \tag{A9}$$

Clearly, the coefficients C_{mn} have to depend on Δ and consequently may be represented as a power series in Δ:

$$C_{mn}(\Delta) = K_{mn}^{(0)} + K_{mn}^{(1)}\Delta + K_{mn}^{(2)}\Delta^2 + \ldots. \tag{A10}$$

It is easy to check that $K_{mn}^{(0)} = 0$. Indeed, the passage has to take place

$$\lim_{\Delta\to 0} P(L_1, L_2) = \delta(L_1 - 1/3)\delta(L_2 - 1/3) \tag{A11}$$

or

$$\begin{aligned} \lim_{\Delta\to 0}\sum_{m,n=1}^{\infty}\{&K_{mn}^{(0)}\delta^{(m)}(L_1 - 1/3)\delta^{(n)}(L_2 - 1/3) + \Delta K_{mn}^{(1)}\delta^{(m)}(L_1 - 1/3)\delta^{(n)}(L_2 - 1/3) \\ &+\Delta^2 K_{mn}^{(2)}\delta^{(m)}(L_1 - 1/3)\delta^{(n)}(L_2 - 1/3) + \ldots\} = 0. \end{aligned} \tag{A12}$$

It follows immediately that $K_{mn}^{(0)}$ has to equal zero. Hence we can write down

$$I = I_1\Delta + I_2\Delta^2 + \ldots \tag{A13}$$

where

$$I_l = \frac{1}{3}\sum_{m,n=1}^{\infty}(-1)^{m+n}K_{mn}^{(l)}\frac{\partial^{m+n}}{\partial^m L_1\partial^n L_2}\Phi(L_1, L_2)\bigg|_{L_1,L_2=1/3}. \tag{A14}$$

Thus, one has for the effective polarizability

$$\langle\alpha\rangle = \langle\alpha\rangle_0 + I_1\Delta + I_2\Delta^2 + \ldots. \tag{A15}$$

As it follows from equations (A1), (A10) and (A14), if

$$\lim_{\Delta\to 0}\left(\frac{\partial P(L_1, L_2)}{\partial\Delta}\right) = 0 \tag{A16}$$

then $I_1 = 0$.

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