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Effective conductivity of a macroscopically inhomogeneous dispersion

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Abstract—Stationary transport of a scalar quantity, such as heat, mass of an admixture or electric charge, through a moderately concentrated composite material that contains identical spherical inclusions imbedded into a continuous matrix is considered. The packing of the spheres is locally random but the material is slightly non-uniform in the sense that there exists a gradient of the mean concentration, the linear scale of which is much larger than the size of the sphere. Conduction is shown to be characterized by concentration dependent bulk conductivity coefficients that make up a second-order tensor. In particular, the occurrence of an additional constituent of the transient flux of the quantity which is directed along the concentrational gradient is possible. The conductivities are expressible in terms of two scalar functions of both concentration and its squared gradient which are found for moderately concentrated materials and an arbitrary relation between the conductivities of a pure matrix and of the material of an inclusion.

1. INTRODUCTION

When the length-scale of a heat transfer process in a heterogeneous medium considerably exceeds the linear microscale of the inner structure of the medium, the process can be described with the help of continuum methods. In such a case the medium is regarded as an arrangement of juxtaposed interacting continua being representatives of different phases or discernable components of the medium. Each fictitious continuum possesses its own mean temperature. It is characterized by a specific effective thermal conductivity. If the medium is spatially uniform in the macroscopic respect, this enables one to express the transient heat fluxes through the continua as vectors proportional to the gradients of the corresponding mean temperatures and, further, to formulate heat transfer equations on the basis of either volume [1, 2] or ensemble [3] averaging technique, heat exchange fluxes between the continua being allowed for in addition. Then there arises the problem of finding out relevant constitutive equations, that is, of obtaining the effective conductivities as well as the quantities that specify the exchange fluxes as functions of both structural characteristics of the medium and physical parameters of its phases. Under steady conditions the mean phase temperatures normally coincide between themselves and the exchange fluxes vanish everywhere save for narrow regions adjacent to the boundaries of the medium. Then merely the phase conductivities remain to be specified.

Similar problems appear when one deals with the stationary transfer of the mass of an admixture and of electric charge, as well as while studying the effective electric and magnetic permeabilities of heterogeneous media at zero frequency. Due to the sameness of the mathematical formulations of all these problems, the final results concerning the effective diffusivity, electric conductivity and indicated permeabilities are basically identical to those having relevance to thermal conductivity. This means that it is quite sufficient to pay attention only to heat transfer, as is the case with the present paper. It is worth mentioning, however, that similar problems identifying rheological equations and determining the effective viscosity of suspensions or elastic moduli of solid bodies are somewhat different since they involve the investigation of the transfer of momentum which is a vector quantity.

A number of different approaches have been put forward in order to get reliable representations of the effective properties of heterogeneous media of different structure and, in particular, of disperse systems. The number of papers written on the subject is enormous; a review of conventional methods suggested so far for this purpose in view is to be found in refs. [4, 5]. In what follows, a modern method will be used which is based on the ensemble averaging technique, and the ideas of the self-consistent field theory which was developed in ref. [6].

A general feature of the great majority of attempts to study the effective thermal conductivity and other bulk properties of disperse mixtures is that a mixture under consideration is commonly viewed, in either an explicit or implicit way, as a macroscopically uniform one. This means that all its macroscopic characteristics are presumed to be independent of coordinates. Then the tensor structure of the transient fluxes of heat, mass, momentum and other quantities

NOMENCLATURE

a	radius of spheres
$\mathbf{I}$	unit tensor
L	length-scale of dispersion inhomogeneity
$\mathbf{l}$	unit vector in direction of temperature gradient
$\mathbf{m}$	unit vector in direction of concentration gradient
P_n^m	Legendre polynomial
$\mathbf{q}$	heat flux
$\mathbf{r}$	dimensionless radius-vector with origin at test sphere centre
$\mathbf{x}$	dimensionless radius-vector
x, z	dimensionless coordinates.

Greek symbols

β, γ	coefficients introduced in equation (3)
ε	small parameter, a/L
θ, φ	angular coordinates
κ	conductivity ratio, λ_1/λ_0
Λ_1, Λ_2	coefficients introduced in equations (2)
$\boldsymbol{\lambda}$	thermal conductivity tensor
λ_0, λ_1	thermal conductivities of matrix and material of spheres
ρ	volume concentration of spherical inclusions
τ	temperature.

Superscripts

*	perturbations of mean temperature caused by test sphere
′	connected with test sphere
^	field inside test sphere.

under question, which play the role of thermodynamic fluxes, are entirely determined by corresponding gradients of temperature, admixture concentration, velocity components, etc., which stand for appropriate thermodynamic forces.

However, it is certainly not so in the opposite case, when averaged structural mixture properties, and primarily the volume concentration of discrete inclusions, are macroscopically non-uniform. Then the gradients of these properties emerge, which can contribute to the fluxes and thereby change their structure in a rather drastic manner. In particular, one can anticipate that a component of the heat flux normal to the direction of the temperature gradient will make its appearance in a disperse mixture if the gradients of the mixture concentration are not parallel or antiparallel to that direction, let alone a possible dependence of the thermal conductivity on the modulus of the latter gradient.

Surprisingly enough, only a few indications to this important effect of principal interest in the current literature are known. First of all, it was stated and proved in ref. [7] that the velocity of hindered setting in a dilute suspension depends on the macroscopic vertical inhomogeneities of the suspension. Later on, a similar model was proposed for heat transfer in a dilute non-uniform dispersion of spherical particles [8]. Lastly, an attempt to revise and refine the modern methods of averaging as applied to slightly inhomogeneous disperse mixtures was recently begun in ref. [9].

In what follows, we shall treat heat transfer under steady conditions in a dispersion of equal spheres randomly distributed in the ambient continuous medium. The volume concentration of the spheres in the dispersion is assumed to be a linear function of one of the Cartesian coordinates, the length-scale of such a dependence being large as compared with the sphere radius. The orientation of the gradient of the mean temperature, which is supposed to be identical for the ambient medium and the embedded spheres relative to that of the concentration may be arbitrary. However, the volume concentration is either small as compared with unity or moderate, thus giving an opportunity to employ a simplified model of transfer, within the frames of which the non-overlapping property of rigid spheres may safely be ignored [6]. Such an assumption suffices for drawing all significant principal conclusions that concern the effect of the spatial inhomogeneity on the effective transfer properties of a dispersion. Generalization to dispersions of higher concentration can be attained at the cost of making the calculations more complicated, but without introducing new difficulties of a principal nature. Since the reasoning and calculation presented below are also called to provide for a guiding manual that indicates how to deal with more complex situations, they are successively set out in some detail.

2. MEAN TEMPERATURE FIELD

Below we make use of dimensionless coordinates x scaled with the radius a of the spheres. Then the volume concentration of the spheres can be written as:

$$\rho(\mathbf{x}_0+\mathbf{r}) = \rho_0+\varepsilon(\mathbf{m}\cdot\mathbf{r}) \quad \rho_0 = \rho(x_0), \quad \mathbf{m} = \nabla\rho/|\nabla\rho|, \qquad (1)$$

in the vicinity of an arbitrary point $\mathbf{x}_0$, where $\varepsilon = |\nabla\rho|$ is a small parameter of the order of a/L, L being the length-scale of the spatial macroscopic inhomogeneity of the dispersion under consideration.

The mean heat flux is to be defined in the form:

$$\mathbf{q} = -\alpha^{-1}\hat{\boldsymbol{\lambda}}\nabla\tau \quad \hat{\boldsymbol{\lambda}} = \Lambda_1\mathbf{I}+\varepsilon^2\Lambda_2(\mathbf{m}*\mathbf{m}), \qquad (2)$$

where $\hat{\lambda}$ stands for a conductivity tensor, $\mathbf{I}$ is the unit tensor, an asterisk signifies the operation of diadic multiplication and $\tau = \tau(\mathbf{x})$ presents the mean temperature field, Λ_j ($j = 1, 2$) being regarded as some unknown coefficients. It follows from equation (2) that:

$$\mathbf{q} = -(\lambda_0/\alpha)[\beta(\rho)\nabla\tau + \varepsilon^2\gamma(\rho)(\mathbf{m}\cdot\nabla\tau)\mathbf{m}], \quad (3)$$

if Λ_j are scaled with the thermal conductivity λ_0 of the ambient medium, $\Lambda_1 = \lambda_0\beta(\rho)$ and $\Lambda_2 = \lambda_0\gamma(\beta)$, β and γ depending additionally on the ratio $\kappa = \lambda_1/\lambda_0$ of the phase thermal conductivities and on the dimensionless parameter ε.

It should be emphasized that the expression of the tensor $\hat{\lambda}$ of the second rank in equation (2) is the most general one which could be constructed on the basis of the unit tensor and of only a vector $\mathbf{m}$ inherent to the dispersion on average, while formula (3) presents the heat flux as the most general linear vector combination that could be formed with the help of vectors $\mathbf{m}$ and $\nabla\tau$, which are only capable of affecting heat transfer in that dispersion.

When confining ourselves to the analysis of the situation accurately to the terms of the order of ε^2 inclusive, we are free, with account of equation (1), to assume that:

$$\beta(\rho) = \beta_0[1+\varepsilon\beta_1(\mathbf{m}\cdot\mathbf{r})^2+\varepsilon^2\beta_2(\mathbf{m}\cdot\mathbf{r})^2] \quad \beta_0 = \beta(\rho_0),$$

$$\beta_1 = \beta_0^{-1}\,\mathrm{d}\beta/\mathrm{d}\rho|_{\rho=\rho_0} \quad \beta_2 = 0.5\beta_0^{-1}\,\mathrm{d}^2\beta/\mathrm{d}\rho^2|_{\rho=\rho_0},$$

$$\gamma(\rho) = \beta_0\gamma_0 \quad \gamma_0 = \beta_0^{-1}\gamma(\rho_0), \quad (4)$$

where again β_0 and γ_0 may depend on ε and κ.

The equation of heat conservation in the dispersion reduces under steady conditions to $\nabla\mathbf{q} = 0$ [3]. With the accuracy up to terms of the order of ε^2 this gives:

$$[1+\varepsilon\beta_1(\mathbf{m}\cdot\mathbf{r})+\varepsilon^2\beta_2(\mathbf{m}\cdot\mathbf{r})^2]\Delta\tau+\varepsilon\beta_1(\mathbf{m}\cdot\nabla\tau)$$
$$+2\varepsilon^2\beta_2(\mathbf{m}\cdot\mathbf{r})(\mathbf{m}\cdot\nabla\tau)+\varepsilon^2\gamma_0(\mathbf{m}\cdot\nabla)(\mathbf{m}\cdot\nabla\tau) = 0. \quad (5)$$

If a solution of this equation is looked for in the form:

$$\tau = \tau_0+\varepsilon\tau_1+\varepsilon^2\tau_2, \quad (6)$$

then equation (5) leads to a set of equations:

$$\Delta\tau_0 = 0 \quad \Delta\tau_1 = -\beta_1(\mathbf{m}\cdot\nabla\tau_0),$$

$$\Delta\tau_2 = (\beta_1^2-2\beta_2)(\mathbf{m}\cdot\mathbf{r})(\mathbf{m}\cdot\nabla\tau_0)$$
$$-\gamma_0(\mathbf{m}\cdot\nabla)(\mathbf{m}\cdot\nabla\tau_0)-\beta_1(\mathbf{m}\cdot\nabla\tau_1). \quad (7)$$

It is convenient to choose one of the coordinate axes (say, z) along unit vector $\mathbf{m}$. If the direction of $\mathbf{q}$ is constant throughout the entire region under study and defined by a unit vector $\mathbf{l}$ lying in the plane (x, z) a general situation with an arbitrary mutual orientation of $\mathbf{m}$ and $\mathbf{l}$ can be subdivided, because of the linearity of heat transfer, into two elementary ones, when $\mathbf{l}$ is directed either along $\mathbf{m}$ ($\mathbf{m}\cdot\mathbf{l} = 1$) or normally to $\mathbf{m}$ ($\mathbf{m}\cdot\mathbf{l} = 0$). In the first case (situation I) we get from equations (2), (6) and (7):

$$\mathbf{q} = -\frac{\lambda_0}{a}\beta_0\mathbf{m} \quad \nabla\tau = \frac{\beta_0}{\beta+\varepsilon^2\gamma}\mathbf{m},$$

$$\tau = \mathrm{const}+z-\varepsilon\beta_1\frac{z^2}{2}-\left[\gamma_0 z+(\beta_2-\beta_1^2)\frac{z^3}{3}\right]\varepsilon^2$$

$$z = \mathbf{m}\cdot\mathbf{r}. \quad (8)$$

In the second case (situation II):

$$\mathbf{q} = -\frac{\lambda_0}{a}\beta_0(1+\varepsilon\beta_1 z+\varepsilon^2\beta_2 z^2)\mathbf{l} \quad \nabla\tau = \mathbf{l},$$

$$\tau = \mathrm{const}+x \quad x = \mathbf{l}\cdot\mathbf{r} \quad z = \mathbf{m}\cdot\mathbf{r}. \quad (9)$$

In both cases the mean temperture gradient is taken to be equal to unity. In a general case, a suitable solution of equation (7) may be written as a sum of those in equations (8) and (9).

In order to get the functions β_0 and γ_0, which are necessary to make equation (3) closed by virtue of equation (4), we shall resort to the method of ref. [6] according to which a special boundary problem about perturbations of the mean temperature field caused by a single test sphere has to be solved. The solution of the problem is to be used while formulating non-linear algebraic equations for the mentioned functions that reflect requirements of the self-consistency of the theory being developed [6]. Formulae (8) and (9) happen to be sufficient to state the test sphere problem in an explicit form.

3. TEST PARTICLE PROBLEM

Let us consider the temperature field around a test sphere with the centre positioned at the point x_0'. If the dispersion concentration is not too high, we may neglect the necessary condition that rigid spheres cannot overlap. It amounts to the concept that regions nearby the test sphere surface do not differ from those far away as pertains to their ability to transport heat, and there is no concentric layer throughout which the effective thermal conductivity varies [6]. Then we arrive at an approximate model of moderately concentrated dispersions. In conformity with this model, the test sphere is regarded as that immersed into a homogeneous fictitious medium whose properties coincide with those of the dispersion as a whole.

The temperature outside the test sphere ($r = |\mathbf{x}-\mathbf{x}_0'| > 1$) can be presented as $\tau+\tau^*$, where τ^* is the perturbation of the mean temperature τ, identified in equation (6) or in equations (8) and (9), due to the presence of that sphere. The temperature inside the sphere ($r < 1$) is denoted as $\hat{\tau}$. Similarly to equation (6) we assume that:

$$\tau^* = \tau_0^*+\varepsilon\tau_1^*+\varepsilon^2\tau_2^* \quad \hat{\tau} = \hat{\tau}_0+\varepsilon\hat{\tau}_1+\varepsilon^2\hat{\tau}_2. \quad (10)$$

It is easy to see that variables τ_j^* satisfy equations of the same form as those listed in equation (7), whereas $\hat{\tau}_j$ are governed by the usual Laplace equation. Pertinent boundary conditions are to be written as follows:

$$\hat{\tau}_j < \infty \quad r = 0; \quad \tau_j^* \to 0 \quad r \to \infty; \quad j = 0, 1, 2, \tag{11}$$

and at $r = 1$:

$$\hat{\tau}_j = \tau_j + \tau_j^* \quad j = 0,1,2; \quad \kappa \mathbf{n}\nabla\hat{\tau}_0 = \beta_0' \mathbf{n}\cdot\nabla(\tau_0 + \tau_0^*),$$

$$\kappa \mathbf{n}\nabla\hat{\tau}_1 = \beta_0'[\mathbf{n}\cdot\nabla(\tau_1+\tau_1^*) + \beta_1'(\mathbf{m}\cdot\mathbf{r})\mathbf{n}\nabla(\tau_0+\tau_0^*)],$$

$$\kappa \mathbf{n}\nabla\hat{\tau}_2 = \beta_0'[\mathbf{n}\cdot\nabla(\tau_2+\tau_2^*) + \beta_1'(\mathbf{m}\cdot\mathbf{r})\mathbf{n}\nabla(\tau_1+\tau_1^*) + \beta_2'(\mathbf{m}\cdot\mathbf{r})^2\mathbf{n}\nabla(\tau_0+\tau_0^*) + \gamma_0'(\mathbf{m}\cdot\nabla(\tau_0+\tau_0^*))(\mathbf{m}\cdot\mathbf{n})]. \tag{12}$$

Here $\mathbf{n}$ is the external normal unit vector at the sphere surface and the prime is introduced to emphasize that β_j' ($j = 0, 1, 2$) and γ_0' refer to the point r_0', where $\rho = \rho_0'$, but not to r_0.

The mentioned equations and the conditions in equations (11) and (12) determine the test sphere problem. It is subdivided into three correct boundary problems for τ_j^* and $\hat{\tau}_j$ ($j = 0, 1, 2$) which correspond to different orders in powers of the small parameter ε. It is convenient to solve these problems separately for situations I and II identified by equations (8) and (9), respectively. It should be noted that merely $\hat{\tau}_j(r)$ is actually needed to formulate the self-consistency equations [6].

3.1. *The zeroth approximation*

When considering situation I with allowance for the last equation in set (8), we get, in a straightforward manner, that:

$$\tau_0 = Az = Ar\cos\theta \quad \hat{\tau}_0 = Br^{-3}z = Br^{-2}\cos\theta, \tag{13}$$

where:

$$A = \frac{3\beta_0'}{2\beta_0'+\kappa} \quad B = \frac{\beta_0'-\kappa}{2\beta_0'+\kappa} \quad \kappa = \frac{\lambda_1}{\lambda_0}. \tag{14}$$

The same expressions holds true also in situation II if $x = r\sin\theta\cos\varphi$ is substituted for z in relations (13), θ and φ being the polar and azimuth angles of the usual spherical coordinate system.

3.2. *The first approximation*

In situation I when expressions (13) are relevant, the right-hand side of the equation for τ_1^* [see the second equation in set (7)] turns out to be:

$$-\beta_1'(\mathbf{m}\cdot\nabla\tau_0^*) = -\beta_1'\frac{\partial}{\partial z}\frac{Bz}{r^3} = -\beta_1'\frac{B}{r^3}(1-3\cos^2\theta),$$

and a partial solution of that equation reads:

$$\tau_{1,\mathrm{p}}^* = -\frac{\beta_1'}{2}\frac{Bz^2}{r^3} = -\frac{\beta_1'}{2}\frac{B}{r}\cos^2\theta = -\frac{\beta_1'}{6}\frac{B}{r}(1+2P_2),$$

P_2 being a Legendre polynomial. A general solution of the Laplace equation for τ_1^* and $\hat{\tau}$ outside and within the test sphere, respectively, that satisfies conditions (11) is well known. Thus in situation I we obtain:

$$\tau_1^* = \beta_1' B\left[-\frac{1}{6r}(1+2P_2) + \frac{C_1}{r^3}P_2 + \frac{C_2}{r}\right]$$

$$\hat{\tau}_1 = \beta_1' B(C_3 r^2 P_2 + C_4). \tag{15}$$

The constants involved in equations (15) have to be found from equations (12), which give:

$$C_1 = \frac{-3\beta_0'+2\kappa}{3(3\beta_0'+2\kappa)} \quad C_2 = -\tfrac{1}{2}$$

$$C_3 = -\frac{2\beta_0'}{3\beta_0'+2\kappa} \quad C_4 = -\tfrac{2}{3}. \tag{16}$$

Quite similarly, in situation II we get in succession:

$$-\beta_1'(\mathbf{m}\cdot\nabla\tau_0^*) = 3\beta_1'\frac{B}{r^3}\cos\theta\sin\theta\cos\varphi,$$

$$\tau_{1,\mathrm{p}}^* = -\frac{\beta'}{6}\frac{B}{r}P_2^1\cos\varphi,$$

and next:

$$\tau_1^* = \beta_1' B\left[-\frac{1}{6r} + \frac{D_1}{r^3}\right]P_2^1\cos\varphi,$$

$$\hat{\tau}_1 = \beta_1' B D_2 r^2 P_2^1 \cos\varphi, \tag{17}$$

with:

$$D_1 = \frac{1}{6B}\frac{2\beta_0' - B(3\beta_0'-2\kappa)}{3\beta_0'+2\kappa} \quad D_2 = \frac{\beta_0'}{3B}\frac{1-3B}{3\beta_0'+2\kappa}. \tag{18}$$

Parameter B involved in both equations (16) and (18) is defined by equation (14).

3.3. *The second approximation*

Consider first, situation I. In this case we derive from equations (13) and (15) the relations:

$$(\mathbf{m}\cdot\mathbf{r})(\mathbf{m}\cdot\nabla\tau_0^*) = \frac{B}{r^3}(1-3\cos^2\theta)\cos\theta,$$

$$(\mathbf{m}\cdot\nabla)(\mathbf{m}\cdot\nabla\tau_0^*) = -\frac{3B}{r^4}(3-5\cos^2\theta)\cos\theta,$$

$$(\mathbf{m}\cdot\nabla\tau_0^*) = \frac{\beta_1' B}{2}\left[-\frac{1}{r}(1-3\cos^2\theta) + \frac{3C_1}{r^4}(3-5\cos^2\theta)\right]\cos\theta,$$

which define the right-hand side of the equation for τ_2^*, a relevant partial solution of which takes the form:

$$\tau_{2,\mathrm{p}}^* = V_1(P_3+4P_1) + V_2 r^{-2}(2P_3+3P_1), \tag{19}$$

$$V_1 = \frac{B}{10}[\tfrac{3}{2}(\beta_1')^2 - 2\beta_2'] \quad V_2 = V_{20} + \gamma_0' V_{21}, \tag{20}$$

$$V_{20} = -\tfrac{3}{20}\gamma_1(\beta_1')^2 \quad V_{21} = \tfrac{3}{10}B.$$

By making use of both the general solution of the uniform equations for τ_2^* and boundary conditions (12) we get, after a simple, however lengthy, calculation:

$$\tau_2^* = \tau_{2,p} + E_1 r^{-2} P_1 + E_2 r^{-4} P_3,$$
$$\hat{\tau}_2 = E_3 r P_1 + E_4 r^3 P_3, \quad (21)$$

where $\tau_{2,p}^*$ is identified in equation (19) and:

$$E_1 = E_{10} + \gamma_0' E_{11} \quad E_2 = E_{20} + \gamma_0' E_{21},$$

$$E_{10} = \frac{1}{2\beta_0' + \kappa}\{\beta_0'[\tfrac{2}{5}(\beta_1')^2(2-2C_1)B - \tfrac{6}{5}\beta_2' B - 6V_{20}] - \kappa(4V_1 + 3V_{20})\},$$

$$E_{11} = -\frac{1}{2\beta_0' + \kappa}\{\beta_0'[\tfrac{4}{5}B + 6V_{21}] + 3\kappa V_{21}\},$$

$$E_3 = E_1 + 4V_1 + 3V_2 \quad E_4 = E_2 + V_1 + 2V_2. \quad (22)$$

Quantities B, C_1 and V_1, V_2, V_{20}, V_{21} are defined in equations (14), (16) and (20), respectively. Expressions of E_{20} and E_{21} are readily obtainable as well. They are not written down, however, since they do not affect the subsequent calculation.

It should be noted that equation (19) does not vanish when $r \to \infty$, so that equation (21) does not satisfy the boundary condition at infinity listed in conditions (11). This is due to the fact that the validity of the expansion of $\beta(\rho)$ in equation (4) is restricted by a space region in which the condition $r < \varepsilon^{-1}$ holds true. Otherwise that expansion is evidently incorrect. This difficulty can easily be avoided by imposing the condition of τ_2^* being equal to zero at a certain radial distance $r = R_m \approx \varepsilon^{-1}$, which happens to be large as compared with unity in view of the supposed smallness of ε. After performing manipulations needed to derive new expressions of τ_2^* and $\hat{\tau}_2$, which depend additionally on R_m, we are free to use the limit transition $R_m \to \infty$. As a result, we again obtain equation (19) which must now be thought of as the zeroth approximation in powers of R_m^{-1}.

A similar calculation can be carried out in situation II as well. We have:

$$(\mathbf{m}\cdot\mathbf{r})(\mathbf{m}\cdot\nabla\tau_0^*) = -\frac{B}{r^2}\cos^2\theta\sin\theta\cos\varphi,$$

$$(\mathbf{m}\cdot\nabla)(\mathbf{m}\cdot\nabla\tau_0^*) = -\frac{3B}{r^4}(1-5\cos^2\theta)\sin\theta\cos\varphi,$$

$$(\mathbf{m}\cdot\nabla\tau_1^*) = -\frac{\beta_1' B}{2r^2}\left\{\left[1-\frac{3D_1}{r^2}\right] - 3\left[1-\frac{5D_1}{r^2}\right]\cos^2\theta\right\}\sin\theta\cos\varphi,$$

and, next:

$$\tau_{2,p}^* = [\tfrac{2}{5}W_1(3P_1^1 + \tfrac{1}{3}P_3^1) + W_2 r^{-2} P_1^1 + \tfrac{1}{5}W_3 r^{-2}(P_1^1 + \tfrac{2}{3}P_3^1)]\cos\varphi, \quad (23)$$

$$W_1 = \frac{B}{24}[5(\beta_1' - 4\beta_2')] \quad W_2 = -\frac{B}{4}(\beta_1')^2,$$

$$W_3 = W_{30} + \gamma_0' W_{31} \quad W_{30} = -\tfrac{3}{4}BD_1(\beta_1')^2$$

$$W_{31} = \tfrac{3}{2}B. \quad (24)$$

Further, when accounting for conditions (11) we arrive at:

$$\tau_2^* = \tau_{2,p}^* + (F_1 r^{-2} P_1^1 + F_2 r^{-4} P_3^1)\cos\varphi,$$

$$\hat{\tau}_2 = (F_3 r P_1^1 + F_4 r^3 P_3^1)\cos\varphi, \quad (25)$$

instead of equations (21) and, by again using the boundary conditions at the sphere surface, we get:

$$F_1 = F_{10} + \gamma_0' F_{11} \quad F_2 = F_{20} + \gamma_0' F_{21},$$

$$F_{10} = \frac{1}{2\beta_0' + \kappa}\{\beta_0'[\tfrac{3}{5}(\beta_1')^2(\tfrac{1}{6} - 3D_1)B + \tfrac{1}{5}\beta_2'(1-2B) - 2W_2] - \kappa[\tfrac{6}{5}W_1 + W_2]\},$$

$$F_{11} = -\frac{0.3}{2\beta_0' + \kappa}(\beta_0' + \kappa)B,$$

$$F_3 = F_1 + \tfrac{6}{5}W_1 + W_2 + \tfrac{1}{5}W_3,$$

$$F_4 = F_2 + \tfrac{2}{15}W_1 + \tfrac{2}{15}W_3, \quad (26)$$

instead of equations (22). Here B, D and W_1, W_2, W_{30}, W_{31} are understood as quantities identified in equations (14), (18) and (24) and expressions for F_{20} and F_{21} are again dropped out.

4. REQUIREMENT OF SELF-CONSISTENCY

In accordance with the method suggested in ref. [6], the mean transient heat flux is expressible in the following form:

$$\mathbf{q}(x_0) = -\frac{\lambda_0}{a}\nabla\tau(x_0) - (\lambda_1 - \lambda_0)\frac{3}{4\pi a} \times \int_{|x_0 - x_0'| \leq 1} \rho(x_0')\nabla x_0 \hat{\tau}(x_0|x_0')\,\mathrm{d}x_0'. \quad (27)$$

Again the dimensionless coordinates are used, and the integration is to be carried out over those positions x_0' of the test sphere centre in which the point x_0, in which the heat flux had to be determined, lies within that sphere. Because of equations (1), the expression:

$$\rho(x_0') = \rho(x_0 - \mathbf{r}) = \rho(x_0) - \varepsilon(\mathbf{m}\cdot\mathbf{r}) \quad \mathbf{r} = x_0 - x_0', \quad (28)$$

is valid and must be used in the integrand of equation (27).

This expression helps one to obtain expansions of quantities β_j' ($j = 0, 1, 2$) and γ_0', regarded as functions of x_0', in the vicinity of the point x_0 in powers of $r = |x_0 - x_0'|$. This leads to writing these quantities in terms of similar ones related to the point x_0. In particular, $\beta_2' = \beta_2$ and $\gamma_0' = \gamma_0$ accurately to the terms of the order of ε^2.

Let us calculate $\nabla_{x_0}\hat{\tau}(x_0|x_0') = \nabla\hat{\tau}(\mathbf{r})$ with the help of equations (10) and of the expressions for $\hat{\tau}_j(\mathbf{r})$, $j = 0, 1, 2$, derived in the preceding section. After simple manipulations we get in situation I with the accepted accuracy:

$$\nabla\hat{\tau}_0 = \frac{3\beta_0'}{2\beta_0'+\kappa} = T_{00}+\varepsilon T_{01} r\cos\theta+\varepsilon^2 T_{02} r^2\cos^2\theta,$$

$$\nabla\hat{\tau}_1 = 2\beta_1' BC_3 r\cos\theta = (T_{11}+\varepsilon T_{12} r\cos\theta) r\cos\theta,$$

$$\nabla\hat{\tau}_2 = T_{21}+3T_{22}r^2(3\cos^2\theta-1), \tag{29}$$

where the following parameters are introduced:

$$T_{00} = \frac{3\beta_0}{2\beta_0+\kappa} \quad T_{01} = \frac{3\beta_0\beta_1^{\kappa}}{(2\beta_0+\kappa)^2},$$

$$T_{02} = \frac{3\beta_0}{(2\beta_0+\kappa)^3}[\beta_2(2\beta_0+\kappa)-2(\beta_1)^2\beta_0],$$

$$T_{11} = \frac{4\beta_0\beta_1(\beta_0-\kappa)}{(2\beta_0+\kappa)(3\beta_0+2\kappa)}$$

$$T_{12} = -4\{[-2\beta_0\beta_2(\beta_0-\kappa)-(\beta_0\beta_1)^2](2\beta_0+\kappa) \times(3\beta_0+2\kappa)+(\beta_0\beta_1)^2(\beta_0-\kappa)(12\beta_0+7\kappa)\} \times[(2\beta_0+\kappa)(3\beta_0+2\kappa)]^{-2}$$

$$T_{21} = T_{210}+\gamma_0 T_{211} \quad T_{210} = E_{10}+4V_1+3V_{20}$$

$$T_{211} = E_{11}+3V_{21}. \tag{30}$$

Here quantities β_j ($j = 0, 1, 2$) and γ_0 referred to the point x_0 are involved, and the above expressions of different coefficients are used.

Similarly, in situation II we obtain:

$$\nabla\hat{\tau}_0 = \frac{3\beta_0'}{2\beta_0'+\kappa} = T_{00}+\varepsilon T_{01} r\cos\theta+\varepsilon^2 T_{02} r^2\cos^2\theta,$$

$$\nabla\hat{\tau}_1 = 3\beta_1' BD_3 r\cos\theta = (\hat{T}_{11}+\varepsilon\hat{T}_{12} r\cos\theta) r\cos\theta,$$

$$\nabla\hat{\tau}_2 = \hat{T}_{21}+\hat{T}_{22}(5\cos^2\theta-1-2\sin^2\theta\cos^2\varphi)r^2, \tag{31}$$

where coefficients T_{00}, T_{01} and T_{02} are the same as those defined in equations (30) and:

$$\hat{T}_{11} = \frac{\beta_0\beta_1(-\beta_0+4\kappa)}{(2\beta_0+\kappa)(3\beta_0+2\kappa)},$$

$$\hat{T}_{12} = \{[-2\beta_0\beta_2(-\beta_0+4\kappa)+(\beta_0\beta_0^2)](2\beta_0+\kappa) \times(3\beta_0+2\kappa)+(\beta_0\beta_1)^2(-\beta_0+4\kappa)(12\beta_0+7\kappa)\} \times[(2\beta_0+\kappa)(3\beta_0+2\kappa)]^{-2},$$

$$\hat{T}_{21} = \hat{T}_{210}+\gamma_0 T_{211},$$

$$\hat{T}_{210} = F_{10}+\tfrac{6}{5}W_1+W_2+\tfrac{1}{5}W_{30},$$

$$\hat{T}_{211} = F_{11}+\tfrac{1}{5}W_{31}. \tag{32}$$

Now we are able to find the integral included in equation (27). When representing the actual heat flux as a superposition of those corresponding to situations I and II and by making use of equations (28), (29) and (31), we in situation I finally get:

$$\frac{3}{4\pi}\int_{|x_0-x_0'|\leq 1}\rho(x_0')\nabla_{x_0}\hat{\tau}(x_0|x_0')\,\mathrm{d}x_0' = \rho_0 T_{00}+\varepsilon^2[0.2(\rho_0 T_{02}-T_{01}) +0.2(\rho_0 T_{12}-T_{11})+\rho_0 T_{21}],\quad \rho_0 = \rho(x_0), \tag{33}$$

where the coefficients T_{00}, T_{02}, T_{01}, T_{12}, T_{11}, T_{21} are defined by relations (30). Similar equations with T_{11}, T_{12} and T_{21} being replaced by $\hat{T}_{11}$, $\hat{T}_{12}$ and $\hat{T}_{21}$ are valid for situation II.

Equations (27) and (33) can be written at any point x_0, so that now there is no sense in retaining a zero subscript in the notation of x_0 and ρ_0. When equating the expressions of $\mathbf{q}$ that result from equations (3) and (27) and allowing for equation (33), we derive equations for situations I and II:

$$\beta(\rho)+\varepsilon^2\gamma(\rho) = 1+(\kappa-1) \times\left\{\rho T_{00}+\frac{\varepsilon^2}{5}[\rho(T_{02}+T_{12}+5T_{21})-T_{01}-T_{11}]\right\}$$

$$\beta(\rho) = 1+(\kappa-1)\left\{\rho T_{00}+\frac{\varepsilon^2}{5}[\rho(T_{02}+\hat{T}_{12} +5\hat{T}_{21})-T_{01}-\hat{T}_{11}]\right\}. \tag{34}$$

Now it is easy to prove that equation (34) is satisfied if the expansion:

$$\beta(\rho) = \beta^{(0)}(\rho)[1+\varepsilon^2\beta^{(1)}(\rho)] \tag{35}$$

is true, in which case the separation of terms of different orders in powers of ε yields:

$$\beta^{(0)} = 1+(\kappa-1)\rho T_{00}, \tag{36}$$

and

$$\beta^{(0)}(\beta^{(1)}+\gamma_0) = (\kappa-1)\{0.2(\rho T_{02}-T_{01}) +0.2(\rho T_{12}-T_{11})+\rho(T_{210}+\gamma_0 T_{211})\},$$

$$\beta^{(0)}\beta^{(1)} = (\kappa-1)\{0.2(\rho T_{02}-T_{01})+0.2(\rho\hat{T}_{12}-\hat{T}_{11}) +\rho(\hat{T}_{210}+\gamma_0\hat{T}_{211})\}. \tag{37}$$

Equation (36) corresponds to retaining terms of the order of ε^0 whereas two equations (37) are obtained by singling out contributions proportional to either **m** or **l** in terms of the order ε^2. An equation stemming from the terms of the order of ε in equation (34) does not arise altogether. It is quite understandable because the functions in equations (2) and (3) determining the thermal conductivity tensor are true scalars, and this implies that they ought not to depend on the direction of **m**, that is, to contain the terms of the order of ε.

It is easy to obtain from equation (36) a familiar formula:

$$\beta^{(0)}(\rho) = \tfrac{1}{4}\{G+\sqrt{G^2+8\kappa}\} \quad G = 2-\kappa+3(\kappa-1)\rho. \tag{38}$$

It describes the effective thermal conductivity of a macroscopically uniform dispersion within the scope of the used model of moderately concentrated dispersions [6].

Equations (37) yield an expression for γ_0 whence:

$$\gamma_0 = \frac{(\kappa-1)[0.2(\rho T_{12}-T_{11}) - 0.2(\rho\hat{T}_{12}-\hat{T}_{11})+(\rho T_{210}-\hat{T}_{210})]}{\beta^{(0)}-(\kappa-1)\rho(T_{211}-\hat{T}_{211})}, \quad (39)$$

and, next:

$$\beta^{(1)} = \frac{(\kappa-1)}{\beta^{(0)}}[0.2(\rho T_{02}-T_{01}) + 0.2(\rho\hat{T}_{12}-\hat{T}_{11})+\rho(\hat{T}_{210}-\gamma_0\hat{T}_{211})]. \quad (40)$$

This defines also β_1 and β_2 introduced in equations (4) in the form:

$$\beta_1 = 3(\kappa-1)(G^2+8\kappa)^{-1/2},$$
$$\beta_2 = 4.5(\kappa-1)^2[G^2+8\kappa]^{-1}[1-G(G^2+8\kappa)^{-1/2}]. \quad (41)$$

Equations (38)–(41) suffice to describe the thermal conductivity tensor (2) in full detail.

5. EFFECTIVE CONDUCTIVITIES

In a physical dimensional reference frame the functions that determine the components of the conductivity tensor from equations (2) can be written as:

$$\Lambda_1 = \lambda_0\beta^{(0)}(1+\varepsilon^2\beta^{(1)}) \quad \Lambda_2 = \lambda_0\beta^{(0)}\gamma_0 \quad \varepsilon = \alpha|\nabla\rho|. \quad (42)$$

Such a structure of this tensor enables us to draw the following principal conclusions. First, the non-uniform dispersion under study represents an anisotropic heat-conducting medium characterized by a symmetric thermal conductivity tensor of the second rank. Second, scalar coefficients of thermal conductivity at a point are dependent on only two functions of the local concentration at the point and of its gradient, both of them including the phase conductivity ratio, $\kappa = \lambda_1/\lambda_0$, as a parameter. Third, values of coefficients relating the heat flux to the mean temperature gradient at a certain point differ from the isotropic bulk thermal conductivity of a corresponding uniform dispersion, the constant concentration of which equals that of the original dispersion at the point under otherwise identical conditions, by quantities of the second order in the concentrational gradient.

It is instructive to consider, by way of example, particular situations when $\nabla\tau$ is either parallel or anti-parallel or normal to $\mathbf{m}$. In these situations we have, respectively:

$$\mathbf{q} = -\lambda_\parallel^+\nabla\tau \quad \lambda_\parallel^+ = \lambda_0\beta^{(0)}[1+\varepsilon^2(\beta^{(1)}+\gamma)] \quad (\mathbf{m}\cdot\nabla\tau) = |\nabla\tau|,$$
$$\mathbf{q} = -\lambda_\parallel^-\nabla\tau \quad \lambda_\parallel^- = \lambda_0\beta^{(0)}[1+\varepsilon^2(\beta^{(1)}-\gamma)] \quad (\mathbf{m}\cdot\nabla\tau) = -|\nabla\tau|,$$
$$\mathbf{q} = -\lambda_\perp\nabla\tau \quad \lambda_\perp = \lambda_0\beta^{(0)}(1+\varepsilon^2\beta^{(1)}) \quad (\mathbf{m}\cdot\nabla\tau) = 0. \quad (43)$$

Coefficients $\lambda_\parallel^+$, $\lambda_\parallel^-$ and $\lambda_\perp$ have the meaning of two longitudinal and of a lateral thermal conductivity with reference to the mutual orientation of $\nabla\tau$ and $\mathbf{m}$.

The anisotropy of heat conduction may also be described by introducing a complementary constituent of the effective heat flux directed along $\mathbf{m}$ irrespective of the direction of $\nabla\tau$, as is the case with definition (3).

Function $\beta^{(0)}(\rho)$ represents the relative thermal conductivity of macroscopically homogeneous dispersions of moderate concentration. It is illustrated in ref. [6], and so there is no need to dwell upon its dependence on ρ and on the parameter κ once more. What is important in the context of this paper is that it either turns to zero at a finite $\rho = \rho^*$, when κ is smaller than unity ($\rho^* = 2/3$ at $\kappa = 0$), or becomes unreally high at large κ (diverges at $\rho^* = 1/3$ when $\kappa = \infty$). This is an obvious consequence of the neglect of the spheres being unable to overlap, and sets limits on the applicability of the employed model of moderately concentrated disperse mixtures. If $\kappa \leqslant 1$, $\beta^{(0)}$ can be shown to give satisfactory results in the range of ρ from zero up to about 0.2–0.3. If $\kappa \gg 1$, the range of approximate validity of this function becomes even more narrow. Anyhow, the above results are surely inapplicable when $\rho \geqslant 0.3$, and so we shall consider $\beta^{(1)}$ and γ only within the interval $0 < \rho < 0.3$.

Dependences of the quantities under question on ρ are rather different in various ranges of κ. They are plotted in Figs. 1–4. When κ is much smaller than unity, both $\beta^{(1)}$ and γ are small and decrease with ρ; they become negative as ρ exceeds a certain value (Fig. 1). As κ grows when remaining smaller than unity, $\beta^{(1)}$ and γ become positive at any ρ (Fig. 2). When κ goes over the threshold $\kappa = 1$, $\beta^{(1)}$ and γ are almost constant and grow with ρ (Fig. 3). After that, an increase in κ results in the occurrence of slight maxima of both $\beta^{(1)}$ and γ as functions of ρ. At last, when κ is very large against unity, these maxima vanish, and $\beta^{(1)}$ and γ transform into increasing positive and decreasing negative functions, respectively (Fig. 4). It is worth noting that the absolute values of these functions become very large when $\kappa \gg 1$.

Thus the coefficients in equations (43) happen to be rather intricate functions of ρ and κ. In a general case, it is difficult to suggest reliable model situations which could be helpful in explaining the behaviour of these coefficients in considerable detail.

In a general case, a complicated picture of the local temperature field develops on the level of individual spheres, and it is not a simple matter to explain the particulars of the dependence of the mean heat flux upon ρ and κ. Numerical investigation of this field around and within several spheres, similar to that in ref. [10], is apparently needed for such an end in view.

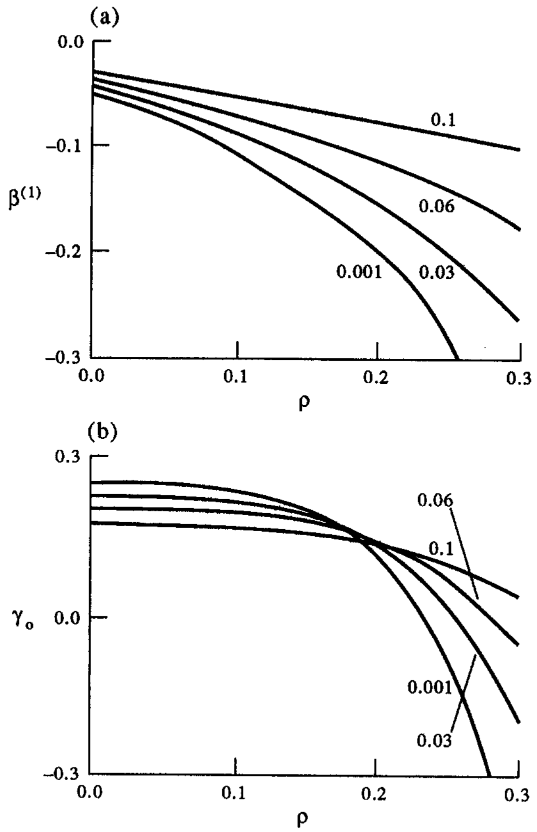

Fig. 1. Dimensionless coefficients $\beta^{(1)}$ (a) and γ_0 (b) as functions of ρ for $\kappa \ll 1$; figures at the curves give values of κ.

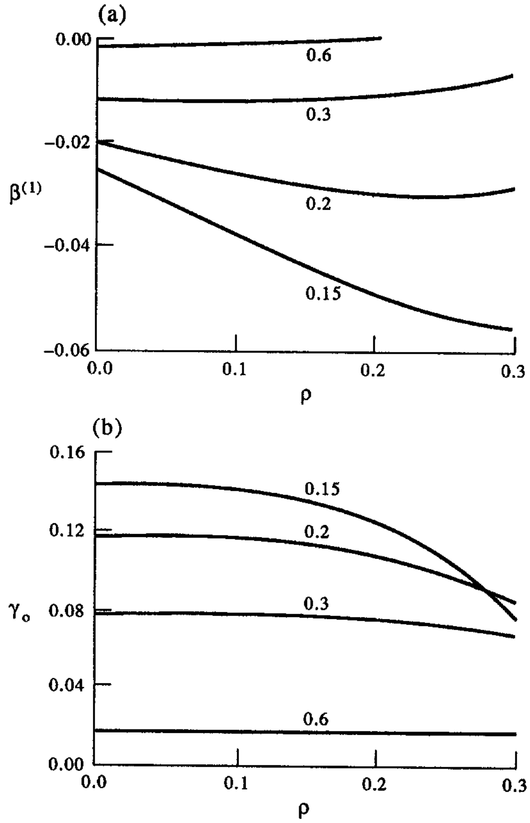

Fig. 2. The same as Fig. 1 for $\kappa < 1$.

6. CONCLUDING REMARKS

The main novel inference of the analysis of the present paper consists in establishing the fact that macroscopic inhomogeneity of either a composite material with discrete inclusions or a dispersion of another origin influences the dispersion conductivity and, in particular, gives rise to new effects of deviation of the mean heat flux direction from that of the mean temperature gradient. As a result, a scalar thermal conductivity happens to be insufficient to describe heat transfer in these media, and a tensor of the second rank is necessary for the purpose.

This effect seems to be undoubtedly important in principal, even though it is insignificant qualitatively, as is the case in disperse mixtures with badly conducting inclusions. However, a quantitative influence of the spatial inhomogeneity on the effective thermal conductivities may not be ignored in dispersions of well conducting inclusions ($\kappa \gg 1$), when $\beta^{(1)}$ and γ can remain large even after multiplication by the small factor ε^2 and essentially contribute to the conductivities identified in equations (43).

Among practical problems in which this effect is important, the heating of either a stationary [11] or a fluidized [12] granular bed by a hot rigid wall is to be mentioned. Because of the inpenetrability of the wall for solid particles of the bed, there originates an adjacent interlayer of excessive porosity, the thickness of which is of the order of the particle size. Due to the ensuing variation in the particle volume concentration across the interlayer, the effective thermal conductivity is monotonously decreasing from its value, specific for the bulk of the bed to that of air or another ambient gas, when the wall is approached. However, just the same variation favours the enlargement of the conductivity for reasons pointed out and discussed in this paper, that is, causes the opposite influence. It is evident that both factors must be taken into account while treating the problem.

It should be noted in this connection that much remains to be done in order to incorporate these factors into an analysis. First, the granular bed under consideration is not of moderate concentration, and a proper generalization of the above results is needed to get a reliable representation of the dependence of a pertinent effective conductivity on the concentration gradient. Second, all the results concerning the conductivity have been derived with the help of the continuum approximation. It is evident that a necessary condition of applicability of that approximation requires $\varepsilon = a/L \ll 1$, which is certainly not true within the wall interlayer. Elucidation of these points indicates tempting directions of future work.

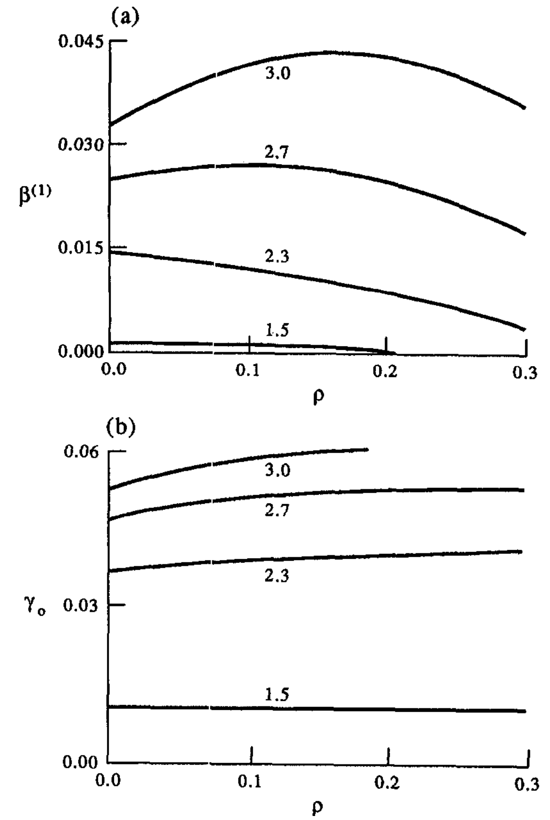

Fig. 3. The same as Fig. 1 for $\kappa > 1$.

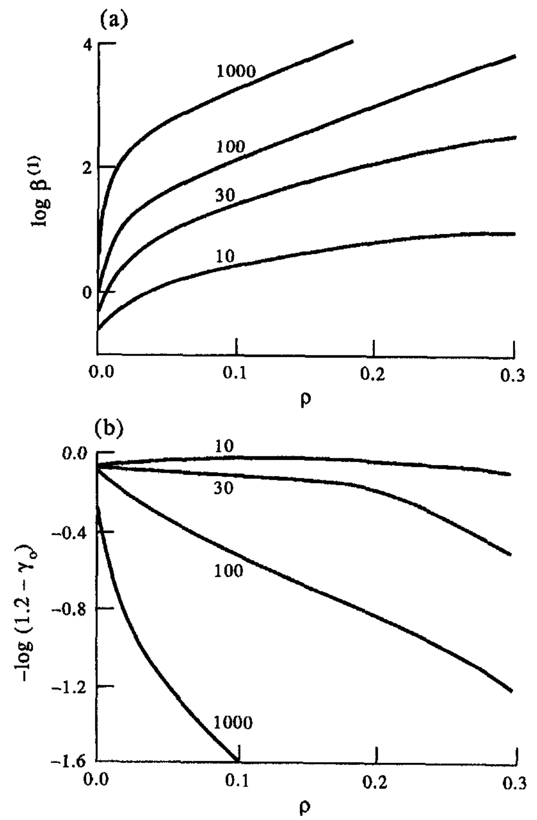

Fig. 4. The same as Fig. 1 for $\kappa \gg 1$.

It is worth noting that there is a distinction between the processes of heating and cooling of a granular bed with the aid of a solid wall. This inference is caused by a difference of the heat conductivities, equations (43), in the direction of the concentration gradient and in the opposite direction. Since the function γ is negative at large κ (Fig. 4), the rate of the former process is somewhat lower than that of the latter one. It is of principal significance because it offers an evident opportunity for an experimental check of the developed theory.

To conclude, we should emphasize once again that all the results obtained have relevance not only to heat transfer but also to transport of other scalar quantities, such as mass of admixture and electric charge.

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