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## THE EFFECTIVE THERMAL CONDUCTIVITY OF A SUSPENSION\*

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The effective thermal conductivity of an inhomogeneous suspension is considered for the case of low and moderate volume densities of randomly distributed spherical particles. A mathematical apparatus of convolutions of the  $\Lambda$ -functions is developed enabling closed formulas to be derived for the dipole moment of a particle in the system. An exact expression for the dipole moment averaged over the ensemble that is accurate to terms of the order of the square of the particle density is given for a spatially homogeneous distribution of particles. The effective thermal conductivity of the suspension is calculated to the same approximation. It is shown that when the region occupied by the spherical particles represents an ellipsoid of revolution and the temperature gradient away from this region tends to a given constant value, the effective thermal conductivity becomes independent of the ratio of the ellipsoid semiaxes, i.e. independent of the form of the region occupied by the system.

The effective thermal conductivity of a homogeneous suspension was studied earlier in /1-7/. Maxwell calculated the effective electrical conductivity of a mixture to terms of the order of the volume concentration of the spherical inclusions. The effective thermal conductivity is easily calculated to the same approximation, since the problems of determining the thermal and electrical conductivity are mathematically equivalent. The same problem is encountered in computing the dielectric permeability of two-phase mixtures /8/ and in determining the effective shear modulus of a homogeneous and isotropic composite material /9, 10/.

A cell model was used in /2-5/ to compute the effective thermal and electrical conductivity of suspensions at moderate and high particle densities. It was assumed that the particle was situated at the centre of a spherical cell, and the medium outside it possessed the required effective thermal conductivity. The drawback of this method lies in the arbitrariness of the choice of the cell boundary. A method of calculating the effective thermal conductivity of the media with spherical inclusions situated at the nodes of various types of cubic lattices at moderate particle densities was given in /6/, where a review of the earlier investigations concerned with computing the thermal conductivity in analogous media at low volume densities was also given. The effective thermal conductivity of a homogeneous suspension with randomly distributed particles was calculated to terms of the order of the square of the particle density in /7/, using the method given earlier in /11/.

1. Formulation of the problem. Let a region of volume  $V$  containing  $N$  identical spherical particles of constant thermal conductivity  $\kappa' \neq \kappa$  exist in an infinite medium filled with a material of constant thermal conductivity  $\kappa$ . We assume that away from  $V$  a steady temperature distribution is given with constant gradient  $k$ . The temperature field  $T$  will depend, at any point  $r$ , on the position of the particle centres determined by the radius vectors  $r_1, \dots, r_N$ . We shall denote the complete set of these radius vectors by  $R_N$ . We will introduce an unconditional correlation function  $f_N(R_N)$  such that

$$\frac{1}{V^N} \int f_N(R_N) dR_N$$

denotes the probability of finding the particle centres, respectively, within the small volumes  $d^3r_1, \dots, d^3r_N$  beside the points  $r_1, \dots, r_N$ . We introduce the conditional correlation function  $f_{N-1}(R_{N-1}; r_N)$  defined in such a manner that

$$\frac{1}{V^{N-1}} \int f_{N-1}(R_{N-1}; r_N) dR_{N-1}$$

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denotes the probability of finding the centres of  $N - 1$  particles within the volumes  $d^3r_1, \dots, d^3r_{N-1}$  beside the points  $r_1, \dots, r_{N-1}$  respectively, provided that the centre of the  $N$ -th particle is at the point  $r_N$ .

In accordance with the above definitions, the correlation functions have the following properties:

$$\begin{aligned} \frac{1}{V^N} \int f_N(R_N) dR_N &= 1, \\ \frac{1}{V^{N-1}} \int f_{N-1}(R_{N-1}; r_N) dR_{N-1} &= 1 \\ f_N(R_N) &= f_{N-1}(R_{N-1}; r_N) f_1(r_N) \\ f_{N-1}(R_{N-1}; r_N) &= f_{N-2}(R_{N-2}; r_{N-1}, r_N) f_1(r_{N-1}, r_N) \end{aligned}$$

and  $f_N(R_N), f_{N-1}(R_{N-1}; r_N), f_{N-2}(R_{N-2}; r_{N-1}, r_N)$  are symmetric functions of the arguments  $R_N, R_{N-1}$  and  $R_{N-2}$  respectively. The mean number of particles per unit volume  $n(r)$  is connected with the one-particle function by the relation

$$n(r) = (N/V) f_1(r)$$

For the spatially homogeneous distribution we have

$$f_1(r) = \begin{cases} 1, & r \in V \\ 0, & r \notin V \end{cases}$$

The mean temperature gradient (over the ensemble) and mean heat flux in the system have the form

$$\begin{aligned} G(r) &= \frac{1}{V^N} \int f_N(R_N) \nabla T'(r, R_N) dR_N \\ F(r) &= \frac{1}{V^N} \int f_N(R_N) \bar{\kappa} \nabla T'(r, R_N) dR_N \\ \bar{\kappa} &= \kappa' \theta(r, R_N) + \kappa [1 - \theta(r, R_N)] \\ \theta(r, R_N) &= \sum_{i=1}^N \eta(a - |\mathbf{r} - \mathbf{r}_i|), \quad \eta(x) = \begin{cases} 1, & x > 0 \\ 0, & x < 0 \end{cases} \end{aligned} \quad (1.1)$$

The tensor  $\kappa_*^{\alpha\beta}$  connecting the components of the averaged vectors of the heat flux  $F$  and the temperature gradient  $G$

$$F^\alpha = \sum_{\beta=1}^3 \kappa_*^{\alpha\beta} G^\beta$$

is called the tensor of effective thermal conductivity of the suspension. Generally speaking, the tensor  $\kappa_*^{\alpha\beta}$  is not spherical /12/ but, as follows from the results of /7/ and of the present paper, in the case of spatially homogeneous system with randomly distributed inclusions the effective thermal conductivity is a scalar quantity:  $\kappa_*^{\alpha\beta} = \kappa_* \delta^{\alpha\beta}$ . Below we shall compute  $\kappa_*$  to terms of the order of  $c^2$  inclusive ( $c = 4/3 \pi a^3 N/V$  is the volume density of the particles). We establish that, although the vectors  $F$  and  $G$  depend on the form of the region occupied by the suspension, the effective heat conductivity  $\kappa_*$  within the approximation stated can be determined in terms of  $\kappa, \kappa'$  and  $c$  only. From (1.1) it follows that

$$F - \kappa G = \frac{\kappa' - \kappa}{V^N} \sum_{i=1}^N \int f_N(R_N) \eta(a - |\mathbf{r} - \mathbf{r}_i|) \nabla T'(r, R_N) dR_N$$

Here and henceforth  $T'$  will denote the temperature at the point  $r$  within the particle. If the observer point  $r$  is situated outside the region occupied by the particles, then the temperature will be denoted by  $T$ .

Using the properties of the correlation functions, we can obtain

$$F - \kappa G = N \frac{\kappa' - \kappa}{V^N} \int dR_{N-1} \int_{|\mathbf{r} - \mathbf{r}_N| < a} f_1(r_N) \times f_{N-1}(R_{N-1}; r_N) \nabla T'(r, R_N) d^3r_N \quad (1.2)$$

and a formula analogous to (1.2) was obtained earlier in /7/.

Let us place the coordinate origin at the centre of the  $N$ -th sphere, and denote by  $R'_{N-1}$  the set of vectors  $r'_1, \dots, r'_{N-1}$  where  $r'_i = r_i - r_N$ , and the difference  $r - r_N$  by  $x_N$ .

If the particle density distribution in the system is almost spatially homogeneous, then we can restrict ourselves to the expansion

$$f_1(\mathbf{r}_N) = f_1(\mathbf{r} - \mathbf{x}_N) \cong f_1(\mathbf{r}) - \mathbf{x}_N \nabla f_1(\mathbf{r}) + \sum_{\alpha, \beta=1}^3 \frac{1}{2} x_N^\alpha x_N^\beta \frac{\partial^2}{\partial r^\alpha \partial r^\beta} f_1(\mathbf{r})$$

The temperature gradient at some point of the space fixed relative to position of the particles, is obviously independent of the choice of the origin of coordinates, therefore we have

$$\nabla T'(\mathbf{r}, R_N) = \frac{\partial}{\partial \mathbf{x}_N} T'(\mathbf{x}_N, R'_{N-1})$$

With reference to the function  $f_{N-1}$ , it is assumed that it is determined by the relative positions of the particles themselves, i.e.

$$f_{N-1}(R_{N-1}; \mathbf{r}_N) = f_{N-1}(R'_{N-1}; 0)$$

Then relation (1.2) can be written in the form

$$\begin{aligned} \mathbf{F} - \kappa \mathbf{G} = N \frac{\kappa' - \kappa}{V_N} \int_{x_N < a} dR'_{N-1} \int [f_1(\mathbf{r}) - \\ \sum_{\alpha=1}^3 x_N^\alpha \frac{\partial f_1(\mathbf{r})}{\partial r^\alpha} + \frac{1}{2} \sum_{\alpha, \beta=1}^3 x_N^\alpha x_N^\beta \frac{\partial^2 f_1(\mathbf{r})}{\partial r^\alpha \partial r^\beta}] \times \\ f_{N-1}(R'_{N-1}; 0) \frac{\partial}{\partial \mathbf{x}_N} T'(\mathbf{x}_N, R'_{N-1}) d^3 \mathbf{x}_N \end{aligned} \quad (1.3)$$

The temperature distribution is found from the solution of the following problem: ( $\mathbf{n}_i$  is the unit normal to the surface of the  $i$ -th sphere)

$$\begin{aligned} \Delta T = 0, \quad \Delta T_i' = 0 \quad (i = 1, \dots, N) \\ T = T_i', \quad \kappa \frac{\partial T}{\partial \mathbf{n}_i} = \kappa' \frac{\partial T_i'}{\partial \mathbf{n}_i}, \quad |\mathbf{r} - \mathbf{r}_i| = a \\ \nabla T \rightarrow \mathbf{k}, \quad |\mathbf{r}| \rightarrow \infty, \quad (|\mathbf{k}| = 1) \end{aligned} \quad (1.4)$$

The temperature is a harmonic function and can be represented (apart from an arbitrary constant omitted from the expression given below), in the neighbourhood and within the volume of the  $i$ -th particle, in the form of a series in terms of the spherical volume functions

$$\begin{aligned} T = (\mathbf{k} \cdot \mathbf{r}_i) + (\mathbf{k} \cdot \mathbf{x}_i) + \sum_{p=1}^{\infty} D_i^{\alpha_1 \dots \alpha_p} \Lambda_i^{\alpha_1 \dots \alpha_p} + \sum_{j \neq i} \sum_{p=1}^{\infty} D_j^{\alpha_1 \dots \alpha_p} \Lambda_j^{\alpha_1 \dots \alpha_p} \\ T_i' = (\mathbf{k} \cdot \mathbf{r}_i) + (\mathbf{k} \cdot \mathbf{x}_i) + \sum_{p=1}^{\infty} D_i^{\alpha_1 \dots \alpha_p} \left(\frac{x_i}{a}\right)^{2p+1} \Lambda_i^{\alpha_1 \dots \alpha_p} + \\ \sum_{j \neq i} \sum_{p=1}^{\infty} D_j^{\alpha_1 \dots \alpha_p} \Lambda_j^{\alpha_1 \dots \alpha_p} \\ \left( \Lambda_i^{\alpha_1 \dots \alpha_p} = \frac{\partial^p}{\partial x_i^{\alpha_1} \dots \partial x_i^{\alpha_p}} \frac{1}{x_i}, \quad \mathbf{x}_i = \mathbf{r} - \mathbf{r}_i, \quad x_i = |\mathbf{x}_i| \right) \end{aligned} \quad (1.5)$$

Here  $x_i^\alpha$  are Cartesian coordinates of the vector  $\mathbf{x}_i$ ,  $D_i^{\alpha_1 \dots \alpha_p}$  are multipolar moments of the  $i$ -th particle are to be determined. The repeated Greek indices  $\alpha_1, \dots, \alpha_p$  in (1.5) and further expressions, taking the value of 1, 2, 3, denote summation. Representing the solution of problem (1.4) in the form (1.5), ensures that the temperatures  $T$  and  $T_i'$  are equal at the surface of the  $i$ -th particle.

We note that the expansions (1.5) deviate from the standard series in spherical functions. As we know [13], for fixed  $p$ ,  $3^p$  functions  $\Lambda_i^{\alpha_1 \dots \alpha_p}$  contain only  $2p + 1$  linearly independent functions. To avoid any ambiguity in the determination of the multipolar moments  $D_i^{\alpha_1 \dots \alpha_p}$ ,

we assume below that they satisfy two conditions: 1)  $D_i^{\alpha_1 \dots \alpha_p}$  are symmetric over any pair of upper indices; 2) the contraction  $D_i^{\alpha_1 \dots \alpha_p - 2\beta\beta} = 0$ . It can be shown that when the above conditions hold, the moments are defined uniquely.

2. Determination of dipole moment. To determine the multipole moments  $D_i^{\alpha_1 \dots \alpha_p}$  we require that the functions (1.5) satisfy the condition of continuity of the normal heat flux component at the surface of the  $i$ -th particle. With this in mind, we use the analyticity of harmonic functions and expand  $\Lambda_j^{\alpha_1 \dots \alpha_p}$  near the  $i$ -th particle in a series in powers of  $x_i^\alpha$ :

$$\Lambda_j^{\alpha_1 \dots \alpha_p} = \Lambda_{ij}^{\alpha_1 \dots \alpha_p} + \sum_{q=1}^{\infty} \frac{1}{q!} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} x_i^{\beta_1} \dots x_i^{\beta_q} \tag{2.1}$$

$$\left( \Lambda_{ij}^{\nu_1 \dots \nu_q} = \frac{\partial^q}{\partial r_{ij}^{\nu_1} \dots \partial r_{ij}^{\nu_q}} \frac{1}{r_{ij}}, \quad r_{ij} = r_i - r_j, \quad r_{ij} = |r_{ij}| \right)$$

Next we take into account the fact that the convolution of the tensor  $\Lambda_i^{\beta_1 \dots \beta_q}$  over any two upper indices is equal to zero and, that  $\Lambda_i^{\beta_1 \dots \beta_q}$  has the form

$$\Lambda_i^{\beta_1 \dots \beta_q} = (-1)^q \frac{(2q-1)!!}{x_i^{2q+1}} x_i^{\beta_1} \dots x_i^{\beta_q} + \dots \tag{2.2}$$

where the terms omitted contain at least one Kronecker delta. The temperature  $T$  near the  $i$ -th particle can be written, taking (2.2) into account, as follows:

$$T = (\mathbf{k} \cdot \mathbf{r}_i) + (\mathbf{k} \cdot \mathbf{x}_i) + \sum_{j \neq i} \sum_{p=1}^{\infty} D_j^{\alpha_1 \dots \alpha_p} \Lambda_{ij}^{\alpha_1 \dots \alpha_p} + \sum_{q=1}^{\infty} \frac{(-1)^q x_i^{2q+1}}{q!(2q-1)!!} \sum_{j \neq i} \sum_{p=1}^{\infty} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} \times D_j^{\alpha_1 \dots \alpha_p} \Lambda_i^{\beta_1 \dots \beta_q} + \sum_{p=1}^{\infty} D_i^{\alpha_1 \dots \alpha_p} \Lambda_i^{\alpha_1 \dots \alpha_p} \tag{2.3}$$

Using the formulas

$$n_i^\alpha \frac{\partial}{\partial x_i^\alpha} \Lambda_i^{\beta_1 \dots \beta_q} = -\frac{q+1}{x_i} \Lambda_i^{\beta_1 \dots \beta_q}$$

$$n_i^\alpha \frac{\partial}{\partial x_i^\alpha} x_i^{2q+1} \Lambda_i^{\beta_1 \dots \beta_q} = q x_i^{2q} \Lambda_i^{\beta_1 \dots \beta_q}$$

which follow from the homogeneity of the  $\Lambda$ -functions, we can obtain the following infinite system of equations connected the multipolar moments of the particles in the system:

$$D_i^{\beta_1 \dots \beta_q} = (-1)^{q+1} \lambda_q a^{2q+1} (\delta^{q1} k^{\beta_1} + \sum_{j \neq i} \sum_{p=1}^{\infty} D_j^{\alpha_1 \dots \alpha_p} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q}) \tag{2.4}$$

$$\lambda_q = \frac{1}{(q-1)!(2q-1)!!} \frac{\kappa' - \kappa}{q\kappa' + (q+1)\kappa}, \quad q = 1, 2, \dots$$

Brown in /14/ studied an analogous problem, restricting himself to the dipole terms and neglecting the influence of the higher-order multipoles on the magnitude of the dipole moments.

If  $|\lambda_1| \ll 1$ , then the solution of (2.4) can be obtained by an iterative method. When carrying out the iterations the following type sums will appear:

$$\sum_{j \neq i}, \sum_{j \neq i} \sum_{n \neq j}, \sum_{j \neq i} \sum_{m \neq j} \sum_{n \neq m}, \dots$$

It should be noted that in averaging the double sums over  $j$  and  $n$  with the help of the conditional correlation function  $f_{N-1}(R_{N-i}, \mathbf{r}_i)$ , we obtain terms proportional to  $c^3$  provided that  $n \neq i$ . Therefore, if we limit ourselves, in the course of computing the multipole moments, to terms of the order of  $c$ , then only the terms  $i = n$  will have to be retained. An analogous situation arises when multiple sums are averaged. Thus in averaging the triple sums over  $j, m, n$ , we must limit ourselves to terms with  $m = i, n = j$  only. Taking all this into account and carrying out the iterations, we obtain

$$D_i^{\beta_1 \dots \beta_q} = \lambda_1 a^2 k^{\beta_1} \delta^{1q} - (-1)^q \lambda_1 \lambda_q \times a^{2q+4} k^\alpha \sum_{j \neq i} [\Lambda_{ij}^{\beta_1 \dots \beta_q \alpha} + \sum_{n=1}^{\infty} M_n^{\beta_1 \dots \beta_q \alpha}] \tag{2.5}$$

$$M_n^{\beta_1 \dots \beta_q \alpha} = \sum_{p_1=1}^{\infty} \dots \sum_{p_n=1}^{\infty} \lambda_{p_1} \dots \lambda_{p_n} a^{2(p_1+\dots+p_n)+n} \times \Lambda_{ij}^{\beta_1 \dots \beta_q (p_1)} \Lambda_{ij}^{(p_2) (p_1)} \dots \Lambda_{ij}^{(p_n) \alpha}$$

In deriving (2.5), we have used the symmetry of the function  $\Lambda_{ij}^{\alpha_1 \dots \alpha_n}$  to interchange of the upper indices and the fact that the function acquires the multiplier  $(-1)^n$  when the lower indices are interchanged. In addition, to reduce the length of the expressions for convolutions of the  $\Lambda$ -functions, we will use the following notation:

$$\Lambda_{ij}^{(p)(q)} \Lambda_{ij}^{(q)\alpha} = \Lambda_{ij}^{\nu_1 \dots \nu_p \varepsilon_1 \dots \varepsilon_q} \Lambda_{ij}^{\varepsilon_1 \dots \varepsilon_q \alpha}$$

To obtain the numerical results we must have a formula for convolution over the repeated upper Greek indices of the  $\Lambda$ -functions in the expression for  $M_n^{\beta_1 \dots \beta_q \alpha}$  in (2.5). With this

in mind, we shall utilize certain properties of the  $\Lambda$ -function. We have the following relation:

$$\Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} \Lambda_{ij}^{\beta_1 \dots \beta_q \mu} = - \frac{(p+q)! (2q-1)!!}{p! r_{ij}^{2q+1}} \left[ \frac{q}{p+1} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \mu} + \frac{2q+1}{r_{ij}} n_{ij}^{\mu} \Lambda_{ij}^{\alpha_1 \dots \alpha_p} \right] \left( n_{ij}^{\mu} = \frac{r_{ij}^{\mu}}{r_{ij}} \right) \quad (2.6)$$

To prove it, we write the second function in the form

$$\Lambda_{ij}^{\beta_1 \dots \beta_q \mu} = \frac{\partial}{\partial r_{ij}^{\mu}} \Lambda_{ij}^{\beta_1 \dots \beta_q} = \frac{\partial}{\partial r_{ij}^{\mu}} \left[ \frac{(-1)^q (2q-1)!!}{r_{ij}^{2q+1}} r_{ij}^{\beta_1} \dots r_{ij}^{\beta_q} + \dots \right] = (-1)^q \frac{(2q-1)!!}{r_{ij}^{2q+2}} \left[ \sum_{m=1}^q n_{ij}^{\beta_m} \dots \delta^{\beta_m \mu} \dots n_{ij}^{\beta_q} - (2q+1) n_{ij}^{\beta_1} \dots n_{ij}^{\beta_q} n_{ij}^{\mu} + \dots \right]$$

Terms omitted contain at least one Kronecker delta and offer no contribution to convolution with the first function of (2.6). We can further show that the following relation holds:

$$n_{ij}^{\mu} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \mu} = - \frac{p+1}{r_{ij}} \Lambda_{ij}^{\alpha_1 \dots \alpha_p}$$

Consecutive application of this formula yields

$$n_{ij}^{\alpha_1} \dots n_{ij}^{\alpha_p} \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} = \frac{(-1)^p (p+q)!}{q! r_{ij}^p} \Lambda_{ij}^{\beta_1 \dots \beta_q}$$

and we have

$$\Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} \Lambda_{ij}^{\beta_1 \dots \beta_q \mu} = \frac{(-1)^q (2q-1)!!}{r_{ij}^{2q+2}} \times [q \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_{q-1} \mu} n_{ij}^{\beta_q} \dots n_{ij}^{\beta_{q-1}} - (2q+1) \Lambda_{ij}^{\alpha_1 \dots \alpha_p \beta_1 \dots \beta_q} n_{ij}^{\mu} \dots n_{ij}^{\beta_q} n_{ij}^{\mu}] = - \frac{(p+q)! (2q-1)!!}{r_{ij}^{2q+1} (p+1)!} \left[ q \Lambda_{ij}^{\alpha_1 \dots \alpha_p \mu} + (p+1) (2q+1) \frac{1}{r_{ij}} \Lambda_{ij}^{\alpha_1 \dots \alpha_p} n_{ij}^{\mu} \right]$$

from which (2.6) follows. The following relations also hold:

$$\begin{aligned} \Lambda_{ij}^{\beta_1 \dots \beta_p} \Lambda_{ij}^{\beta_1 \dots \beta_p} &= \frac{p! (2p-1)!!}{r_{ij}^{2p+2}} \quad (2.7) \\ \Lambda_{ij}^{\alpha \beta_1 \dots \beta_p} \Lambda_{ij}^{\beta_1 \dots \beta_p} &= - \frac{(p+1)! (2p-1)!!}{r_{ij}^{2p+2}} n_{ij}^{\alpha} \\ \Lambda_{ij}^{\alpha \beta \beta_1 \dots \beta_p} \Lambda_{ij}^{\beta_1 \dots \beta_p} &= \frac{(p+2)! (2p-1)!!}{2 r_{ij}^{2p+1}} \Lambda_{ij}^{\alpha \beta} \end{aligned}$$

and are proved in a similar way.

Below we shall need an expression for the dipole moment  $D_i^{\beta}$  which can be found from (2.5) with  $q=1$ . Using (2.6) and (2.7), we can obtain the following fundamental formula which yields the result of convolution of an arbitrary number of  $\Lambda$ -functions:

$$\begin{aligned} \Lambda_{ij}^{\alpha(p_1)} \Lambda_{ij}^{(p_2)(p_3)} \dots \Lambda_{ij}^{(p_n)\beta} &= \quad (2.8) \\ \frac{(p_1+p_2) \dots (p_{n-1}+p_n)! (2p_1-1)!! \dots (2p_n-1)!!}{2(p_1+1) \dots (p_{n-1}+1)! r_{ij}^{2(p_1+\dots+p_n)+n+2}} \times \\ & \left( (-1)^{n+1} p_1 \dots p_n \delta^{\alpha\beta} + [(-1)^n p_1 \dots p_n + 2(p_1+1) \dots (p_n+1)] n_{ij}^{\alpha} n_{ij}^{\beta} \right) \end{aligned}$$

3. Calculation of the effective thermal conductivity. The results obtained enable us to determine the integrals over the volume of the  $N$ -th particle appearing in (1.3). Indeed, using expansion (2.3) near the particle with index  $i=N$  and taking into account the mutual orthogonality of the functions  $\Lambda_N^{\beta_1 \dots \beta_p}$  with different numbers of upper indices, we can obtain

$$\begin{aligned} \int_{x_N \leq a} \frac{\partial T_N'}{\partial x_N^{\alpha}} d^3 x_N &= \oint_{x_N=a} n_N^{\alpha} T_N dS = \oint_{x_N=a} n_N^{\alpha} \left[ (\mathbf{k} \cdot \mathbf{x}_N) - \right. \\ & a^3 \sum_{j=N} \sum_{p=1}^{\infty} \Lambda_{Nj}^{\alpha_1 \dots \alpha_p \beta} D_j^{\alpha_1 \dots \alpha_p} \Lambda_N^{\beta} + D_N^{\beta} \Lambda_N^{\beta} \left. \right] dS = \\ & \frac{4}{3} \pi a^3 \left[ k^{\alpha} + \sum_{j=N} \sum_{p=1}^{\infty} D_j^{\alpha_1 \dots \alpha_p} \Lambda_{Nj}^{\alpha_1 \dots \alpha_p \alpha} - \frac{1}{a^3} D_N^{\alpha} \right] \end{aligned}$$

Further, using (2.4) we can obtain

$$\int_{x_N \leq a} \frac{\partial T_{N'}'}{\partial x_N^\alpha} d^3 x_N = -\frac{4\pi}{3} \left( \frac{1}{\lambda_1} - 1 \right) D_N^\alpha$$

We can evaluate the following integrals by similar methods:

$$\begin{aligned} \int_{x_N \leq a} x_N^\alpha \frac{\partial T_{N'}'}{\partial x_N^\beta} d^3 x_N &= \frac{8\pi}{5} \left( 1 - \frac{1}{6\lambda_2} \right) D_N^{\alpha\beta} \\ \int_{x_N \leq a} x_N^\alpha x_N^\beta \frac{\partial T_{N'}'}{\partial x_N^\gamma} d^3 x_N &= a^2 \oint_{x_N=a} n_N^\alpha n_N^\beta n_N^\gamma T_N dS - \\ &\int_{x_N \leq a} x_N (\delta^{\alpha\gamma} n_N^\beta + \delta^{\beta\gamma} n_N^\alpha) T_N' d^3 x_N = \\ &a^2 \oint_{x_N=a} \left[ n_N^\alpha n_N^\beta n_N^\gamma - \frac{1}{5} (\delta^{\alpha\gamma} n_N^\beta + n_N^\alpha \delta^{\beta\gamma}) \right] T dS = \\ &-\frac{8\pi}{45} a^2 \left( \frac{1}{\lambda_1} - 1 \right) \delta^{\alpha\beta} D_N^\gamma + \dots \end{aligned}$$

In evaluating the last integral, higher-order terms proportional to  $D_N^{\alpha\beta\gamma}$  were omitted. Substituting these expressions into (1.3), we arrive at the following result:

$$\begin{aligned} F^\alpha - \kappa G^\alpha &= \frac{4\pi}{3} \frac{N}{V} (\kappa' - \kappa) \left[ \left( \frac{1}{\lambda_1} - 1 \right) \left( f_1 - \frac{a^2}{15} \Delta f_1 \right) \langle D_N^\alpha \rangle - \right. \\ &\left. \frac{2}{5} \left( 3 - \frac{1}{2\lambda_2} \right) \frac{\partial f_1}{\partial r^\beta} \langle D_N^{\alpha\beta} \rangle + \dots \right] \\ \langle D_N^{\alpha_1 \dots \alpha_p} \rangle &= \frac{1}{V^{N-1}} \int dR_{N-1} f_{N-1}(R_{N-1}; 0) D_N^{\alpha_1 \dots \alpha_p}(R_{N-1}) \end{aligned} \tag{3.1}$$

In computing the averages  $\langle D_N^\alpha \rangle$  and  $\langle D_N^{\alpha\beta} \rangle$  to terms of the order of the mean volume density  $c = 4/3 \pi N a^3 / V$  inclusive, we will use the representation of the correlation function  $f_1(\mathbf{r}; 0)$  in the form

$$f_1(\mathbf{r}; 0) = \eta (|\mathbf{r}| - 2a) f_1(\mathbf{r})$$

Let us further limit ourselves to the case of a spatially homogeneous distribution. Then the averaging and use of (2.5), (2.8) yields

$$\langle D_N^\alpha \rangle = \lambda_1 a^3 [k^\alpha + \lambda_1 a^3 k^\beta \langle \sum_{j \neq N} \Lambda_{Nj}^{\alpha\beta} \rangle + c \lambda_1 k^\alpha B], \quad B = \sum_{n=1}^{\infty} b_n \tag{3.2}$$

$$\begin{aligned} b_1 &= \frac{1}{2} \sum_{p=1}^{\infty} \frac{p(p+1)}{4^p} \frac{\kappa' - \kappa}{p\kappa' + (p+1)\kappa} \\ b_2 &= \frac{1}{8} \sum_{p=1}^{\infty} \sum_{q=1}^{\infty} \frac{(p+q)!}{4^{p+q} (p-1)! (q-1)!} \frac{(\kappa' - \kappa)^2}{[p\kappa' + (p+1)\kappa][q\kappa' + (q+1)\kappa]} \\ b_n &= \sum_{p_1=1}^{\infty} \dots \sum_{p_n=1}^{\infty} \frac{(p_1+p_2)! \dots (p_{n-1}+p_n)!}{(p_1+1)! \dots (p_{n-1}+1)!} \times \\ &\frac{\lambda_{p_1} \dots \lambda_{p_n} (2p_1-1)! \dots (2p_n-1)!}{[2(p_1+\dots+p_n)+n] 2^{n(p_1+\dots+p_n)+n}} \times \\ &[(p_1+1) \dots (p_n+1) - (-1)^n p_1 \dots p_n], \quad n \geq 3 \end{aligned}$$

Using the properties of the correlation functions, we can reduce the process of computing the averages of the sum  $\Lambda_{Nj}^{\alpha\beta}$  to computing the integral over the volume  $V$ , excluding the region  $|\mathbf{r}_{Nj}| \leq 2a$

$$\left\langle \sum_{j \neq N} \Lambda_{Nj}^{\alpha\beta} \right\rangle = \frac{N}{V} \int d^3 \mathbf{r}_{Nj} \Lambda_{Nj}^{\alpha\beta} \tag{3.3}$$

When the region occupied by the particles with constant volume density  $c$  is an ellipsoid, evaluating the last integral is equivalent to solving the well-known problem of electrostatics, of calculating the depolarizing field  $/15/$ . If two semiaxes of the ellipsoid are equal while the third one represents the axis of symmetry and is directed along the vector  $\mathbf{k}$ , then

$$k^\beta \left\langle \sum_{j \neq N} \Lambda_{Nj}^{\alpha\beta} \right\rangle = \frac{N}{V} \frac{4\pi}{3} (1 - K_D) k^\alpha$$

Here  $4/3\pi K_D$  is the depolarisation coefficient equal, in particular, to  $4/3\pi$  for a sphere, to  $4\pi$  for a thin plate, and to 0 for a circular cylinder.

There effective thermal conductivity is obtained in /7/ for an infinite medium. Conditional convergence of the integrals representing the mean dipole moment means that the computations can be reduced, in accordance with the method described in /4/, to calculating the finite difference between two, conditionally converging integrals. In the present paper the problem does not arise, since the particles are distributed through a finite volume of the space.

It can be shown that we have the following expression for the homogeneous and isotropic distribution apart from terms of order  $O(c)$ :

$$\langle D_N^{\alpha\beta} \rangle = 0 \quad (3.4)$$

We prove this relation using the formula (2.5). We see that the quantities  $M_n^{\beta\alpha\beta\alpha}$  in (2.5) have the form

$$M_n^{\beta\alpha\beta\alpha} = A_1(n) \Lambda_{ij}^{\beta\alpha\beta\alpha} + A_2(n) n_{i,j} \Lambda_{ij}^{\beta\alpha\beta\alpha}$$

and integration over the angular part yields, by virtue of the orthogonality of the  $\Lambda$ -functions with the same indices, (3.4).

To compute the effective thermal conductivity we require, in addition to (3.1), an expression for the mean temperature gradient in the system. If the region occupied by the particles represents an ellipsoid, as was assumed when calculating the dipole moment, then

$$\mathbf{G}(\mathbf{r}) = \mathbf{k} - \frac{4\pi}{3} \frac{N}{V} K_D \langle \mathbf{D}_N \rangle = \mathbf{k} - c\lambda_1 K_D [1 + c\lambda_1(1 - K_D + B)] \mathbf{k} \quad (3.5)$$

Substituting (3.5) into (3.1) and taking into account (3.2) and (3.3), we arrive at the following expression for the three-dimensional homogeneous distribution corresponding to the case  $f_1 \equiv 1$ :

$$\mathbf{F} = \kappa_* \mathbf{G} = \kappa \mathbf{G} - c(\lambda_1 - 1)(\kappa' - \kappa)[1 + c\lambda_1(1 - K_D + B)] \mathbf{k}$$

Thus the effective thermal conductivity, up to and including terms of order  $c^2$ , is equal to

$$\kappa_* = \kappa - c(\kappa' - \kappa)(\lambda_1 - 1) - c^2\lambda_1(\kappa' - \kappa)(\lambda_1 - 1)(1 + B)$$

The above results show that the effective thermal conductivity is independent of the form of the region occupied by the particles up to and including terms of order  $c^2$ , although the temperature and heat flux averaged over the ensemble depend not only on the volume density, but also on the form of the region occupied by the suspension.

The effective thermal conductivity is identical with the Maxwell formula to terms proportional to  $c$ . Below we give the results of calculations of the coefficient accompanying  $c^2$  in the expression  $\kappa_*/\kappa$  (the number on the left corresponds to the number of first terms taken into account in B)

$\kappa'/\kappa = 0$	0.1	0.5	2.0	5.0	50	$\infty$
1	0.583	0.447	0.110	0.207	1.20	3.64 4.17
3	0.587	0.450	0.110	0.208	1.23	3.82 4.39
5	0.588	0.450	0.110	0.208	1.23	3.86 4.45

The results of the computations carried out earlier in /7/ by another method agree with the values given in the last column except for the last two values (according to /7/ those values are equal 3.90 and 4.51 respectively).

It is clear that the series for determining the coefficient of  $c^2$  converges fairly rapidly except in the case when  $\kappa' \gg \kappa$ . Restricting ourselves to two terms of the series only, we find that the error in the value of the coefficient of  $c^2$  does not exceed 5% for any value of the ratio of  $\kappa'$  and  $\kappa$ .

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## EFFECT OF HYDRODYNAMIC INTERACTIONS BETWEEN THE PARTICLES ON THE RHEOLOGICAL PROPERTIES OF DILUTE EMULSIONS \*

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The form of mean stress tensor in monodisperse emulsions is studied in the second-order approximation with respect to the volume density of the particles, for a number of flows which are of rheological interest. It is shown how the particular features of the two-particle interaction between liquid spheres, especially the non-zero differences between the normal stresses in shear flows, give rise to non-Newtonian properties of the emulsion.

We know /1, 2/ that in the case of the second-order approximation with respect to the volume concentration of the suspended disperse phase the mean stress tensor is expressed in terms of two-particle interactions in a linear velocity field, and of the binary correlation function. The binary function is formed under the action of the macroscopic flow. Specific results, however, were obtained only for suspensions and rigid spheres /1, 2/. The present paper deals with the structural model of fluid spheres of equal radius, with hydrodynamic and "contact" interactions. A number of fundamental deviations from /1/ exist in the case of rheologically strong flows, since drop flocculation-deflocculation processes must be considered (i.e. the formation and disruption of aggregates). A strict analysis is given within the framework of the model, of the effect of these processes on the binary correlation function. A connection between the model of "contact" interaction and the result of the D.L.V.O. theory /3-5/ is considered. Numerical values are obtained for the Trouton viscosity in strong rheologically axisymmetric expanding flows. The differences in normal stresses in a strong shear flow are obtained and an approximate estimate is given for the shear viscosity and compared with experimental data /6/. A method given in /2/ is used to compute the effective viscosity of the emulsion in arbitrary, rheologically weak flows in which Brownian motion predominates. Considerable use is made of the exact computational methods and asymptotic representations of hydrodynamic functions determining the pairwise interaction of fluid spheres /7-9/.

1. A general expression for the mean stress tensor. Consider a locally homogeneous monodisperse emulsion of drops of radius  $a$  and viscosity  $\mu'$  freely suspended in a

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