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THE PHYSICAL SIGNIFICANCE OF NON-CONVERGENT INTEGRALS IN EXPRESSIONS FOR EFFECTIVE TRANSPORT PROPERTIES

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Introduction

The idea of assigning effective properties to an inhomogeneous medium, for example a composite material or a suspension of particles in fluid, is well known [4, 9, 14], but some methods used to calculate these effective properties have been sources of controversy over the years. In particular, the appearance of non-convergent integrals (or sums) in several calculational schemes has generated many discussions on the relative merits of the different ways of calculating effective properties. Unfortunately these discussions remain largely unappreciated, with the result that new-comers to the field usually reinterpret for themselves the non-convergence, to the despair of those who have heard it all before. This paper presents a discussion of the significance of the non-convergent integrals which may help new workers understand the existing literature more easily. There is space here for only the briefest descriptions of the original papers, and consequently the details in this article will be intelligible only to those who have read or will read the original papers. For the general reader, the main conclusions of this paper are that

(1) long-range effects will produce non-convergent integrals in incorrectly formulated calculations of effective properties,

(2) these integrals can easily pass unnoticed or be assigned some non-unique finite value, and

(3) the correct formulation of calculations of effective properties is now known (and is described here).

Quite a few researchers still think that concern over non-convergent integrals is only a mathematical quibble, for the following seductive reasons. First, it has seemed at times in the past that all proposed schemes arrive at the same, presumably correct, answers and thus there has been no incentive, such as conflicting results would provide, to examine the various schemes critically. This paper shows, however, that cases do exist in which different schemes lead to conflicting results and that the prevailing idea that all methods give the same answers is a result of the particular selection of problems studied in the past. Second, those who have written on convergence difficulties have not had, until now, a sufficiently secure physical interpretation of the causes of non-convergence to be able to win over the unconvinced, who in the past seem to have been more numerous. I hope the weight of the arguments presented here will balance the weight of their numbers. Third, the fact that finite, but non-unique, values can be found for some of the non-convergent multiple integrals has strongly tempted many people to think that what is needed is some way of picking the 'correct' or 'physically significant' finite value. This has led to a narrow view of the problem. For a start, it is by no means always possible to find such finite values for the integrals in question. Here the view will be put forward that non-convergent integrals present a problem in interpretation and not just a problem in correct evaluation.

Although the discussion in this paper is confined to inhomogeneous media that consist of a particulate phase suspended in a continuous matrix phase, the ideas presented are valid in more general situations, and accordingly references to similar studies in related areas are given in the last section of this paper. If we denote by c the volume fraction of particles in a suspension or composite material, the calculations we shall discuss are those accurate to $O(c)$ or $O(c^2)$. The points which have not been discussed at length in print before and which will receive particular attention here are (a) the various possible averages of the 'applied field', (b) why non-convergent integrals arise and the incompleteness of early approaches, (c) nearest-neighbour and any-neighbour formulations, and (d) the characteristics of problems in which the interactions are too strong for the methods described here to be successful.

Non-convergent integrals in Einstein's work

Einstein's work [7, 8] was virtually the first on the subject. We start with it partly because his method is still sometimes used [1] and partly because the points arising from a consideration of his approach will recur when we examine later work. The reconstruction of Einstein's argument will obviously be coloured by present knowledge; however, we are not interested primarily in historical accuracy, but rather in a general framework for different approaches. Einstein calculated the effective viscosity of a suspension of spherical particles in a Newtonian fluid of viscosity μ correct to first order in the volume fraction of particles c. He proceeded by calculating the average rate of energy dissipation $\overline{W}$ in a suspension which is subjected to a uniform average rate of strain $\overline{e_{ij}}$. (Tensors will be indicated by subscripts or double underlining as convenient.) We have $\overline{W} = 2\mu\overline{e_{ij}e_{ij}}$ and

$$\overline{\underline{\underline{e}}} = V^{-1} \int \underline{\underline{e}} \, \mathrm{d}V \ ,$$

where V is the volume occupied by the suspension, and the expression for $\overline{W}$ uses the fact that inside a rigid particle $e_{ij} = 0$. Einstein actually expressed $\overline{W}$ as a surface integral, but the argument is much clearer in terms of volume integrals. We now substitute $\underline{\underline{e}} = \overline{\underline{\underline{e}}} + \underline{\underline{e}}'$ into the definition of W and obtain

$$\overline{W} = 2\mu\overline{e_{ij}}\ \overline{e_{ij}} + 4\mu\overline{e_{ij}}\ \overline{e'_{ij}} + 2\mu\overline{e'_{ij}e'_{ij}}\ .$$

Although it is clear that by definition $\overline{\underline{\underline{e}}'} = 0$, Einstein retained the second term because his expression for $\overline{W}$ in terms of surface integrals did not allow him to see this obvious simplification. To obtain an expression for $\underline{\underline{e}}$ in the neighbourhood of a particle, Einstein considered a subsidiary problem of flow around an isolated particle when there is a rate of strain $\overline{\underline{\underline{e}}}$ at infinity. Next he assumed that a volume integral over the suspension (i.e., over V) could be estimated by adding up independently volume integrals over regions V_0 which defined the region of influence of each particle on the flow, that is he assumed that for any quantity Z which tends to zero with increasing distance from a particle

$$\overline{Z} = V^{-1}\int Z\ \mathrm{d}V \approx V^{-1}\sum_{\text{all particles}}\int_{V_0} Z\ \mathrm{d}V = n\int_{V_0} Z\ \mathrm{d}V\ ,$$

where n is the number density of particles. The only restriction on the size of V_0 was that the volumes could not overlap. In making this assumption, Einstein apparently did not notice that $\underline{\underline{e}}'$ is $O(r^{-3})$ at large distances r from a particle and that the assumption led to an expression for $\overline{\underline{\underline{e}}'}$ in the form of a non-convergent integral. By taking V_0 to be a sphere and integrating first over angular co-ordinates, Einstein obtained a finite value for the integral of $\underline{\underline{e}}'$ (in the present formulation, zero from the fluid part of V_0 and $-c\overline{\underline{\underline{e}}}$ from the particle part); he also made several arithmetic errors in evaluating his integrals and obtained as an estimate for $\overline{W}$ the expression $2\mu\overline{e_{ij}}\;\overline{e_{ij}}(1-c)$ instead of $2\mu\overline{e_{ij}}\;\overline{e_{ij}}(1+\frac{1}{2}c)$. In a sentence omitted from the English translation [8], Einstein interpreted his incorrect expression for $\overline{W}$ as meaning that the only influence of the particles was to reduce by a factor $(1-c)$ the volume in which the rate of strain $\overline{\underline{\underline{e}}}$ dissipated energy. (Last sentence of [7, section 1]: Es ist bemerkenswert ... würde.) To understand the next steps in his argument, it is helpful to have in mind the picture shown in Figure 1. The region bounded by the surface Γ contains the suspension, i.e., it has volume V and we can identify it with Einstein's region G; the subregion bounded by Γ^* is a large sphere containing many particles and can be identified with Einstein's K. Einstein decided that his procedure for calculating $\overline{W}$ had given him not $\overline{W}$ but instead an average over the subregion bounded by Γ^*; we can denote this new average by W^* and write $W^* = 2\mu\overline{e_{ij}}\;\overline{e_{ij}}(1-c)$ if we retain the arithmetic error. Einstein argued that the average rate of strain over the subregion will be different from $\overline{\underline{\underline{e}}}$ and equal to some $\underline{\underline{e}}^*$ which can be calculated by using the same volumes V_0 as above. Thus he wrote

$$\underline{\underline{e}}^* = \overline{\underline{\underline{e}}} + n\int_{V_0} \underline{\underline{e}}'\,\mathrm{d}V = (1-c)\overline{\underline{\underline{e}}}\ .$$

The non-convergent integral in the definition of W^* reappears in the definition of $\underline{\underline{e}}^*$ and is given the same finite value. Eliminating $\overline{\underline{\underline{e}}}$ from the equations for W^* and $\underline{\underline{e}}^*$ gave $W^* = 2\mu e^*_{ij}e^*_{ij}(1+c)$ which became $W^* = 2\mu e^*_{ij}e^*_{ij}(1+\frac{5}{2}c)$ when the arithmetic error was corrected five years later. The effective viscosity finally became $\mu^{\mathrm{e}} = \mu(1+\frac{5}{2}c)$.

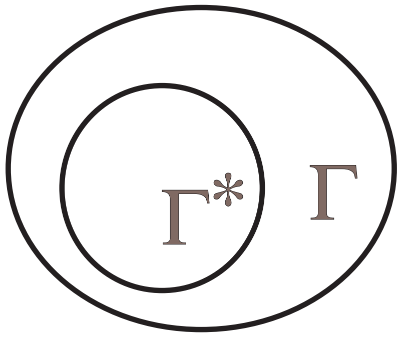

Figure 1: The two regions used in Einstein's calculation.

Saitô [25] and Mooney [19] noticed the non-convergent integral in the definition of $\underline{\underline{e}}^*$ but not the one in W^*; Saitô re-evaluated $\underline{\underline{e}}^*$ using a parallel-plate (as opposed to spherical) geometry but used Einstein's W^* unchanged and obtained a different result for the effective viscosity. It is possible to improve Einstein's procedure so that only convergent integrals appear and only averages over the full suspension bounded by Γ are used. We simply note that $\overline{\underline{\underline{e}}'} = 0$ and hence

$$\overline{W} = 2\mu\overline{e_{ij}}\,\overline{e_{ij}} + 2\mu\overline{e'_{ij}e'_{ij}}\ .$$

The quantity $e'_{ij}e'_{ij}$ is $O(r^{-6})$ far from a particle and the average can legitimately be approximated using Einstein's method. The result is

$$\overline{W} = 2\mu\overline{e_{ij}}\,\overline{e_{ij}}(1 + \frac{5}{2}c)\ .$$

Several of the themes of this paper appear in the above description. The distinction between the averages $\underline{\underline{e}}^*$ and $\overline{\underline{\underline{e}}}$ taken over different regions had been anticipated by Rayleigh [23] and was later used by Brown [5] and many others. The fact that non-convergent integrals occur in pairs and that one is effectively subtracted from the other is important because otherwise, as Saitô showed, the effective viscosity would depend upon the shape of the averaging volume. Because of the possibility of shape dependence, it is necessary to show that the result is independent of the shape and this is the advantage of the second method described which approximates only those quantities that can be expressed as convergent integrals. It should be noted finally that Einstein offered no proof that the error in his calculation was $O(c^2)$; we shall describe work later in which error estimates become crucial. It has been suggested in the past that Einstein's choice of a spherical shape for V_0 was based on this shape being 'physically significant'. It is clear, however, that any other shape, although giving different expressions for W^* and $\underline{\underline{e}}^*$ as functions of $\overline{\underline{\underline{e}}}$, would give the same expression for effective viscosity.

The examples to be studied

The advantages that are gained from viewing together all the transport problems in media with particulate structure have been explained by Batchelor [4]. For our present purposes the advantages are particularly marked because, quite simply, some problems are harder than others and the harder ones force on us a greater understanding of the easier ones. Our aim is to arrive at a single interpretation and method of solution that will apply to all problems and accordingly we here concentrate on two problems which have proved more difficult than the rest and which show up the inadequacies of simple expedients. We shall call the first problem the sedimentation problem; the quantity to be calculated is $\langle \boldsymbol{U} \rangle$ the velocity of sedimentation averaged over all particles in the suspension. The same force acts on each particle, and if we denote by $\boldsymbol{U}^{(0)}$ the velocity that corresponds to an isolated particle moving under this force, then the quantity we shall be devoting our attention to is $\langle \boldsymbol{U} - \boldsymbol{U}^{(0)} \rangle$.

The second problem is an effective-modulus problem and is the linearly elastic analogue of Einstein's calculation. We wish to relate the (volume) average stress $\overline{\sigma_{ij}}$ in a linearly elastic material to the average strain $\overline{e_{ij}}$ through effective moduli L^{e}_{ijkl} [6, 9, 10, 27]. Using the idea of polarization stress, due to Eshelby and Kröner [16], we have the stress at any point in the composite material given by

$$\sigma_{ij} = L^1_{ijkl} e_{kl} + \tau_{ij} \ ,$$

where L^1_{ijkl} is the tensor of elastic moduli of the matrix and $\underline{\underline{\tau}}$ is the polarization stress which is defined to be zero at any point in the matrix and $(L^2_{ijkl} - L^1_{ijkl})e_{kl}$ at a point in one of the inclusions, the inclusions having elastic moduli L^2_{ijkl} (more general definitions of $\underline{\underline{\tau}}$ are possible but this is the most convenient in this context). Since $\underline{\underline{\tau}}$ is non-zero only inside an inclusion, we can introduce a quantity $\underline{\underline{S}}$ [4, 6] which is defined for any inclusion by

$$\underline{\underline{S}} = \int_{\text{inclusion}} \underline{\underline{\tau}} \, \mathrm{d}V \ ,$$

where as indicated the integration is over the volume of an inclusion. Thus each inclusion has associated with it a value of $\underline{\underline{S}}$. Averaging the equation for σ_{ij} then gives

$$\overline{\sigma_{ij}} = L^1_{ijkl} \overline{e_{kl}} + \overline{\tau_{ij}} = L^1_{ijkl} \overline{e_{kl}} + n \langle \, S_{ij} \, \rangle \ .$$

Here n is the number density of inclusions and the angle brackets have been used to show that, to find the average value of $\underline{\underline{S}}$, we must use ensemble averaging over configurations of particles. The relation between $n\langle \, \underline{\underline{S}} \, \rangle$ and $\overline{\underline{\underline{e}}}$ will introduce the concentration tensors of Hill [10]. Having expressed $\overline{\underline{\underline{\sigma}}}$ in terms of $n\langle \, \underline{\underline{S}} \, \rangle$, we see that the statistical problem we are now faced with is analogous to the one facing us in the sedimentation problem. We can make the analogy more specific if we denote by $\underline{\underline{S}}^{(0)}$ the value of $\underline{\underline{S}}$

calculated for an inclusion placed alone in a matrix in which the strain 'at infinity' is $\bar{\underline{\underline{e}}}$. Then $\langle\, \underline{\underline{S}}\, \rangle \approx \underline{\underline{S}}^{(0)}$ corresponds to Einstein's approximation and we wish to calcu1ate $\langle\, \underline{\underline{S}} - \underline{\underline{S}}^{(0)}\, \rangle$.

The features of these two problems that make them of special interest are as follows. In the sedimentation problem the non-convergent integrals one is faced with are of the type $\int r^{-1}\, \mathrm{d}V$ which cannot be assigned any finite value at all no matter what geometry one tries. Questions of 'physically significant' volumes cannot, therefore, even be raised. Another consequence of the $O(r^{-1})$ integrand is that a naive nearest-neighbour approach (see next section) produces qualitatively different answers from an any-neighbour one and this shows dramatically that long-range interactions exist in the suspension in addition to interactions between neighbouring particles. The interest in the effective-modulus problem comes from ambiguities which appear in the subtraction method devised by Batchelor [3, 4] when the calculation is restricted to finding the compression modulus for a composite medium subjected to a simple compression.

Nearest neighbour or any neighbour?

The estimates $\underline{\underline{S}}^{(0)}$ for $\langle\, \underline{\underline{S}}\, \rangle$ and $\boldsymbol{U}^{(0)}$ for $\langle \boldsymbol{U} \rangle$ were obtained using the solutions of problems in which one particle was alone in an infinite matrix or fluid and it has long been supposed that improved estimates could be obtained by using solutions of problems involving two particles. In particular it has been assumed that estimates for $\langle\, \underline{\underline{S}} - \underline{\underline{S}}^{(0)}\, \rangle$ and $\langle\, \boldsymbol{U} - \boldsymbol{U}^{(0)}\, \rangle$ correct to $O(c)$ could be obtained from knowledge of two-particle interactions. In calculating these estimates, some authors [3, 4, 12, 13] have used an 'any-neighbour' probability density function, which we shall denote $P_A(\boldsymbol{r}|\boldsymbol{o})$, while others [18, 28] have used a 'nearest-neighbour' one $P_N(\boldsymbol{r}|\boldsymbol{o})$. For spherical particles these are defined as follows: we place the origin of a set of co-ordinates at the centre of the test particle and select a point $\boldsymbol{r}$; $P_A(\boldsymbol{r}|\boldsymbol{o})\mathrm{d}\boldsymbol{r}$ is then the probability that the centre of any particle of the suspension will be within the volume element $\mathrm{d}\boldsymbol{r}$ while $P_N(\boldsymbol{r}|\boldsymbol{o})\mathrm{d}\boldsymbol{r}$ is the probability that the centre of the nearest neighbour to the test particle will be within $\mathrm{d}\boldsymbol{r}$. When the point $\boldsymbol{r}$ is near the test particle the two functions are equal, but when it is far away P_N tends to zero while P_A tends to the number density n.

We use the sedimentation problem to show how the functions P_A or P_N arise in calculations. By definition, $\boldsymbol{U}-\boldsymbol{U}^{(0)}$ is the contribution to the velocity of sedimentation of the test particle due to the presence of other particles, and as such we expect particles close to the test particle to have greater influence on it than those further away. We might hope that an estimate of $\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle$ could be obtained by averaging the effect of just one other particle on the test particle and write

$$\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle \approx \int (\boldsymbol{U}-\boldsymbol{U}^{(0)}) P(\boldsymbol{r}|\boldsymbol{o}) d\boldsymbol{r} \ .$$

Since $\boldsymbol{U}-\boldsymbol{U}^{(0)}$ is for two particles an $O(r^{-1})$ quantity, it is important to know whether P_A or P_N is used in the integral. If P_A is used the integral is non-convergent, while if P_N is used the integral is convergent and gives an $O(c^{1/3})$ approximation for $\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle$. This last result is in conflict with work [3, 24] which finds an $O(c)$ result. The reason for the conflict is the incorrectness of the assumption that only local interactions between close particles affect $\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle$. Two arguments show us that long-range interactions are present in the suspension and affect $\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle$: (1) the very fact that it makes a difference to our answer whether we use P_A or P_N shows that other factors are important. The change from P_N to P_A can be regarded as a test of the assumption that only nearest neighbours contribute to $\langle\,\boldsymbol{U}-\boldsymbol{U}^{(0)}\,\rangle$. (2) Simple situations can be constructed in which conditions far from the test particle affect its velocity. For example, consider a cloud of sedimenting particles placed first in a container which it completely fills and secondly in one which has a substantial layer of clear fluid between the cloud and the walls. In the first case the fluid displaced as the particles sediment will have to flow through the cloud whereas in the second case it will flow around the cloud. The velocity $\langle \boldsymbol{U} - \boldsymbol{U}^{(0)} \rangle$ will be different for the two cases and cannot be calculated until the updraft of displaced fluid is taken into account. As explained in [3], the only quantity that can be calculated is $\langle \boldsymbol{U} - \boldsymbol{U}^{(0)} \rangle - \langle \boldsymbol{u} \rangle$, where $\langle \boldsymbol{u} \rangle$ is the average velocity of material (either fluid or solid) within the cloud and is determined by the overall specification of the problem and not by interactions between pairs of particles. The description in [22] of the calculation of $\langle \boldsymbol{U} - \boldsymbol{U}^{(0)} \rangle - \langle \boldsymbol{u} \rangle$ as being a change of reference frame is unfortunate because it could easily generate misleading ideas about changing from one set of axes to another. What is meant is that one is calculating the velocity of sedimentation relative to the updraft of displaced fluid.

An attempt such as the one above to use P_N functions as part of a calculational scheme would be a naive one; recent work using these functions [18, 28] recognizes the points made above and arrives at the same conclusions as are reached here, namely

(1) any estimate of $\langle \underline{\underline{S}} - \underline{\underline{S}}^{(0)} \rangle$ or $\langle \boldsymbol{U} - \boldsymbol{U}^{(0)} \rangle$ must include separate contributions from long-range interactions and from interactions between close particles,

(2) the long-range interactions manifest themselves mathematically as the terms which lead to non-convergent integrals when the P_A functions are used for averaging,

(3) for short-range interactions, P_N and P_A functions lead to the same results [12].

We now turn to the problem of calculating the effect of long-range interactions.

Calculating long-range interactions

The method used in [18] to calculate long-range interactions was an extension of the ideas described in the section on Einstein in that a distinction was made between the field ‘at infinity’ and the average field. The formulation in [28] is equivalent to [18]. For reasons which are given in the note added in proof at the end of this paper we shall concentrate here on discussing the subtraction method for calculating long-range interactions devised by Batchelor [3, 4]. The method uses only fields ‘at infinity’ which equal the average field, and thus is similar to the method offered earlier as an alternative to Einstein’s calculation. The aspect of the method which is most misunderstood is the way in which it apparently calculates long-range interactions using only the interactions between two particles. This impression is an understandable result of the form taken by the final integrals. Now, however, an example has been found [6] which shows that at least sometimes a correct application of the method requires knowledge of interactions between larger groups of particles even though the final integral still appears to require only two-particle interactions. The example also shows that estimates of the error made in the calculation, which are usually not given, are needed to ensure that the correct answer is obtained. In discussing the example I shall have to assume the reader is familiar with the basic subtraction device used by the method.

The example is a calculation to $O(c^2)$ of the compression modulus of a composite material containing spherical particles. Chen and Acrivos [6] chose a pure compression for their mean strain, i.e., $\overline{e_{ij}} = \Delta\, \delta_{ij}$. They then found three ways to obtain convergent two-particle approximations to the trace of $\langle\, S_{ij} - S_{ij}^{(0)} \,\rangle$, which led to three different results. The ways were:

(1) Take the trace of $S_{ij} - S_{ij}^{(0)}$ before averaging. The resulting convergent integral contained no long-range effects at all.

(2) Form the quantity $\langle\, S_{ij} - S_{ij}^{(0)} \,\rangle - A\langle\, e_{ij} - \Delta\delta_{ij} \,\rangle$, using the fact that $\langle\, e_{ij} - \Delta\delta_{ij} \,\rangle = 0$ and then approximate to two particles. The constant A is chosen so that the two-particle integral is convergent, but the long-range effects so calculated are not the correct ones.

(3) Approximate the quantity $\langle\, S_{ij} - S_{ij}^{(0)} \,\rangle - A_{ijkl}\langle\, e_{kl} - \Delta\delta_{kl} \,\rangle$ where the tensor A_{ijkl} is chosen so that the two-particle integral would converge for more general choices of the mean field than $\overline{e_{ij}} = \Delta\delta_{ij}$. This last choice gives the long-range effects correctly.

Chen and Acrivos found that the lack of uniqueness in the calculation arose because the disturbance strain field outside a spherical particle in a matrix in hydrostatic compression has the special form of a pure strain without dilatation, i.e., it has zero trace, and the constant A which was successful in producing a convergent integral was the component of A_{ijkl} appropriate to this state of affairs. In more general situations both pure strain and dilatation are present and the full A_{ijkl} tensor must be used. The correct choice is proved by considering the three-particle term in the general series expansion given by Jeffrey [13] and showing that only one choice gives a convergent integral at this higher order. This is the same as supplying an estimate of the error made in the calculation. The fact that one has a convergent integral, then, does not prove that long-range interactions have been accounted for correctly. Other less straightforward uses of Batchelor's method exist [20] which have yet to be made rigorous.

The macroscopic boundary and the infinite-volume limit

The above discussions have helped to elucidate the calculational procedures used in the past and also have established the interpretation of non-convergent integrals as consequences of long-range, multiparticle interactions which can nevertheless be reduced to integrals requiring knowledge only of two-particle interactions, provided the cautionary note of the last section is remembered. What we still need is a physical picture of the long-range interactions. Developing such a picture is the main aim here. An approach developed independently in [15, 27, 2l] which builds on earlier work [2, 9, 26] provides us with the required picture and at the same time provides a link between our considerations and the well-known 'self-consistent scheme' [16]. The new idea is to formulate the problem so that the bounding surface Γ and the manner in which it becomes infinitely large are considered explicitly.

Again using the elasticity problem as a specific example, the starting point is a finite sample of our composite material with displacements exactly equal to $\overline{e_{ij}}\, x_j$ on the bounding surface Γ. We write down an exact integral expression for the displacement at a point inside the sample, using the idea of polarization stress introduced earlier together with a Green's function for the matrix. We can use either the Green's function for an unbounded matrix or one for the volume bounded by Γ, different integrals over Γ being needed for the different cases. We shall use the one for an unbounded matrix so that the role of the integral over Γ is made clearer, but it must be emphasized that the final equation does not depend on this choice. The integral equation is [15,21]

$$u_i(\boldsymbol{x}) = \int_V G_{ij,k}(\boldsymbol{x}-\boldsymbol{x}')\tau_{jk}(\boldsymbol{x}')\,\mathrm{d}V(\boldsymbol{x}') + \int_\Gamma (G_{ij}\sigma_{jk} - u_j J_{jki}) n_k \,\mathrm{d}A(\boldsymbol{x}') \ ,$$

where G_{ij} is the Green's function for the unbounded medium and $J_{jki} = \frac{1}{2} L^1_{jklm}(G_{mi,l} + G_{li,m})$. Now to the crudest approximation $\underline{\underline{\tau}}$ is constant inside an inclusion and zero in the matrix, so the volume integral is a sum of terms $O(r^{-2})$ (because $G_{ij,k}$ is $O(r^{-2})$) where r is the distance from $\boldsymbol{x}$ to an inclusion. Therefore it is not possible to proceed to a limit of infinite volume and ignore the effect of the surface Γ without first casting the surface integral into a more suitable form.

Before proceeding to the manipulation of the equation, we note that because our equation is in terms of the Green's function for an unbounded medium, we require $\underline{\underline{\sigma}}$ as well as $\boldsymbol{u}$ on the bounding surface Γ. We know from the definition of the problem that $u_i = \overline{e_{ij}}\, x_j$ on Γ but $\underline{\underline{\sigma}}$ is unknown. It may seem then that the new formulation of the problem has too many unknowns in it, $\underline{\underline{\sigma}}$ on Γ as well as L^{e}. We shall find, however, that to solve the equations and find L^{e} to any order in the volume fraction c (say c^p), the stress is needed on the boundary only to $O(c^{p-1})$ and a simple iterative procedure is then available to us. A further consequence of $\underline{\underline{\sigma}}$ appearing in our integral equation is that our equations will be implicit ones for $\overline{\underline{\underline{\tau}}}$ (or equivalently $\langle\, \underline{\underline{S}}\, \rangle$). We start our transformation of the equation by replacing u_i in the integral over Γ by $\overline{e_{ij}} x'_j$ and arguing that, provided Γ is suitably large and smooth, we can approximate $\underline{\underline{\sigma}}$ by $\overline{\underline{\underline{\sigma}}}$ (the conditions under which this approximation is valid require further study):

$$u_i = \int_V G_{ij,k} \tau_{jk}\, \mathrm{d}V + \int_\Gamma (G_{ij} \overline{\sigma_{jk}} - \overline{e_{jm}} x'_m J_{jki}) n_k \, \mathrm{d}A \ .$$

The divergence theorem can now be used to transform the surface integral to a volume integral and the limit $V \to \infty$ taken; the details can be found in [15, 21]. Here we shall quote the results of the transformation. The surface integral is replaced by two terms, one of these simply ensures that $\langle\, u_i\, \rangle = \overline{e_{ij}} x_j$ while the other one is

$$- \int_V G_{ij,k} \overline{\tau_{jk}}\, \mathrm{d}V = -n \int G_{ij,k} \langle\, S_{jk}\, \rangle \, \mathrm{d}V \ ,$$

where n is again the number density of inclusions. When we combine this volume integral with the one already in our equation for

u_i we obtain the term

$$\int_V G_{ij,k}(\tau_{jk} - \overline{\tau_{jk}})\,\mathrm{d}V \ ,$$

which has been shown in [27] to be absolutely convergent.

It is important to realize that the above considerations are not in conflict with other approaches [29, 30]. By using the Green's function for the medium bounded by Γ, Kröner and Koch [30, equation 5] seem to obtain an equation which does not contain the $\overline{\underline{\underline{\tau}}}$ term. Before solving their equation in the $V \to \infty$ limit, however, they modify their equation using an operator P [30, equation 19] and this step is equivalent to introducing the $\overline{\underline{\underline{\tau}}}$ term. Similarly in [29] the use of [29, equation 13] in preference to [29, equation 17] is closely connected with the need for the $\overline{\underline{\underline{\tau}}}$ term here to ensure convergence in the $V \to \infty$ limit. See [17] for further discussion.

The reader is referred for the method of solution of the transformed equation, including the iterative procedure for handling the appearance of the unknown $\overline{\underline{\underline{\tau}}}$ in the equation, to [21] and [27]. Note, however, that in [27] the authors separate the two terms which together guarantee the convergence of the integrals in the formulation and evaluate them for a specific (elliptical) outer boundary Γ; their proof earlier in their paper of the convergence of the combined integral allows them to justify this, but it is an unfortunate way to present the calculation.

A physical picture for convergence difficulties

The equations given above allow us finally to present a physical picture to explain the occurrence of non-convergent integrals. This picture is inevitably given in terms of the particular examples that were chosen for study here, but the principles should be clear enough for their application to other examples to be straightforward. We remember first that part of the picture was given earlier in connection with the discussion of the sedimentation problem. There it was pointed out that the $O(r^{-1})$ integrands that arise in the calculations correspond to the fact that a statistically non-homogeneous suspension of particles (for example, a cloud of particles surrounded by clear fluid) and a statistically homogeneous suspension will settle at different speeds and this difference cannot be calculated from local interactions between particles. We now turn to the $O(r^{-3})$ integrands of the elasticity problem and to the ambiguity discovered by Chen and Acrivos [6].

Referring to the equation for $\boldsymbol{u}$ derived above, we can think of the two terms that appeared in the equation when the surface integral over Γ was transformed to a volume integral as corresponding to two different aspects of long-range interactions. The first aspect is simply that the average field $\overline{\underline{\underline{e}}}$ is supplied through displacements on the boundary Γ and it is reflected in the $\overline{e_{ij}}x_j$ term. In Batchelor's subtraction procedure it is reflected in the choice of $\langle\, e_{ij} - \overline{e_{ij}}\,\rangle$ as the quantity to be subtracted and it is also reflected in the difference between $\overline{\underline{\underline{e}}}$ and $\underline{\underline{e}}^*$ in Einstein's calculation; it has long been known also in the theory of dielectrics [32, section 5]. The second aspect is reflected in the volume integral of $\overline{\underline{\underline{\tau}}}$. To interpret this we note that the volume integral gives the displacement that would be produced in a matrix in which the polarization stress $\underline{\underline{\tau}}$ was uniform and equal to $\overline{\underline{\underline{\tau}}}$. By definition, however, a matrix which has elastic moduli L^1 and which has throughout it a uniform polarization stress $\overline{\underline{\underline{\tau}}}$ is equivalent to a matrix with moduli L^{e}. Thus our second aspect arises because we initially formulated our calculation supposing that an inclusion “saw”, far from itself, the matrix L^1, whereas it actually sees an effective matrix L^{e}. This view of the problem is the link with the ‘self-consistent’ scheme mentioned above, and in fact the results of the two approaches can be made to tally so far as the long-range effects alone are concerned [6, 12, 15]. The present formulation allows us to go further than the self-consistent scheme and calculate the additional modifications to L^{e} due to interactions between neighbouring inclusions (and we can use either nearest neighbours or any neighbours for this part of the calculation). In Batchelor's subtraction procedure this second aspect of the long-range effects is reflected in the constant that is chosen to ensure that the final integral expression is convergent. Moreover, it has been shown in [21] that using the present interpretation gives another way of choosing the correct constant in the Chen and Acrivos problem. A similar idea of embedding in effective media is used in the theory of dielectrics, for example, in deriving the Clausius-Mosotti equation [31, section 33].

Other types of non-convergent integrals

In this final section, we point out two directions in which work is progressing. First, in [17] the above considerations have been extended to media with continuous variations in properties. In such cases, an approach often adopted is to seek a perturbation solution in terms of the strength of the property fluctuations. For example, in the elastic problem used earlier, the elastic moduli L are taken as random functions of position and the definition of polarization stress used earlier is replaced by the new definition $\sigma_{ij} = \langle L_{ijkl} \rangle e_{kl} + \tau_{ij}$ where $\tau_{ij} = (L_{ijkl} - \langle L_{ijkl} \rangle) e_{kl}$. If the fluctuations $L - \langle L \rangle$ can be assigned a magnitude ϵ, then it can be shown that non-convergent integrals can appear in expressions for the effective moduli at $O(\epsilon^4)$. The high order at which non-convergence first appears stands in contrast to the examples above; the reason lies in the fact that in our examples $\underline{\underline{\tau}}$ was defined using the quantity $L - L^1$ which has the non-zero mean $c(L^2 - L^1)$ whereas in the perturbation approach the quantity used,

$L - \langle L \rangle$, has zero mean [17].

The second direction in which new work is progressing is motivated by the existence of problems for which the methods described above fail to handle all the long-range effects present. These new effects show up mathematically as non-convergent integrals still present in the equations after the reformulations above have removed the familiar troublesome terms [4, 11]. It is obviously desirable to be able to recognize these more difficult problems. One means of recognition has been given in [13] which examines the interactions between pairs of particles using the 'method of ref1exions'. The key step is to determine the number of reflexions which lead to non-convergent interactions. A more physical idea is an extension of the 'self-consistent' ideas discussed above. We shall use the example of flow through a bed of fixed particles to illustrate this. Suppose fluid is flowing through an array of particles, each of which is held fixed in space. Near any one particle, the problem is one of flow of a viscous fluid around a particle and the equations are the familiar Stokes equations for creeping flow. Far from the particle, however, the problem is one of flow through a porous medium and the equations are Darcy's equations for a porous medium. The methods described above assume that the small-scale problem around any particle, and the large-scale problem far from anyone particle are governed by the same equations with possibly different constants (i.e., for the elastic problem L^1 near a particle and L^{e} far from it). Problems in which the governing equations themselves change require a more subtle formulation.

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Note Added in Proof

An additional comment is required on the nearest-neighbour formulation since it has been advocated elsewhere in this volume [28]. Consider the problem of flow past fixed particles which was discussed briefly in the last main paragraph of this paper and studied extensively in [11, 24]. Both the nearest-neighbour approach and the any-neighbour subtraction approach fail to obtain the correct average drag on a particle (both miss the $c^{1/2}$ term), but the ways in which they fail are quite different. The any-neighbour approach fails visibly because the non-convergent integrals cannot be removed, while the nearest-neighbour approach obtains a wrong answer without giving any indication of having failed. This underlines again the point made in the text that any method must include some way of estimating errors and the fact that an answer is obtained is no proof that the answer is correct. The any-neighbour approach has been extended to include estimating errors [13] but not the nearest-neighbour approach. Consequently there is always the danger that the approach will be used unwittingly for problems, such as the fixed particle one, to which it cannot be applied.