This is a re-typed version of a paper published in the conference proceedings *Continuum Models of Discrete Systems*, editor J.W. Provan, published by The University of Waterloo Press in 1977. The page numbering is the same as the original, but the line breaks are not the same.

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

D. J. Je®rey
Department of Applied Mathematics and Theoretical Physics
Cambridge University, Silver Street

(1) long-range e®ects will produce non-convergent integrals in incorrectly formulated calculations of e®ective properties,

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

(3) the correct formulation of calculations of e®ective 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 con°icting results would provide, to examine the various schemes critically. This paper shows, however, that cases do exist in which di®erent schemes lead to con°icting 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 di±culties have not had, until now, a su±ciently 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 ¯nite, 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 signi¯cant' ¯nite value. This has led to a narrow view of the problem. For a start, it is by no means always possible to ¯nd such ¯nite 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 con¯ned 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 $\underline{\underline{e}}$

as an estimate for $\overline{W}$ the expression $2^{1}\overline{e_{ij}}\ \overline{e_{ij}}$(1

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 ¡ 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 e®ective 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 ¯nally that Einstein o®ered 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 signi¯cant'. It is clear, however, that any other shape, although giving di®erent expressions for $W^{¤}$ and $\underline{\underline{e}}^{¤}$ as functions of $\bar{\underline{\underline{e}}}$, would give the same expression for e®ective 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

The second problem is an e®ective-modulus problem and is the linearly elastic analogue of Einstein's calculation. We wish to relate the (volume) average stress $\overline{¾_{ij}}$ in a linearly elastic material to the average strain $\overline{e_{ij}}$ through e®ective moduli L^{e}_{ijkl} [6, 9, 10, 27]. Using the idea of polarization stress, due to Eshelby and KrÄoner [16], we have the stress at any point in the composite material given by

$$¾_{ij} = L^1_{ijkl} e_{kl} + ¿_{ij} \; ;$$

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

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

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 $¾_{ij}$ then gives

$$\overline{¾_{ij}} = L^1_{ijkl} \overline{e_{kl}} + \overline{¿_{ij}} = L^1_{ijkl} \overline{e_{kl}} + n h S_{ij} \, i \; :$$

Here n is the number density of inclusions and the angle brackets have been used to show that, to ¯nd the average value of $\underline{\underline{S}}$, we must use ensemble averaging over con¯gurations of particles. The relation between $nh\underline{\underline{S}}\,i$ and $\overline{\underline{\underline{e}}}$ will introduce the concentration tensors of Hill [10]. Having expressed $\overline{\underline{\underline{¾}}}$ in terms of $nh\underline{\underline{S}}\,i$, 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 speci¯c 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 $\underline{\underline{\bar{e}}}$. Then $\langle \underline{\underline{S}} \rangle = \underline{\underline{S}}^{(0)}$ 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 subat the centre of the test particle and select a point $\mathbf{r}$; $P_A(\mathbf{r}|\ )d\mathbf{r}$ is then the probability that the centre of any particle of the suspension will be within the volume element $d\mathbf{r}$ while $P_N(\mathbf{r}|\ )d\mathbf{r}$ is the probability that the centre of the nearest neighbour to the test particle will be within $d\mathbf{r}$. When the point $\mathbf{r}$ 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, $\mathbf{U}-\mathbf{U}^{(0)}$ 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 $\langle\mathbf{U}-\mathbf{U}^{(0)}\rangle$ could be obtained by averaging the effect of just one other particle on the test particle and write

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

Since $\mathbf{U}-\mathbf{U}^{(0)}$ is for two [redacted]rticles [redacted]n $O(r^{-1})$ quantity, it is important to know whether P_A or P_N is use[redacted] the [redacted]ntegral. If P_A is used the integral is non-convergent, while if P_N is [redacted] use[redacted] th[redacted] integral is convergent and gives an $O(c^{1/3})$ approximation for $\langle\mathbf{U}-$[redacted]$\mathbf{U}^{(0)}$ [redacted]his last result is in conflict with work [3, 24] which finds an $O(c$ [redacted] s$_A$ used [redacted]. This cal p(2,nprobabi35 h w b4canb402(b)26(y(b4c)-3tru 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

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 ¯eld `at in¯nity' and the average ¯eld. 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 ¯elds `at in¯nity' which equal the average ¯eld, and thus is similar to the method o®ered 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 ¯nal 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 ¯nal 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., $\overline{e_{ij}} = ¢ \pm_{ij}$. They then found three ways to obtain convergent two-particle approximations to the trace of $h S_{ij}$ ¡ $S_{ij}^{(0)} i$, which led to three di®erent results. The ways were:

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

(2) Form the quantity $\langle S_{ij} - S_{ij}^{(0)} \rangle - A \langle e_{ij} - \cdot \delta_{ij} \rangle$, using the fact that $\langle e_{ij} - \cdot \delta_{ij} \rangle = 0$ 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 $\langle S_{ij} - S_{ij}^{(0)} \rangle - A_{ijkl} \langle e_{kl} - \cdot \delta_{kl} \rangle$ where the tensor A_{ijkl} is chosen so that the two-particle integral would converge for more general choices of the mean field than $\overline{e_{ij}} = \cdot \delta_{ij}$. 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 interpre384(fullln6.full26(es=on-con)26(v)27(ergen 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 ¡ and the manner in which it becomes in¯nitely large are considered explicitly.

Again using the elasticity problem as a speci¯c example, the starting point is a ¯nite sample of our composite material with displacements exactly equal to $\overline{e_{ij}}\, x_j$

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 $\langle \underline{\underline{S}} \rangle$).

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 $\overline{\underline{\tau}}$ 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 $\overline{\underline{\tau}}$ term. Similarly in [29] the use of [29, equation 13] in preference to [29, equation 17] is closely connected with the need for the $\overline{\underline{\tau}}$ 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 $\overline{\underline{\tau}}$ 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

L ¡ hL i, 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 e®ects present. These new e®ects 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 di±cult 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 re°exions 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 °ow through a bed of ¯xed particles to illustrate this. Suppose °uid is °owing through an array of particles, each of which is held ¯xed in space. Near any one particle, the problem is one of °ow of a viscous °uid around a particle and the equations are the familiar Stokes equations for creeping °ow. Far from the particle, however, the problem is one of °ow 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 di®erent 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.

Acknowledgements

I gratefully thank Professor G. K. Batchelor for his comments on earlier drafts of this paper; also Professor J. J. McCoy for many stimulating discussions.

References

1. AERO, E.L., BULYGIN, A.N. and KUVSHINSKII, E.V., *Prikl.Mat.Mekh.*, Vol. 29, 1965, p. 297.
2. BATCHELOR, G. K., *Introduction to Fluid Dynamics*, Cambridge, 1967.
3. BATCHELOR, G. K., *J. Fluid Mech.*, Vol. 52, 1972, p. 245.
4. BATCHELOR, G. K., *Ann. Rev. Fluid Mech.*, Vol. 6, 1974, p. 227.
5. BROWN, W. F., *Trans. Soc. Rheol.*, Vol. 9, 1965, p. 357.
6. CHEN, H.-S. and ACRIVOS, A., *Int.J.Solids Struc.*, Vol.14, 1978, p.349.
7. EINSTEIN, A., *Ann. Phys.*, Vol. 19, 1906, p. 289 and Vol. 34, 1911, p. 591.
8. EINSTEIN, A., *Theory of Brownian Movement*, Dover, 1956.
9. 9H-1S5ETBTS0 p.8i28AppdVRnf.i0Dover,

22. PYUN, C, W. and FIXMAN, M., *J. Chem. Phys.*, Vol. 41, 1964, p. 937.

23. RAYLEIGH, LORD, *Phil. Mag.*, Vol. 34, 1892, p. 481.

24. SAFFMAN, P. G., *Stud. Appl. Math.*, Vol. 52, 1973, p. 115.

25. SAITÔ, N., *J. Phys. Soc. Japan*, Vol. 7, 1952, p. 447.

26. SPENCER, A. J. M., *Int. J. Engineering Sci.*, Vol. 3, 1965, p. 441.

27. WILLIS, J. R. and ACTON, J. R., *Q. J. Mech. Appl. Math.*, Vol. 29, 1976, p. 163.

28. GODDARD, J. D., Proceedings of the Second International Symposium on Continuum Models of Discrete Systems, SM Study No. 12, Solid Mechanics Division, Univ. of Waterloo Press, 1977. p. 605.

29. GUBERNATIS, J. E., Proceedings of the Second International Symposium on Continuum Models of Discrete Systems, SM Study No. 12, Solid Mechanics Division, Univ. of Waterloo Press, 1977. p. 25

30. KRÖNER, E. and KOCH, H., *SM Archives*, Vol. 1, 1976, p. 183.

31. BÖTTCHER, C. J. F., *Theory of Electric Polarisation*, Elsevier, 1952.

32. FRÖHLICH, H., *Theory of Dielectrics*, Oxford, 1949.

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.