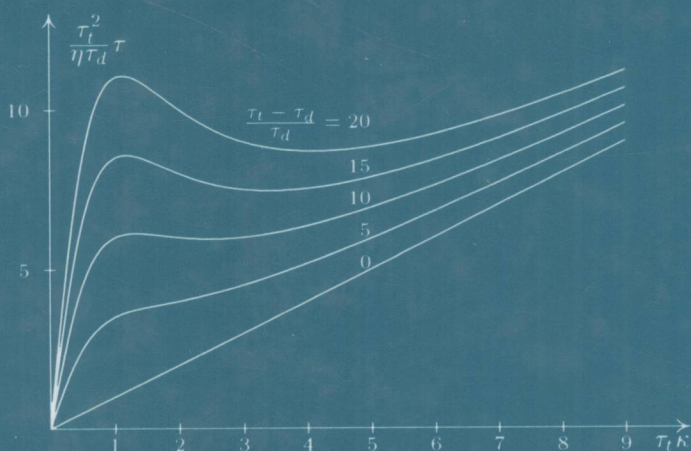


J. Verhás

Thermodynamics and Rheology



AKADÉMIAI KIADÓ, BUDAPEST

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THERMODYNAMICS AND RHEOLOGY



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THERMODYNAMICS AND RHEOLOGY

PREFACE

To facilitate the application of physical theories in practice, researchers today must develop new methods not only for the systematization and interpretation of increasing experimental data but also for the simplification, unification and combination of already existing and well-functioning theories. A step toward this goal is served by the present text which attempts to compose a general theory of classical field incorporating continuum mechanics, electrodynamics and thermodynamics. Although looking mainly at the mechanical motion of media, the methods described by this text point far beyond the rheological applications (as indicated in the title) to an exact theory of non-equilibrium thermodynamics, thus making possible a unified description of mechanical, electromagnetic and thermic phenomena together with the interrelations between them. As a result, this theory offers a strong organizational force and an extraordinarily wide range of applications. Knowledge of non-equilibrium thermodynamics is indispensable for the physicists dealing with transport processes, physical chemistry, plasma physics or energetics, for the chemical engineer and even for biophysicist and biologist. To the aforementioned enumeration can be added electrical, mechanical and civil engineers as well as architects engaged in dielectrics, structural materials, colloid agents or even in liquid crystals. This wide range of application of irreversible thermodynamics arises from the fact that in nature any macroscopic process is irreversible.

This book deals both with the complicated and far-reaching forms of motion of the materials continuously filling up the universe and with establishing principles in joining classical field theoretical methods with irreversible thermodynamics, using macroscopic methods but not forgetting the corpuscular structure of the agents. By elaborating on these methods in detail, the proper means for the quantitative description of irreversible phenomena—means formerly lacking from the viewpoint of generality, mathematical exactness and direct applicability—are now evident.

The results outlined in this book speak for themselves. The general feature of the correlations presented and—at the same time—their simplicity will surprise even the reader familiar with literature of the topics. To those not acquainted with the subject matter of deformation and flow or with the relevant optical and electromagnetic phenomena, it serves as an introduction.

The harmonic coordination of the succinctly expressed vast knowledge assures—beyond the aesthetic experience—its comprehension, understanding and immediate applicability.

Successful striving for mathematical simplicity is an advantage of the book. The author avoided complicated methods too often seen in the literature (for example, he did not apply the Ricci calculus that—considering the Euclidean structure of surrounding classical space—has an advantage only for numerical solutions of special problems). The importance of the topics presented and the precision and applicability of the new thermodynamic theory (the so-called dynamic variables introduced by the author) will usher in a new epoch in the literature of the topics.

István Gyarmati

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INTRODUCTION

Thermodynamics as a field theory has a history of a few decades only. Its methods—we daresay—have been elaborated but are not final at all. They are changing and transforming even at present. At the same time, the rapid development in technology increases the demand for a unified and general theory of classical field incorporating continuum mechanics, electrodynamics, and thermodynamics. The need for practical applications has led to the development of several methods and ideas differing from each other according to the different branches of applications, nevertheless, overlapping and similarity are not rare. Unification is rather hard, partly due to the different intentions, partly due to the enormous amount of knowledge involved.

I intend to contribute to the unification following Gyarmati [75] who made the first consistent steps in his book in 1970. The starting point of my work is the field theoretical view of thermodynamics—founded by Onsager in 1931 [137, 138]—of irreversible processes. I have fitted the aspects of non-equilibrium thermodynamics to classical and modern concepts and methods of mechanics and electrodynamics.

In this work I, of course, had to compromise as well. For example, I avoid the difficulties due to the complexity of mathematical apparatus of general relativity (and not to be hindered in my work by the undeveloped theory of relativistic thermodynamics), I limited my considerations to slowly moving media and accordingly used the correlations of electrodynamics in approximate form.

As my work may only be the first step in the elaborating of this theory, in this book I give only those consequences I deemed the most important. First of all, the linear constitutive equations of Onsager's thermodynamics are applied (although from time to time the linear and non-linear alternatives come in question as well, but only to show the possibilities for continuing this work). The majority of methods and concepts applied in my work belong to non-equilibrium thermodynamics: however, I relied considerably on the methods of continuum mechanics, rheology and electrodynamics. For example, I borrowed the often applied complex formalism from the linear theory of electric networks.

The composition of this book reflects the synthesizing character of my work. Chapters 1 and 2 include the principles of continuum mechanics to the extent needed to expound a unified theory by giving the definitions taken from continuum mechanics together with their correlations. The apparatus required to consider the effects of gravitational and electromagnetic fields is so defined that it can be applied even in the case of micropolar media.

Chapter 3 outlines the well-known methods relating to stress and strain. This Chapter aims partly at summarizing the experimental results and partly at indicating the ideas taken from rheology and continuum mechanics, respectively.

Chapter 4 includes the setting forth of irreversible thermodynamics, acquainting the reader with balance equations and “linear” laws of irreversible thermodynamics as well as with the Onsager-Casimir reciprocal relations, and, further, with Gyarmati’s variational principle. The new theory and applications of dynamic variables is described in this chapter. This chapter sums up some results of irreversible thermodynamics partly reached by me, partly born while writing this book, so their direct application—at least as to rheology—could hardly come on as yet.

Chapter 5 is based on the hypothesis of local equilibrium. It also incorporates theories of classical elasticity, thermoelasticity and Newtonian fluids into the framework of thermodynamics.

Chapter 6 studies media far from equilibrium, rejecting the hypothesis of local equilibrium. New scientific results follow the solution of the linear constitutive equations of Onsager’s thermodynamics, thus enabling models of rheology to begin now from a single uniform basic principle. The viscoelastic and plastic response, Ostwald’s curve characterizing the generalized Newtonian fluids, the effect of creep, the elastic features preceding plastic flow, the basic interrelations of rheoptics, etc. are interpreted with phenomenological methods, quantitatively. The consistent character of the method is shown by the self-evident theoretical proof of the empirical Cox-Mertz rule [30]. It is confirmed that its limit of validity coincides with that of strictly linear rules. This book over-rides the limits of the linear theory, but only in the simplest case to show the direction for the future.

Chapter 7 deals with electromagnetic phenomena including the irreversible thermodynamic theory of streaming birefringence and photoelasticity. Here the *raison d’être* of quasi-linear theory is verified.

Chapter 8 outlines some practical applications of the aforesaid theory. Several colloids, polymers as well as the liquid crystals are described here. In case of colloids—after thermodynamic considerations—the equivalent theory based on microscopic structure is mentioned. The two methods do not exclude each other; in fact, the microscopic considerations complete well those of thermodynamics giving the graphic meaning of phenomenological conductive coefficients. Here I draw the attention of the reader to the thermodynamic method which gives the mathematical form of the functions studied more briefly and elegantly than the mathematical methods based on partial differential equations that are essential for other approximations; moreover it is free from the uncertainties due to not perfect reliability in each case separately of hypothesis regarding the microscopic structure of a particular material.

The Appendix sums up the applied mathematical apparatus, but only to facilitate the interpretation of used notations.

It is a pleasure to thank Prof. I. Gyarmati for his introducing me—at a high international level—to the basis of irreversible thermodynamics. During my three-decade research work, he always inspired me to reach considerable results, and placed his extensive knowledge at my disposal.

The Author is deeply obliged to the Hungarian National Scientific Research Fund, OTKA (1949, T-17000) and the EC (Contract No: ERBCIPDCT 940005) for supporting the research work included into the book, moreover, the preparation of the manuscript and the edition.

KINEMATICS

1.1. Motion of continua

The motion of a body is kinematically known if we know the position of any point at any time. Many methods are known to label the points of the body. One possibility is to use the position occupied at a given time t_0 to denote the moving point. To give the positions, some kind of frame of reference is applied. The usual mathematical means are a system of coordinates fixed to a frame of reference. The mode of selecting the reference and coordinate system may be optional in principle: however, the proper selection can simplify the calculus to be done. We can use Cartesian coordinates in Euclidean space; the vectors of the Euclidean space are suitable for indicating positions.

Select a moving body and choose its point P_0 . Denote the rectangular coordinates of the point in time t_0 with x_1, x_2, x_3 in the coordinate system defined by orthonormal base vectors (unit vectors) $\mathbf{j}_1, \mathbf{j}_2, \mathbf{j}_3$. Then call the vector

$$\mathbf{X} = X_1\mathbf{j}_1 + X_2\mathbf{j}_2 + X_3\mathbf{j}_3 \quad (1.1)$$

the position vector of point P_0 in time t_0 . Denote the coordinates of points P_0 in time t with x_1, x_2, x_3 . The vector

$$\mathbf{x} = x_1\mathbf{j}_1 + x_2\mathbf{j}_2 + x_3\mathbf{j}_3 \quad (1.2)$$

is the position vector of point P_0 . It is changing in time. We can regard the motion of point P_0 as kinematically known if we know the function

$$\mathbf{x} = \mathbf{x}(t). \quad (1.3)$$

If we choose another point P instead of point P_0 then the vector \mathbf{X} will be quite different and even the function $\mathbf{x} = \mathbf{x}(t)$ will be replaced. Therefore, we can say that the position vector \mathbf{x} depends not only on time but on the point whose motion it describes. As we apply the \mathbf{X} vector to specify the points of the body, we can mathematically give the motion by the so-called motion function

$$\mathbf{x} = \mathbf{x}(\mathbf{X}, t). \quad (1.4)$$

We can call the numbers x_1, x_2, x_3 as Eulerian or space coordinates and the numbers X_1, X_2, X_3 as material or Lagrange coordinates [6, 29, 44, 81, 109, 169]. The mapping (1.4) is assumed to be single valued and to have continuous partial derivatives except possibly at some singular points, curves and surfaces. Furthermore, its Jacobian

$$j = \frac{\partial(x_1, x_2, x_3)}{\partial(X_1, X_2, X_3)} = \begin{vmatrix} \frac{\partial x_1}{\partial X_1} & \frac{\partial x_1}{\partial X_2} & \frac{\partial x_1}{\partial X_3} \\ \frac{\partial x_2}{\partial X_1} & \frac{\partial x_2}{\partial X_2} & \frac{\partial x_2}{\partial X_3} \\ \frac{\partial x_3}{\partial X_1} & \frac{\partial x_3}{\partial X_2} & \frac{\partial x_3}{\partial X_3} \end{vmatrix} \quad (1.5)$$

is positive, i.e.,

$$0 < j < \infty. \quad (1.6)$$

This inequality physically means the indestructibility of the material. It shows that during motion the body of finite volume was always of finite volume and will remain under any circumstances. To understand this, consider an infinitesimal parallelepiped with edges dX_1, dX_2, dX_3 around the point X_1, X_2, X_3 . During the motion, the edges of the parallelepiped $dX_1\mathbf{j}_1, dX_2\mathbf{j}_2, dX_3\mathbf{j}_3$ deform to edges $\frac{\partial \mathbf{x}}{\partial X_1}dX_1, \frac{\partial \mathbf{x}}{\partial X_2}dX_2, \frac{\partial \mathbf{x}}{\partial X_3}dX_3$, while the parallelepiped remains parallelepiped, the volume of which can be given by

$$dV = \left(\frac{\partial \mathbf{x}}{\partial X_1}dX_1 \times \frac{\partial \mathbf{x}}{\partial X_2}dX_2 \right) \frac{\partial \mathbf{x}}{\partial X_3}dX_3 \quad (1.7)$$

In coordinates, it makes

$$dV = \begin{vmatrix} \frac{\partial x_1}{\partial X_1} & \frac{\partial x_1}{\partial X_2} & \frac{\partial x_1}{\partial X_3} \\ \frac{\partial x_2}{\partial X_1} & \frac{\partial x_2}{\partial X_2} & \frac{\partial x_2}{\partial X_3} \\ \frac{\partial x_3}{\partial X_1} & \frac{\partial x_3}{\partial X_2} & \frac{\partial x_3}{\partial X_3} \end{vmatrix} dX_1 dX_2 dX_3 = j dV_0 \quad (1.8)$$

where dV_0 is the original volume in time t_0 . It is very important beyond the fundamental physical meaning that the inequality (1.6) assures the invertability of the deformation function, i.e., the inverse function

$$\mathbf{X} = \mathbf{X}(\mathbf{x}, t) \quad (1.9)$$

exists.

In many important cases, making distinction between certain points is only of theoretical importance: therefore the use of spatial or Eulerian description is common, especially when discussing liquids. This means that we regard the motion as known if we know the velocity as a function of time and place:

$$\mathbf{v} = \mathbf{v}(\mathbf{x}, t). \quad (1.10)$$

The connection between these two kinds of description can be summed up as follows: The time derivative of the motion gives the velocity in function of time and starting point:

$$\mathbf{v} = \frac{\partial \mathbf{x}(\mathbf{X}, t)}{\partial t}. \quad (1.11)$$

However, the starting point can be given with the help of (1.9) as a function of the instantaneous position and time. So considering (1.9), we turn (1.11) into (1.10) as

$$\mathbf{v} = \frac{\partial \mathbf{x}[\mathbf{X}(\mathbf{x}, t), t]}{\partial t} = \mathbf{v}(\mathbf{x}, t). \quad (1.12)$$

Doing the reverse—from (1.12) to (1.04)—is more complicated. Let us regard the function (1.10) as a vectorial differential equation for the position of a selected point P_0

$$\frac{d\mathbf{x}}{dt} = \mathbf{v}(\mathbf{x}, t). \quad (1.13)$$

In the general solution, three arbitrary numbers appear, the value of which are determined by the initial condition

$$\mathbf{x} = \mathbf{X}, \quad \text{if} \quad t = t_0. \quad (1.14)$$

In this way, we can get the motion function

$$\mathbf{x} = \mathbf{x}(\mathbf{X}, t).$$

From the aforesaid it can be seen that from a physical point of view the two descriptions are equivalent: we can change over from one to the other at any time.

1.2. Strain

The motion of a body, as we mentioned before, is described in general with a function (1.4). As not any motion of the body results in deformation, it is more precise to apply the term "motion" instead of the frequently used term "deformation".

A characteristic of motions without strain is that during motion the distance between any pair of points remains unchanged. These motions are called rigid body motions.

Let us examine which of the functions (1.4) indicate motion without strain. Consider two different, but otherwise arbitrary points of the medium P_1 and P_2 . The distance between them does not change, so

$$(\mathbf{x}_1 - \mathbf{x}_2)^2 = (\mathbf{X}_1 - \mathbf{X}_2)^2 \quad (1.15)$$

that can be written as

$$\sum_{i=1}^3 (x_{1i} - x_{2i})^2 = \sum_{J=1}^3 (X_{1J} - X_{2J})^2 \quad (1.16)$$

in rectangular coordinates. Making use of the inverse function (1.9), (1.16) becomes:

$$\sum_{i=1}^3 (x_{1i} - x_{2i})^2 = \sum_{J=1}^3 [X_{1J}(\mathbf{x}_1, t) - X_{2J}(\mathbf{x}_2, t)]^2. \quad (1.17)$$

As the point P_1 is arbitrary, this equation holds for any \mathbf{x}_1 ; and also the partial time derivatives with respect to x_{1i} of the right and left sides are equal:

$$x_{1i} - x_{2i} = \sum_{J=1}^3 (X_{1J} - X_{2J}) \frac{\partial X_{1J}}{\partial x_{1i}}. \quad (1.18)$$

Introducing the form

$$Q_{iJ} = \frac{\partial X_{1J}}{\partial x_{1i}} \quad (1.19)$$

and rearranging (1.18), we can write

$$x_{2i} = x_{1i} + \sum_{J=1}^3 Q_{iJ}(X_{2J} - X_{1J}) \quad (1.20)$$

whose vector form is

$$\mathbf{x}_2 = \mathbf{x}_1 + \mathbf{Q}(\mathbf{X}_2 - \mathbf{X}_1). \quad (1.21)$$

The Q_{ij} quantities (i.e., the tensor \mathbf{Q}) does not depend on how point P_2 has been chosen: namely, its components can be calculated by (1.19) only from the coordinates belonging to point P_1 . Exchanging P_1 and P_2 we obtain

$$\mathbf{x}_2 = \mathbf{x}_1 + \mathbf{Q}'(\mathbf{X}_2 - \mathbf{X}_1) \quad (1.22)$$

being fully similar to (1.21) where \mathbf{Q} does not depend on the selection of point P_1 . The equivalence of (1.21) and (1.22) results in $\mathbf{Q} = \mathbf{Q}'$, i.e., \mathbf{Q} is identical for any P_1 and P_2 .

On the basis of (1.15) and (1.21), the tensor \mathbf{Q} leaves the absolute value of any vector unchanged. Based on this and using the equations

$$(\mathbf{Q}\mathbf{A})^2 = \mathbf{A}^2; \quad (\mathbf{Q}\mathbf{B})^2 = \mathbf{B}^2; \quad [\mathbf{Q}(\mathbf{A} + \mathbf{B})]^2 = (\mathbf{A} + \mathbf{B})^2,$$

it is seen that tensor \mathbf{Q} leaves the scalar product of any two vectors unchanged:

$$(\mathbf{Q}\mathbf{A}, \mathbf{Q}\mathbf{B}) = (\mathbf{A}, \mathbf{B}), \quad (1.23)$$

so \mathbf{Q} is orthogonal, i.e., $\mathbf{Q}^T = \mathbf{Q}^{-1}$ and, therefore, $\mathbf{Q}\mathbf{Q}^T = \delta$. On the other hand, it can be understood that during any motion given by the function

$$\mathbf{x} = \mathbf{a}(t) + \mathbf{Q}(t)\mathbf{X} \quad (1.24)$$

(\mathbf{Q} orthogonal tensor), the distance of any two points is constant. The class of functions which do not result in strain is thus evident. The $\mathbf{a}(t)$ function shows the translation of the body, while the tensor $\mathbf{Q}(t)$, the rotation. The $\mathbf{Q}(t)$ may not mean reflection as the determinant of reflections is -1 ; and the determinant of $\mathbf{Q}(t)$ is non-negative in accordance with (1.6).

The motions which can be written in a form similar to (1.24)

$$\mathbf{x} = \mathbf{a}(t) + \mathbf{x}\mathbf{X} \quad (1.25)$$

(where \mathbf{x} is not orthogonal) are called homogeneous deformations.

The tensor \mathbf{x} is called the deformation gradient. The deformation gradient describes the strain and rotation of an environment of point P_0 .

To separate the rotation and strain, Cauchy's polar decomposition theorem is used. On the basis of this theorem, any non-singular, and invertible second order tensor \mathbf{x} can be factorized in form

$$\mathbf{x} = \mathbf{R}\mathbf{D} = \mathbf{d}\mathbf{R} \quad (1.26)$$

where \mathbf{D} and \mathbf{d} are symmetric tensors whose proper values are positive, and \mathbf{R} is an orthogonal tensor, i.e., the relations

$$\mathbf{D} = \mathbf{D}^T, \quad \mathbf{d} = \mathbf{d}^T, \quad \mathbf{R}\mathbf{R}^T = \boldsymbol{\delta} \quad (1.27)$$

hold. The decomposition is unique.

By analyzing the deformation gradient tensor on the ground of polar decomposition, it is seen that \mathbf{R} shows the rotation while \mathbf{D} and \mathbf{d} , respectively, the local deformation in the sense that in a rotation-free case \mathbf{R} , in a strain-free case \mathbf{D} and \mathbf{d} equal the unit tensor. Indeed, at a motion without strain, the deformation gradient is orthogonal, i.e., on the basis of polar decomposition

$$\mathbf{x} = \mathbf{Q} = \mathbf{R}\boldsymbol{\delta} = \boldsymbol{\delta}\mathbf{R} = \mathbf{R} \quad (1.28)$$

can be written.

To simplify the notation, we use the following conventions:

- a) the indices denoted by minuscules refer to x coordinates; those denoted by majuscules refer to X coordinates.
- b) in every case when an index occurs twice in an expression we omit the sign of summation \sum and sum up from 1 to 3 for the dummy index (Einstein convention).
- c) the indices following the comma mean the partial derivatives with respect to the relevant variable.

Now study the case where the deformation gradient tensor does not depend on position, and the orthogonal factor received during polar decomposition corresponds to the unit tensor.

Then, according to (1.25), we can write the motion as

$$\mathbf{x} = \mathbf{a}(t) + \mathbf{D}(t)\mathbf{X} \quad (1.29)$$

and after a transformation to principal axis as

$$\begin{aligned} x_1 &= a_1 + D_1 X_1, \\ x_2 &= a_2 + D_2 X_2, \\ x_3 &= a_3 + D_3 X_3. \end{aligned} \quad (1.30)$$

where D_1 , D_2 and D_3 indicate the eigenvalues of the tensor \mathbf{D} . This means that, apart from translations determined by a_1 , a_2 , a_3 , stretchings parallel to the proper vectors have also appeared. We should image the whole process so that, first, we realize the three translations in arbitrary sequence and, afterwards, the stretching. So, in case of homogeneous deformations, when the function (1.4) can be given in form

$$\mathbf{x} = \mathbf{a}(t) + \mathbf{R}(t)\mathbf{D}(t)\mathbf{X}, \quad (1.31)$$

the instantaneous deformation is defined by the tensor \mathbf{D} . The motion to the present configuration can be decomposed to a succession of deformation, rotation

and translation. The decomposition in equation (1.31) corresponds to this succession: \mathbf{D} defines the deformation, \mathbf{R} the rotation and \mathbf{a} the translation. Of course, the order of the steps of the motion is important as can be seen

$$\begin{aligned}\mathbf{r} &= \mathbf{a} + \mathbf{R}\mathbf{D}\mathbf{X} = \mathbf{a} + d\mathbf{R}\mathbf{X} = \mathbf{R}\mathbf{D}(\mathbf{A} + \mathbf{X}) = \\ &= d\mathbf{R}(\mathbf{A} + \mathbf{X}) = \mathbf{R}(\mathbf{a}^* + \mathbf{D}\mathbf{X}) = d(\mathbf{A}^* + \mathbf{R}\mathbf{X}).\end{aligned}\quad (1.32)$$

Here we have introduced the notations

$$\mathbf{A} = \mathbf{D}^{-1}\mathbf{R}^T\mathbf{a}, \quad \mathbf{a}^* = \mathbf{R}^T\mathbf{a}, \quad \mathbf{A}^* = \mathbf{R}\mathbf{D}^{-1}\mathbf{R}^T\mathbf{a}$$

as well as having used the identity

$$d = \mathbf{R}\mathbf{D}\mathbf{R}^T$$

due to polar decomposition. We mention that tensor \mathbf{D} changes during motion, as do the directions of its proper vectors which means we can regard the foregoing as an instantaneous exposure. The considerations on homogeneous deformation can be applied for arbitrary motion. For that, only a sufficiently small neighborhood of a selected position \mathbf{X}_0 should be regarded.

It is important to mention that the resultant of two sequential deformations without rotation is generally not a strain without rotation as the product of two symmetric tensors is generally not symmetric. The situation is quite different if the deformations are small, i.e., when the tensor \mathbf{D} describing the deformations slightly differs from the unit tensor. In this case it is advisable to introduce the notation

$$\mathbf{D} = \boldsymbol{\delta} + \mathbf{E}. \quad (1.33)$$

Tensor \mathbf{E} should be called the deformation tensor, as it is the zero tensor in the case of motions without strain. In case of two sequential deformations without rotation, we get the relation

$$\mathbf{x} = (\boldsymbol{\delta} + \mathbf{E}_1)(\boldsymbol{\delta} + \mathbf{E}_2) \approx \boldsymbol{\delta} + \mathbf{E}_1 + \mathbf{E}_2 \quad (1.34)$$

for the resultant deformation gradient, if we neglect the product $\mathbf{E}_1\mathbf{E}_2$ due to its smallness. The sum of two symmetric tensors is, however, also symmetric, so we can regard the resultant deformation as the sum of two strains. The formula (1.34) can be applied even in cases when the small strains are accompanied by rotations. In this case the resultant deformation gradient is determined from the equation

$$\mathbf{x} = \mathbf{R}_2(\boldsymbol{\delta} + \mathbf{E}_2)\mathbf{R}_1(\boldsymbol{\delta} + \mathbf{E}_1) = \mathbf{R}_2\mathbf{R}_1(\boldsymbol{\delta} + \mathbf{R}_1^T\mathbf{E}_2\mathbf{R}_1 + \mathbf{E}_1) \quad (1.35)$$

which shows that the deformation tensors can be added even now; however, due to the rotations, a common reference-frame should be provided. The additivity of small deformations tempts one to believe that any deformation can be composed of small deformations. Although this misconception is very common in the old theory of infinitesimal deformations, it is, nevertheless, false as the sum of the terms neglected from (1.34) is not negligible anymore in case of finite resultant.