

Turbulent Dufour effect and Onsager-type relations

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(Received 20 January 1999)

In this paper we extend the work of Elperin *et al.* [Phys. Rev. Lett. **80**, 69 (1998)] by showing that a turbulent Dufour-type effect is also present in chemically nonreacting gaseous admixtures. This result is used to analyze the possibility of obtaining turbulent analogs of the Onsager relations, a fundamental result for molecular crossed effects in irreversible thermodynamics. [S1063-651X(99)11708-2]

PACS number(s): 47.27.Qb, 05.70.Ln

I. INTRODUCTION

In recent times the possibility of turbulent cross-type effects has been considered in two different approaches [1–6]. In the approach taken by Elperin *et al.* [2–4] there is, in the case of chemically nonreacting admixtures (and the absence of phase transitions), a noticeable asymmetry between the turbulent Soret and Dufour effects. As shown in Refs. [3] and [4], a turbulent Soret effect is present in both reacting and nonreacting admixtures; however, in this approach the turbulent Dufour effect only exists for chemically reacting admixtures. In this paper we want to show that this result is due to the approximation used in Ref. [4]. When the terms neglected in this approximation are taken into account, we obtain a turbulent Dufour effect even in chemically nonreacting admixtures. This analysis is presented in Sec. II.

This result is important because it extends to turbulent crossed effects the important symmetry found in the Soret and Dufour effects in the molecular case. Probably the most relevant consequence of this symmetry in the molecular case is the existence of the Onsager relations between the coefficients of the Soret and Dufour effects [7,8]. These relations have a deep physical meaning, as they reflect the time-reversal invariance of the underlying microscopic dynamics in the macroscopic level. Using the results of Sec. II (simultaneous existence of turbulent Soret and Dufour effects), we see that these turbulent effects also play a symmetric role in the turbulent case. It is then interesting to study if this symmetric role also implies the existence of some type of relationship between the coefficients of the turbulent fluxes. We shall show in Sec. III that the coefficients of the turbulent Soret and Dufour effects indeed obey a relation of the Onsager type. Finally, in the Discussion we shall analyze the main implications of these results.

II. TURBULENT DUFOUR EFFECT IN NONREACTING ADMIXTURES

Let us consider a mixture of chemically nonreacting gases advected by a compressible turbulent flow. The number density n_i of the components in the admixture and the temperature field T obey the evolution equations

$$\frac{\partial n_i}{\partial t} + \vec{v}_i \cdot \nabla n_i + n_i \nabla \cdot \vec{v}_i = \nabla \cdot (\kappa_i \nabla n_i) \quad (1)$$

and

$$\frac{\partial T}{\partial t} + \vec{v} \cdot \nabla T + (\gamma - 1) T \nabla \cdot \vec{v} = \nabla \cdot (\kappa \nabla T), \quad (2)$$

where \vec{v}_i is the random velocity field of the i th component of the admixture in the turbulent fluid velocity field \vec{v} , γ is the specific-heat ratio, κ_i is the coefficient of molecular diffusion of component i , and κ is the coefficient of molecular thermal diffusivity.

It is assumed that the densities of gaseous admixtures are much smaller than the total fluid density ρ . The continuity equation for the fluid is

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0. \quad (3)$$

We do not take into account the diffusion of the fluid in the gaseous admixtures because $n_i \ll n$.

Now, we want to derive the equations for the mean concentration of the gaseous components of the admixture and temperature. In order to obtain these equations, Eqs. (1) and (2) must be averaged over the ensemble of random velocity fluctuations. Following Refs. [2–4], this is done using the stochastic calculus. In the stochastic calculus the solution of the equation

$$\frac{\partial \eta}{\partial t} + \vec{v} \cdot \nabla \eta + A \eta \nabla \cdot \vec{v} = \nabla \cdot (\kappa_\eta \nabla \eta) \quad (4)$$

with the initial condition $\eta(t=t_0, \vec{x}) = \eta_0(\vec{x})$ is given by

$$\eta(t, \vec{x}) = M[G(t, t_0) \eta_0(\vec{\xi}(t, t_0))], \quad (5)$$

where

$$G(t, t_0) = \exp\left(-\int_{t_0}^t B(\sigma, \vec{\xi}(t, \sigma)) d\sigma\right) \quad (6)$$

with

$$B = A \nabla \cdot \vec{v} \quad (7)$$

and $M[\]$ the mathematical expectation over the Wiener paths $\vec{\xi}$,

$$\tilde{\xi}(t, t_0) = \vec{x} - \int_0^{t-t_0} \tilde{\mathbf{v}}(t_s, \tilde{\xi}_s) ds + (2\kappa_\eta)^{1/2} \tilde{\mathbf{w}}(t-t_0), \quad (8)$$

where $t_s = t - s$, $\tilde{\xi}_s = \tilde{\xi}(t, t-s)$, and $\tilde{\mathbf{w}}$ is the Wiener random process.

The validity of these relations can be easily verified. Following Ref. [3], one obtains for $\eta(t+\Delta t, \vec{x})$ the following expression (accurate up to $\sim \Delta t$ terms):

$$\eta(t+\Delta t, \vec{x}) = \eta(t, \vec{x}) - \left(V_m \frac{\partial \eta}{\partial x_m} + B \eta - \kappa_\eta \Delta \eta \right) \Delta t. \quad (9)$$

Now, calculating the limit of this expression for $\Delta t \rightarrow 0$, we recover Eq. (4).

Using a similar procedure, we can derive the equation for the mean value of the field, $\Lambda = \langle \eta \rangle$. Following again the procedure of Ref. [3], we obtain the following expression [using now an expression accurate up to $\sim (\Delta t)^2$ terms]:

$$\begin{aligned} \frac{\partial \Lambda}{\partial t} + \{ [\tilde{V} - 2A \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle - \langle \tau(\tilde{\mathbf{u}} \cdot \nabla) \tilde{\mathbf{u}} \rangle] \cdot \nabla \} \Lambda \\ = \Lambda [-\nabla \cdot \tilde{V} + A \langle \tau(\tilde{\mathbf{u}} \cdot \nabla)(\nabla \cdot \tilde{\mathbf{u}}) \rangle + A^2 \langle \tau(\nabla \cdot \tilde{\mathbf{u}})^2 \rangle] \\ + D_{pm}^* \frac{\partial^2 \Lambda}{\partial x_p \partial x_m}, \end{aligned} \quad (10)$$

where

$$\tilde{\mathbf{v}} = \tilde{V} + \tilde{\mathbf{u}} \quad (11)$$

with $\tilde{V} = \langle \tilde{v} \rangle$ the mean velocity and $\tilde{\mathbf{u}}$ the turbulent component of the velocity. The parameter τ is the characteristic time of turbulent motions. The expression for the matrix D is

$$D_{pm} = \kappa_\eta \delta_{pm} + \langle \tau u_p u_m \rangle. \quad (12)$$

In a straightforward way we can obtain the following relations:

$$\begin{aligned} \tilde{V} - 2A \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle - \langle \tau(\tilde{\mathbf{u}} \cdot \nabla) \tilde{\mathbf{u}} \rangle \\ = \tilde{V}_{\text{eff}} - (A-1) \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle - \frac{\partial}{\partial x_p} \langle \tau u_p u_m \rangle \end{aligned} \quad (13)$$

and

$$\begin{aligned} A \langle \tau(\tilde{\mathbf{u}} \cdot \nabla)(\nabla \cdot \tilde{\mathbf{u}}) \rangle + A^2 \langle \tau(\nabla \cdot \tilde{\mathbf{u}})^2 \rangle \\ = A \nabla \cdot \langle \tau \tilde{\mathbf{u}}(\nabla \cdot \tilde{\mathbf{u}}) \rangle + A(A-1) \langle \tau(\nabla \cdot \tilde{\mathbf{u}})^2 \rangle, \end{aligned} \quad (14)$$

where

$$\tilde{V}_{\text{eff}} = \tilde{V} - A \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle. \quad (15)$$

Using Eqs. (13) and (14) we easily obtain the equation for the mean field:

$$\begin{aligned} \frac{\partial \Lambda}{\partial t} + \frac{\partial}{\partial x_p} \left(\Lambda (\tilde{V}_{\text{eff}})_p - D_{pm}^* \frac{\partial \Lambda}{\partial x_m} \right) \\ = (A-1) \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle \cdot \nabla \Lambda + A(A-1) \Lambda \langle \tau(\nabla \cdot \tilde{\mathbf{u}})^2 \rangle. \end{aligned} \quad (16)$$

Note that when $A=1$, Eq. (16) becomes a conservation law and we recover the expression obtained in [3]. The first term on the right-hand side of Eq. (16) represents an advection of the mean field Λ by the ‘‘turbulent velocity’’ $\langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle$. On the other hand, the second term is only present when $A \neq 1$, i.e., in the case of the temperature equation, becoming null in the case of the concentration (that is a passive scalar). When $A > 1$, this term leads to a local exponential growth of the mean field in regions of strong convergence or divergence.

Using these expressions, the equations for the mean number density and temperature fields become

$$\frac{\partial N_i}{\partial t} + \frac{\partial}{\partial x_p} \left((\tilde{V}_{\text{eff}}^i)_p N_i - D_{pm}^i \frac{\partial N_i}{\partial x_m} \right) = 0 \quad (17)$$

and

$$\begin{aligned} \frac{\partial \bar{T}}{\partial t} + \frac{\partial}{\partial x_p} \left((\tilde{V}_{\text{eff}}^i)_p \bar{T} - D_{pm}^* \frac{\partial \bar{T}}{\partial x_m} \right) \\ = (\gamma-2) \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle \cdot \nabla \bar{T} + (\gamma-1)(\gamma-2) \bar{T} \langle \tau(\nabla \cdot \tilde{\mathbf{u}})^2 \rangle, \end{aligned} \quad (18)$$

where $N_i = \langle n_i \rangle$, $\bar{T} = \langle T \rangle$, $D_{pm}^i = \kappa_i \delta_{pm} + \langle \tau(\tilde{u}_i)_p (\tilde{u}_i)_m \rangle$, $D_{pm}^* = \kappa \delta_{pm} + \langle \tau(\tilde{u})_p (\tilde{u})_m \rangle$, $\tilde{V}_{\text{eff}}^i = \tilde{V}_i - \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{u}_i \rangle$, and $\tilde{V}_{\text{eff}} = \tilde{V} - (\gamma-1) \langle \tau(\nabla \cdot \tilde{\mathbf{u}}) \tilde{\mathbf{u}} \rangle$.

Note that the time parameter τ characterizing the turbulent motions is the same for the components of the admixture and the fluid because the turbulent velocity of the components of the admixture coincides with that of the surrounding fluid [3].

These equations are the basis for the introduction of the crossed effects. In the case of low Mach numbers, Eq. (3) reduces to [3]

$$\nabla \cdot \tilde{\mathbf{u}} \approx -\frac{1}{\bar{\rho}} (\tilde{\mathbf{u}} \cdot \nabla) \bar{\rho}, \quad (19)$$

where $\bar{\rho} = \langle \rho \rangle$ is the mean fluid density.

Note that Eq. (19) can be rewritten using the turbulent velocity of any of the admixtures, $\nabla \cdot \tilde{\mathbf{u}}_i = -(\bar{\rho})^{-1} (\tilde{\mathbf{u}}_i \cdot \nabla) \bar{\rho}$, because, as remarked earlier, the turbulent velocity of the admixtures coincides with that of the surrounding fluid [3].

On the other hand, using the equation of state for the mean fields $\bar{P} \sim \bar{\rho} \bar{T}$ (P is the pressure), we have $(\bar{T})^{-1} \nabla \bar{T} - (\bar{P})^{-1} \nabla \bar{P} = -(\bar{\rho})^{-1} \nabla \bar{\rho}$. Using this expression, Eq. (17) can be rewritten as

$$\frac{\partial N_i}{\partial t} + \nabla \cdot (\tilde{J}_i^c + \tilde{J}_i) = 0, \quad (20)$$

where \tilde{J}_i^c , the turbulent crossed Soret effect, is defined as

$$\tilde{J}_i^c = -\frac{N_i}{\bar{T}} \langle \tau \tilde{u}_i \tilde{u}_i \rangle \cdot \nabla \bar{T} \quad (21)$$

and \tilde{J}_i contains the remaining terms, i.e., the direct flux and the turbulent barodiffusion [3].

Let us consider now the turbulent crossed term for Eq. (18). This equation can be rewritten as

$$\frac{\partial \bar{T}}{\partial t} + \nabla \cdot (\vec{J}^c + \vec{J}) = (\gamma - 2) \langle \tau (\nabla \cdot \vec{u}) \vec{u} \rangle \cdot \nabla \bar{T} + (\gamma - 1)(\gamma - 2) \bar{T} \langle \tau (\nabla \cdot \vec{u})^2 \rangle, \quad (22)$$

where the crossed part of the flux is

$$\vec{J}^c = \frac{\gamma - 1}{\bar{\rho}} \langle \tau \vec{u} \vec{u} \rangle \cdot \nabla \bar{\rho}. \quad (23)$$

On the other hand, \vec{J} contains the other terms, which will not be considered in this paper. The density of the fluid is the sum of the densities of the different components, $\rho = \sum_i \rho_i$, which implies $\bar{\rho} = \sum_i \bar{\rho}_i$. Remembering the definition of the number density of every component $n_i = \rho_i / m_i$ (with m_i the mass of the molecules of component i), Eq. (22) transforms into

$$\vec{J}^c = \sum_i \frac{(\gamma - 1) m_i}{\bar{\rho}} \langle \tau \vec{u} \vec{u} \rangle \cdot \nabla N_i. \quad (24)$$

Equation (24) shows clearly the existence of a turbulent Dufour effect: the mean gradients of the number density induce heat fluxes. This effect is even present in the absence of chemical reactions. As in the case of the turbulent Soret effect in chemically nonreacting admixtures [3], the compressibility of the fluid results in a new heat flux caused by turbulent transport when gradients of the number density of particles are present.

In Ref. [4] the authors have not obtained this term due to the approximations introduced in the analysis, namely to neglect their term $(\gamma - 1) T_f \nabla \cdot \vec{v}$. It is simple to show that their analysis, taking into account the neglected term, also gives the term represented in Eq. (24).

Note that the turbulent Dufour effect in the absence of chemical reactions (or phase transitions) is a small effect as compared with the Dufour effect in the presence of chemical reactions. Indeed, the density of the admixture is much less than the density of the complete fluid, and the ratio of these densities is a small parameter in the problem. The order of magnitude of these heat fluxes in Eq. (24) must be proportional to this small parameter. Consequently, the turbulent Dufour effect in the absence of chemical reactions is small compared to the same effect in the presence of the reactions, because in the second case the effect does not depend on this type of small parameter. In spite of the smallness of the new effect, we shall show in the next section that it can be of much interest to study the possible extension to turbulence of some relations usual within irreversible thermodynamics (in addition to the intrinsic interest of the existence of the effect).

III. TURBULENT ONSAGER-TYPE RELATIONS

In the preceding section we have shown the existence of a turbulent Dufour effect for chemically nonreacting systems. This result is important because it extends the symmetric role played by the Soret and Dufour effects in the molecular

regime to the turbulent regime. The most relevant consequence of this symmetric role in molecular theory is the existence of reciprocal relations between the crossed coefficients, the well-known Onsager relations [7,8]. These relations are one of the fundamental results in irreversible thermodynamics. Now that we know that the symmetry between the Soret and Dufour effects can be extended to turbulence, we must investigate to see whether some type of extension of the Onsager relations is also possible in turbulence. In this section we shall show that it is indeed possible. Moreover, in contrast with the theory of irreversible processes, we shall show that it is possible to obtain this result without referring to the microscopic dynamics of the system.

In the beginning, we shall review the Onsager relations for the Soret and Dufour effects in the theory of irreversible processes [8]. In the molecular regime, the heat and mass fluxes in a binary mixture are

$$\vec{J}_q = -\lambda \nabla T - \rho_1 \mu_{11}^c T D'' \nabla c_1 \quad (25)$$

and

$$\vec{J}_{m1} = -\rho c_1 c_2 D' \nabla T - \rho D \nabla c_1. \quad (26)$$

In these equations c_i refers to the concentration of component i , μ to the chemical potential, λ to the heat conductivity, D' to the Dufour coefficient, D'' to the thermal diffusion coefficient, and D to the diffusion coefficient. The Onsager relations establish in this case that the crossed coefficients are equal, $D' = D''$. These relations have been subjected to severe experimental scrutiny, confirming their validity within reasonable limits of experimental errors.

The turbulent crossed fluxes can be expressed as

$$(\vec{J}_i^c)_k = -C_{k1}^S \frac{N_i}{\bar{T}} \frac{\partial \bar{T}}{\partial x_1} \quad (27)$$

and

$$(\vec{J}_c)_k = C_{k1}^D (\gamma - 1) \frac{m_i}{\bar{\rho}} \frac{\partial N_i}{\partial x_1} \quad (28)$$

with the turbulent crossed coefficients given by

$$C_{k1}^S = \langle \tau (\vec{u}_i)_k (\vec{u}_i)_1 \rangle \quad (29)$$

and

$$C_{k1}^D = \langle \tau (\vec{u})_k (\vec{u})_1 \rangle. \quad (30)$$

Note that in the turbulent case the crossed coefficients are not scalars but the components of two tensors. This is also true in the molecular case when we consider nonisotropic systems.

The above choice of the turbulent crossed coefficients can be justified by analogy with the molecular case. The coefficients must express the strength of the flux and are numerical values independent of the actual values of the thermodynamic variables $(\rho, T, n_i, \vec{u}_i, \vec{u})$. Moreover, the coefficients cannot depend on thermodynamic parameters (γ, m_i) because these parameters refer to fluid properties, and the strength is a flow property. When these parameters are not

constant, but functions of the fundamental thermodynamic variables, for instance $\gamma = \gamma(T)$, they play the same role as the thermodynamic variables. The coefficients are obtained as mean properties of the flow velocity. Note that the turbulent coefficients have the dimension L^2T^{-1} , as do the thermal diffusivity and the diffusion coefficient. This is in contrast to the crossed molecular coefficients, which have dimension $L^2T^{-1} \text{ deg}^{-1}$.

Direct inspection of Eqs. (29) and (30) shows that in the case $\vec{u}_i = \vec{u}$, which as we have remarked earlier is the situation we are considering, we obtain

$$C_{k1}^S = C_{k1}^D. \quad (31)$$

These relations are the turbulent extension of the Onsager relations.

Note that the equality $\vec{u}_i = \vec{u}$, giving rise to the turbulent Onsager relations, does not, however, arise in a trivial way. For instance, in the case of small inertial particles, the fluctuating component of the velocity of the particle and the turbulent velocity of the fluid are, in general, different [2].

The extended Onsager relation is even valid when the turbulent Soret effect is relatively large and the turbulent Dufour effect very small. This case is also sometimes present in molecular flows when the Soret flux is easy to measure and the Dufour effect very hard to detect, but the Onsager relations remain valid.

There is a fundamental difference in the derivation of the relations between the molecular and turbulent cases. In the first case, all the derivations are based on microscopic theories, whereas in the second case the derivation is of a completely macroscopic nature. Let us briefly review the derivation in the molecular case [7,8]. Under the conditions that (i) the fluxes are identified as time rates of state variables, (ii) the forces are identified as the derivatives of the entropy with respect to the state variables, and (iii) there exists between these so-defined fluxes and forces linear constitutive relations, then the coefficients of these linear relations obey the Onsager reciprocity relations. This result was shown to be a consequence of the time-reversal invariance of the microscopic dynamics. A crucial point in the derivation of the Onsager relations is that regressions of fluctuations are assumed to follow the same linear dynamical laws as the macroscopic equations (this assumption has been criticized by many authors, see Ref. [9] and references therein). Although various proofs of the Onsager reciprocal relations have been proposed so far, all of them are based on microscopic theories, statistical mechanics of fluctuations, or kinetic theory. According to some authors [9], the absence of a macroscopic derivation of the relations is a serious drawback. These authors have expressed the opinion that, unless a complete macroscopic proof of the relations is proposed, one should regard them as postulates at the macroscopic level. The situation is completely different in the turbulent example presented here. The derivation of the turbulent Onsager-type relations is of a macroscopic nature. It does not rest upon assumptions on the microscopic dynamics of the system; we only deal with the macroscopic equations of the system. An important advantage is that we do not need to assume, as in

the molecular case, a particular behavior for the regression of the fluctuations. Moreover, the time-reversal invariance of the microscopic dynamics does not play any role in our derivation. Finally, none of the necessary conditions (i)–(iii) in the theory of irreversible processes is fulfilled. The last point emerges in a natural way because these conditions represent the behavior of fluxes close to the thermodynamic equilibrium, whereas turbulent flows are systems which are very far from the thermodynamic equilibrium.

The above considerations show that although Eq. (31) is the turbulent analog of the molecular Onsager relations, the physical basis behind both relations is very different.

IV. DISCUSSION

In this paper we have extended the results of Elperin *et al.* [2–4] by showing the existence of a turbulent Dufour effect even in the case of chemically nonreacting admixtures (and in the absence of phase transitions). Although, as we have remarked earlier, this effect is small in comparison with other crossed effects (for instance the turbulent Dufour effect in chemically reacting admixtures), the importance of the new effect is twofold.

(a) The turbulent Soret and Dufour effects recover the symmetric role played by their molecular counterparts.

(b) It can be used to show the existence of a turbulent analog of the Onsager reciprocal relations, typical of irreversible thermodynamics.

These two points are important because they show, in a particular example, that some fundamental results of the theory of irreversible processes have turbulent analogs, although turbulent flows take place very far from the thermodynamic equilibrium. These results suggest the possibility of generalizing in some sense the theory of irreversible processes to physical regions far from the equilibrium as, for instance, turbulent flows. Moreover, these results provide clues to possible ways of exploring such generalizations.

Another important characteristic of the example of turbulent Onsager-type relations presented in this paper is that they have been derived from a macroscopic framework, in contrast to the microscopic derivations of the molecular relations. As previously stressed, several authors have indicated the necessity of macroscopic derivations of these relations to avoid the difficulties present in the microscopic derivations, in particular the necessity of assuming that the regressions of fluctuations follow the same linear dynamical laws as the macroscopic equations.

By no means do the above considerations on the importance of a macroscopic derivation of the relations suggest that we underestimate the role of a possible microscopic derivation of the turbulent relations. A microscopic derivation would allow us to understand the role of the time-reversal invariance of the microscopic dynamics in the turbulent relations. However, this approach is problematic as no microscopic theory of such a general character, as exists for equilibrium phenomena, is available for nonequilibrium processes [8].

The turbulent Onsager-type relations derived in this paper are, in principle, experimentally verifiable. An experimental verification of the relations would be an important support

for the theory of turbulent crossed fluxes. On the other hand, it would also be important to study the validity of the relations when the restriction to low Mach numbers [Eq. (19) is valid under these restriction] is relaxed. We must also study the validity of these types of relations in the case of other turbulent crossed fluxes, for instance the Soret and Dufour effects in chemically reacting admixtures. However, the experimental verification of the relations in these more general

cases would be more difficult due to the superposition of different effects.

ACKNOWLEDGMENTS

This work has been partially supported by the Spanish Ministry of Education and Science under Contract No. PB96.0451. I am grateful to Professor Tov Elperin for discussions on the problem.

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