

Turbulent Thermal Diffusion of Small Inertial Particles

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A new physical effect, turbulent thermal diffusion, is discussed. This phenomenon is related to the dynamics of small inertial particles in incompressible turbulent fluid flow. At large Reynolds and Peclet numbers the turbulent thermal diffusion is much stronger than the molecular thermal diffusion. It is shown that inertia of particles under certain conditions can cause a large-scale instability of spatial distribution of particles. Inertial particles are concentrated in the vicinity of the minimum (or maximum) of the mean temperature of the surrounding fluid depending on the ratio of material particle density to that of the surrounding fluid.

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The main purpose of this Letter is to discuss a new physical effect, i.e., the turbulent thermal diffusion. The phenomenon of molecular thermal diffusion in gases was predicted by Enskog and confirmed experimentally by Chapman and Dootson long ago [1] (see also [2]). The equation for the number density n_p of particles taking into account this effect reads $\partial n_p / \partial t = -\vec{\nabla} \cdot \mathbf{J}_M$, where the flux of particles \mathbf{J}_M is given by $\mathbf{J}_M = -D[\vec{\nabla} n_p + k_t \vec{\nabla} T_f / T_f]$. The first term in the formula for the flux of particles describes molecular diffusion, while the second term accounts for the flux of particles caused by the temperature gradient $\vec{\nabla} T_f$ (molecular thermal diffusion). Here D is the coefficient of molecular diffusion, $k_t \propto n_p$ is the thermal diffusion ratio, and $D_M = Dk_t$ is the coefficient of thermal diffusion.

When the velocity field of a surrounding fluid is nonzero and turbulent, the nature of diffusion drastically changes at large Reynolds and Peclet numbers. It was demonstrated by Taylor that turbulence results in a sharp increase of the effective diffusion coefficient [3]. Detailed discussion of turbulent transport can be found in [4]. Recently it was shown that the contribution of molecular thermal diffusion in gases into the total mass transfer rate can be significant even in a turbulent flow [5].

In this Letter it is shown that in a turbulent fluid flow with a nonzero mean temperature gradient there appears an additional mean flux of particles in the direction opposite to that of the mean temperature gradient. For large Reynolds and Peclet numbers the turbulent thermal diffusion is much stronger than the molecular thermal diffusion. Evolution of the number density $n_p(t, \mathbf{r})$ of small particles in a turbulent flow is determined by

$$\frac{\partial n_p}{\partial t} + \vec{\nabla} \cdot (n_p \mathbf{v}_p) = -\vec{\nabla} \cdot \mathbf{J}_M, \quad (1)$$

where \mathbf{v}_p is a random velocity field of the particles which they acquire in a turbulent fluid velocity field. We

consider the case of large Reynolds and Peclet numbers. The velocity of particles \mathbf{v}_p depends on the velocity of the surrounding fluid, and it can be determined from the equation of motion for a particle. This equation represents a balance of particle inertia with the fluid drag force produced by the motion of the particle relative to the surrounding fluid. Solution of the equation of motion for small particles with $\rho_p \gg \rho$ yields [6]

$$\mathbf{v}_p = \mathbf{v}(t, \mathbf{Y}(t)) - \tau_p \left[\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \vec{\nabla}) \mathbf{v} \right] + O(\tau_p^2), \quad (2)$$

where \mathbf{v} is the velocity of the surrounding fluid, $\mathbf{Y}(t)$ is the position of the particle, τ_p is the characteristic time of coupling between the particle and surrounding fluid (Stokes time), ρ_p is the material density of particles, and ρ is the density of the fluid. For instance, for spherical particles of radius a_* the Stokes time is $\tau_p = m_p / 6\pi a_* \rho \nu$, where ν is the kinematic viscosity of the surrounding fluid and m_p is the particle mass. The second term in (2) describes the difference between the local fluid velocity and particle velocity arising due to the small but finite inertia of the particle.

In this study we consider incompressible turbulent flow $\vec{\nabla} \cdot \mathbf{v} = 0$. However, the velocity field of particles is assumed to be compressible, i.e., $\vec{\nabla} \cdot \mathbf{v}_p \neq 0$. Indeed, Eq. (2) for the velocity of particles and Navier-Stokes equation for the fluid yield $\vec{\nabla} \cdot \mathbf{v}_p = -\tau_p \vec{\nabla} \cdot d\mathbf{v}/dt = \tau_p \vec{\nabla} \cdot \vec{\nabla} P / \rho$. We study the large-scale dynamics of small inertial particles and average Eq. (1) over an ensemble of random velocity fluctuations. For this purpose we use the stochastic calculus which has been previously employed in magnetohydrodynamics [7,8] and in the problems of passive scalar transport in incompressible [7,9] and compressible [10] turbulent flows. The main object of the stochastic calculus is a Wiener random process defined by the properties $M\{\mathbf{w}\} = 0$ and $M\{w_i w_j\} = t \delta_{ij}$, where $M\{\cdot\}$ denotes the mathematical expectation over the Wiener paths.

Within the stochastic calculus the solution of Eq. (1) is reduced to the analysis of the evolution of the concentration field $n_p(t, \mathbf{r})$ along the Wiener path, $\vec{\xi}$:

$$\vec{\xi}(t, t_0) = \mathbf{x} - \int_0^{t-t_0} \mathbf{v}_p[t_s, \vec{\xi}(t, t_s)] ds + (2D)^{1/2} \mathbf{w}(t - t_0), \quad (3)$$

where $t_s = t - s$. Equation (3) describes a set of random trajectories which pass through the point \mathbf{x} at time t . Because $\mathbf{w}(t)$ is a Wiener process, the initial coordinates of every trajectory $\vec{\xi}$ are random. It can be proved (see [7,10]) that the solution of Eq. (1) with the initial condition $n_p(t = t_0, \mathbf{x}) = n_0(\mathbf{x})$ is given by $n_p(t, \mathbf{x}) = M\{G(t, t_0)n_0[\vec{\xi}(t, t_0)]\}$, where the Green function is $G(t, t_0) = \exp[-\int_{t_0}^t b_*(\sigma, \vec{\xi}(t, \sigma)) d\sigma]$ and $b_* \equiv \vec{\nabla} \cdot \mathbf{v}_p$. Hereafter we neglect small molecular thermal diffusion (see below). It can be taken into account in mean velocity of particles. The use of the technique described in [7,10] allows us to derive equations for the mean field $N = \langle n_p \rangle$:

$$\frac{\partial N}{\partial t} + \vec{\nabla} \cdot [N \mathbf{V}_{\text{eff}} - \hat{D} \vec{\nabla}_m N] = 0, \quad (4)$$

where $\hat{D} \equiv D_{pm} = D \delta_{pm} + \langle \tau u_p u_m \rangle$, $\mathbf{V}_{\text{eff}} = \mathbf{V} - \langle \tau b \mathbf{u} \rangle$, and $\mathbf{v}_p = \mathbf{V} + \mathbf{u}$, $\mathbf{V} = \langle \mathbf{v}_p \rangle$ is the mean velocity, \mathbf{u} is the random component of the velocity of particles, $b = \vec{\nabla} \cdot \mathbf{u}$, and τ is the momentum relaxation time of random velocity field \mathbf{u} , which depends on scale of turbulent motion. We use here for simplicity the δ correlated in time random process to describe a turbulent velocity field. However, the results remain valid also for the velocity field with a finite correlation time, if the mean number density of the particles varies slowly in comparison with the correlation time of the turbulent flow (see, e.g., [11]). Equation (4) was derived for $\text{Pe} \gg 1$. It can be shown that for $\text{Pe} \ll 1$ and arbitrary velocity field the equation for the mean field coincides with Eq. (4). Now we derive an equation for N^2 . Multiplication of Eq. (4) by N and simple manipulations yield

$$\frac{\partial N^2}{\partial t} + (\vec{\nabla} \cdot \mathbf{S}) = -N^2 (\vec{\nabla} \cdot \mathbf{V}_{\text{eff}}) - I_D,$$

where $\mathbf{S}_m = N^2 (\mathbf{V}_{\text{eff}})_m - D_{mp} \vec{\nabla}_p N^2$ and $I_D = 2D_{mp} (\vec{\nabla}_m N) (\vec{\nabla}_p N)$. The latter equation implies that if $\vec{\nabla} \cdot \mathbf{V}_{\text{eff}} < 0$, a perturbation of the equilibrium distribution of inertial particles can grow in time, i.e., $(\partial/\partial t) \int N^2 d^3r > 0$. However, the total number of particles is conserved. Therefore the growth of N^2 when $\vec{\nabla} \cdot \mathbf{V}_{\text{eff}} < 0$ is accompanied by formation of an inhomogeneous spatial distribution of the inertial particles whereby regions with an increased concentration of particles coexist with regions depleted from particles.

Now we calculate the velocity \mathbf{V}_{eff} . Using the equation of state $P = \kappa T_f \rho / m_\mu$ we obtain the expression for $b_* \equiv \vec{\nabla} \cdot \mathbf{v}_p$:

$$b_* = \frac{\tau_p v_T^2}{T_*} \left[\Delta T_f + T_f \frac{\Delta \rho}{\rho} + \left(\frac{\vec{\nabla} \rho}{\rho} \right) \cdot \vec{\nabla} T_f - \left| \frac{\vec{\nabla} \rho}{\rho} \right|^2 T_f \right], \quad (5)$$

where $v_T^2 = \kappa T_* / m_\mu$, m_μ is the mass of molecules of surrounding fluid, and $T_f(t, \mathbf{r})$ is the temperature field with a characteristic value T_* . Then we use Eq. (5) to calculate $\langle \tau \mathbf{u} b \rangle \approx \langle \tau \mathbf{u} \Delta \Theta \rangle = (\tau_p v_T^2 / T_*) \langle \tau \tilde{\mathbf{u}} \Delta \Theta \rangle$, where Θ are fluctuations of temperature. We neglect the second moments $\sim \langle \tilde{\mathbf{u}} \tilde{\rho} \rangle$, since the mean turbulent flux of mass of the surrounding fluid vanishes in a finite domain surrounded by solid boundaries. Here $\tilde{\rho}$ and $\tilde{\mathbf{u}}$ are fluctuations of the density and velocity of the fluid. On the other hand, the mean turbulent heat flux is nonzero in the presence of an external mean temperature gradient $\vec{\nabla} T \neq 0$ (see below). To obtain an equation for $\langle \tau \mathbf{u} b \rangle$, we take into account that the fluctuating component of the particle velocity \mathbf{u} can be expressed in terms of the turbulent velocity of fluid $\tilde{\mathbf{u}}$: $\mathbf{u} = \tilde{\mathbf{u}} - \tau_p d\tilde{\mathbf{u}}/dt$ [see Eq. (2)]. Note that $\vec{\nabla} \cdot \mathbf{u} \neq 0$, whereas $\vec{\nabla} \cdot \tilde{\mathbf{u}} = 0$. Therefore the velocity \mathbf{V}_{eff} is given by $\mathbf{V}_{\text{eff}} = \mathbf{V} - (\tau_p v_T^2 / T_*) \langle \tau \tilde{\mathbf{u}} \Delta \Theta \rangle$, where we neglect terms $\sim O(\tau_p^2)$. The latter formula shows that \mathbf{V}_{eff} depends on the mean turbulent heat flux $\langle \tilde{\mathbf{u}} \Theta \rangle$ that is determined by the well-known equation $\langle \tilde{\mathbf{u}}(\mathbf{x}) \Theta(\mathbf{x}) \rangle = -\chi_T \vec{\nabla} T$ (see, e.g., [4]), where the total temperature is $T_f = T + \Theta$, $T = \langle T_f \rangle$ is the mean temperature field, $\chi_T \sim u_0 l_0 / 3$ is the coefficient of turbulent thermal conductivity. Note that herein we do not consider the situation with very high gradients when gradient transport assumption is violated. The above formula for the mean turbulent heat flux is written in \mathbf{r} space. The corresponding second moment in \mathbf{k} space is given by $\langle \tilde{u}_m(\mathbf{k}) \Theta(-\mathbf{k}) \rangle = -\tau(k) \langle \tilde{u}_m(\mathbf{k}) \tilde{u}_n(-\mathbf{k}) \rangle \partial T / \partial R_n$, where \mathbf{R} is a large-scale variable and a spectrum of the turbulent velocity field and correlation time $\tau(k)$ can be chosen as Kolmogorov's spectrum:

$$\langle \tilde{u}_m \tilde{u}_n \rangle = \frac{2}{3k_0} \left(\frac{\langle \tilde{u}^2 \rangle}{8\pi k^2} \right) \left(\frac{k}{k_0} \right)^{-5/3} \left(\delta_{mn} - \frac{k_m k_n}{k^2} \right),$$

$$\tau(k) = 2\tau_0 \left(\frac{k}{k_0} \right)^{-2/3},$$

where $k_0 < k < k_0 \text{Re}_*^{3/4}$ (see, e.g., [4]), $\text{Re}_* = \min\{\text{Re}, \text{Pe}_T\}$, $\text{Re} = l_0 u_0 / \nu$ is the Reynolds number, $\text{Pe}_T = l_0 u_0 / \chi$ is the thermal Peclet number, $l_0 = k_0^{-1}$ is the maximum scale of turbulent motions, u_0 is the characteristic velocity in this scale, and χ is the coefficient of molecular thermal conductivity. Multiplying the equation for $\langle \tilde{u}_m(\mathbf{k}) \Theta(-\mathbf{k}) \rangle$ by $-k^2 \tau(k)$ and integrating in \mathbf{k} space we obtain $\langle \tau \tilde{u}_m \Delta \Theta \rangle = \alpha \ln(\text{Re}_*) \vec{\nabla} T$, where $\alpha = 2/3$. Finally we arrive at the following equation for velocity:

$$\mathbf{V}_{\text{eff}} = \mathbf{V} - \frac{\alpha \tau_p v_T^2}{T_*} \ln(\text{Re}_*) \vec{\nabla} T.$$

Equation (4) with this effective velocity \mathbf{V}_{eff} can be rewritten in the form

$$\frac{\partial N}{\partial t} + \vec{\nabla} \cdot (N\mathbf{V}) = -\vec{\nabla} \cdot (\mathbf{J}_T + \mathbf{J}_M), \quad (6)$$

where

$$\mathbf{J}_T = -D_T \left[\frac{k_T}{T} \vec{\nabla} T + \vec{\nabla} N \right], \quad (7)$$

$$k_T = N \frac{3\alpha}{\text{Pe}} \left(\frac{m_p}{m_\mu} \right) \left(\frac{T}{T_*} \right) \ln \text{Re}_*, \quad (8)$$

where $D_T = u_0 l_0 / 3$ is the coefficient of turbulent diffusion, k_T can be interpreted as turbulent thermal diffusion ratio, and $D_T k_T$ is the coefficient of turbulent thermal diffusion. We use here an identity

$$\frac{\tau_p \nu_T^2}{l_0 u_0} = \frac{1}{\text{Pe}} \left(\frac{m_p}{m_\mu} \right),$$

and $\text{Pe} = u_0 l_0 / D_*$ is the Peclet number and the molecular diffusion coefficient $D_* = \kappa T_* / 6\pi a_* \rho \nu$. Note that for $\text{Re}_* \gg 1$ and $\text{Pe} \gg 1$ both turbulent diffusion coefficients are much larger than the corresponding molecular coefficients (i.e., $D_T \gg D$ and $D_T k_T \gg D k_t$).

Now we will show that turbulent thermal diffusion results in large-scale pattern formation, whereby initially spatial distribution of particles in a turbulent incompressible flow of fluid evolves under certain conditions into large-scale inhomogeneous distribution due to excitation of an instability. One of the most important conditions for the instability is inhomogeneous spatial distribution of mean temperature of surrounding fluid. In particular, the instability can be excited in the vicinity of the minimum in the mean temperature distribution.

Now let us discuss the mechanism of the instability. The inertia effect results in $\vec{\nabla} \cdot \mathbf{v}_p \propto \tau_p \Delta P \neq 0$. On the other hand, for large Peclet numbers $\vec{\nabla} \cdot \mathbf{v}_p \propto -dn_p/dt$. This means that in regions with maximum pressure of turbulent fluid (i.e., where $\Delta P < 0$) there is an accumulation of inertial particles (i.e., $dn_p/dt > 0$). Similarly there is an outflow of inertial particles from regions with minimum pressure of fluid. In a homogeneous and isotropic turbulence without large-scale external gradients of temperature a drift from regions with increased (decreased) concentration of inertial particles by a turbulent flow of fluid is equiprobable in all directions. Location of these regions is not correlated with turbulent velocity field. Therefore they do not contribute in large-scale flow of inertial particles.

The situation is drastically changed when there is a large-scale inhomogeneity of the temperature of the turbulent flow. In this case the mean heat flux $\langle \tilde{\mathbf{u}} \Theta \rangle \neq 0$. Therefore fluctuations of both temperature and velocity of fluid are correlated. Fluctuations of temperature cause fluctuations of pressure of fluid. The pressure fluctuations result in fluctuations of the concentration of inertial

particles. Indeed, an increase (decrease) of the pressure of the surrounding fluid is accompanied by an accumulation (outflow) of the particles. Therefore the direction of mean flux of particles coincides with that of heat flux, i.e., $\langle \tilde{\mathbf{u}} n \rangle \propto \langle \tilde{\mathbf{u}} \Theta \rangle \propto -\vec{\nabla} T$. Therefore the mean flux of the inertial particles is directed to the minimum of the mean temperature and the inertial particles are accumulated in this region. This effect is more pronounced when turbulent fluid flow is inhomogeneous in the direction of the mean temperature gradient.

Let us study this effect in more detail. Evolution of the mean field N is determined by Eq. (6). Substitution

$$N(t, \mathbf{r}) = N_* \Psi_0(Z) \exp(\gamma_0 t) \times \exp \left[-\frac{1}{2} \int \chi_0(Z) dZ + i\mathbf{k} \cdot \mathbf{r}_\perp \right] + N_0(\mathbf{r})$$

reduces Eq. (6) to the eigenvalue problem for the Schrödinger equation

$$\frac{1}{m_0} \Psi_0''(Z) + [W_0 - U_0(Z)] \Psi_0(Z) = 0, \quad (9)$$

where $W_0 = -\gamma_0$, $A' = dA/dZ$, and the potential U_0 is given by

$$U_0 = \frac{1}{m_0} \left(\frac{\chi_0^2}{4} + \frac{\chi_0'}{2} + \kappa_0 \right),$$

and $\chi_0 = f'(Z) + \eta_0 \exp(-f) T'(Z)$, $\kappa_0 = k^2 - \eta_0 \exp(-f) T''$, and $m_0 = \exp[-f(Z)]$. Here the axis Z is directed along mean temperature gradient, the wave vector \mathbf{k} is perpendicular to the axis Z , and

$$\eta_0 = \frac{3\alpha}{\text{Pe}} \left(\frac{m_p}{m_\mu} \right) \left(\frac{\delta T}{T_*} \right) \ln \text{Re}_*.$$

In the derivation of Eq. (9) we take into account that for an isotropic turbulence $\langle v_m(\mathbf{x}) v_n(\mathbf{x}) \rangle = u_0^2 \exp(f) \delta_{mn} / 3$. The equilibrium distribution of the mean number density $N_0(\mathbf{r})$ is determined by equation $\hat{D} \vec{\nabla}_m N_0 = \mathbf{V}_{\text{eff}} N_0$. Equation (9) is written in dimensionless form, the coordinate is measured in units Λ_T , time t is measured in units Λ_T^2 / D_T , wave number k is measured in units Λ_T^{-1} , the temperature T is measured in units of temperature difference δT in the scale Λ_T , and concentration N is measured in units N_* .

Now we use the quantum mechanical analogy for the analysis of the large-scale pattern formation in the concentration field N of the inertial particles. The instability can be excited ($\gamma_0 > 0$) if there is a region of potential well where $U_0 < 0$. A positive value of W_0 corresponds to turbulent diffusion, whereas a negative value of W_0 results in the excitation of the instability. The condition when the potential U_0 can be negative is given by $\chi_0'/2 + \kappa_0 < 0$. For instance, this condition can be satisfied in the vicinity of both the temperature minimum ($T'' > 0$) and the maximum in $\langle \mathbf{u}^2 \rangle$ ($f'' < 0$). Expansion of functions $T(Z)$ and $f(Z)$ in the Taylor series $T(Z) \sim T_m + Z^2 + \dots$ and

$f(Z) \sim -a_0 Z^2 + \dots$ in the vicinity of $Z = 0$ reduces the problem to that of a harmonic oscillator in quantum mechanics. For simplicity we assume that the minimum temperature at the point $Z = 0$ coincides with the maximum $f(Z)$, and the value $\ln\langle \mathbf{u}^2 \rangle$ is measured in the units of value f at the point $Z = 0$. Discrete levels of energy of the oscillator are determined by condition $E - 1 = 2p$ ($p = 0, 1, 2, \dots$). This yields the growth rate of the instability for $p = 0$:

$$\gamma_0 = \eta_0 + \frac{3}{2} a_0 - \left[\eta_0^2 + \frac{9}{4} a_0^2 - a_0 k^2 \right]^{1/2} - k^2.$$

The solution for Ψ_0 can be expressed in terms of the Hermite polynomials. Thus the initially spatial distribution of the concentration of the inertial particles evolves into a pattern containing regions with increased (decreased) concentration of particles. The characteristic vertical size of the inhomogeneity when $\eta_0 \geq a_0$ is of the order of

$$l_z \sim \Lambda_T \left[\frac{3\alpha}{\text{Pe}} \left(\frac{m_p}{m_\mu} \right) \left(\frac{\delta T}{T_*} \right) \ln \text{Re}_* \right]^{-1/2}.$$

Remarkably $l_z \rightarrow \infty$ when $\text{Pe} \rightarrow \infty$; i.e., this effect exists for large but finite Peclet numbers.

The obtained results are valid in the case when the density of the surrounding fluid is much less than the material density of particles ($\rho \ll \rho_p$). However, the results of this study can be easily generalized to include the case $\rho \geq \rho_p$ using the equation of motion of particles in fluid flow presented in [4,12]. This equation of motion takes into account contributions due to the pressure gradient in the fluid surrounding the particle (caused by acceleration of the fluid) and the virtual ("added") mass of the particles relative to the ambient fluid. The solution of this equation coincides with Eq. (2) except for the transformation $\tau_p \rightarrow \beta \tau_p$, where

$$\beta = \left(1 + \frac{\rho}{\rho_p} \right) \left(1 - \frac{3\rho}{2\rho_p + \rho} \right).$$

For $\rho \geq \rho_p$ the turbulent thermal diffusion ratio k_T in Eq. (8) must be multiplied by β . Therefore the additional mass flux of particles is directed towards the mean temperature gradient [see Eq. (7)], and particles are accumulated in the vicinity of the maximum of mean temperature of surrounding fluid since $\beta < 0$. In the opposite case when $\rho \ll \rho_p$, $\beta \approx 1$ and particles are accumulated in the vicinity of the mean temperature minimum.

The analyzed effect of turbulent thermal diffusion may be of relevance in some atmospheric phenomena (e.g., atmospheric aerosols, cloud formation, and smog formation) and combustion. Most aerosol particles are trapped in the vicinity of mean temperature minimum. Observations of the vertical distributions of aerosols in the atmosphere show that maximum concentrations can occur within temperature inversion layers

(see, e.g., [13]). This is in agreement with the necessary condition ($T'' > 0$, i.e., the temperature inversion) for the instability considered in the present study. Using the parameters of the atmospheric turbulent boundary layer $u_0 \sim 30\text{--}100$ cm/s, $l_0 \sim 10^3\text{--}10^4$ cm and of the temperature inversion $\Lambda_T \sim 3 \times 10^4$ cm and $\delta T/T_* \sim (1 - 3) \times 10^{-2}$ (see, e.g., [13]), we obtain that the characteristic time of excitation of the instability of concentration distribution of aerosols with $\rho_p \sim 2$ g/cm³ and $a_* = 10$ μm varies in the range from 0.3 to 3 h. Note that this time $\sim a_*^{-2}$. This value is in compliance with the characteristic time of growth of inhomogeneous structures in atmosphere.

In summary, we have analyzed a new phenomenon of turbulent thermal diffusion. This phenomenon is caused by the correlation between temperature and velocity fluctuations of the surrounding fluid and leads to relatively strong mean flux of small inertial particles in regions with mean temperature gradients. It is conceivable that similar phenomena can occur in deterministic flows with chaotic behavior.

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