Nonequilibrium structures in reacting fluids subject to external forces

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It is shown that a reacting fluid under the action of external forces can develop nonequilibrium stationary structures. The main difference between these nonequilibrium states and the hydrostatic one is the appearance of a velocity field that drives the fluid from the zones where particles are created by reactions to the regions where they annihilate. Although this mechanism is derived here for a perfect fluid subject to gravity, within a particular reaction model and a partial-linearization scheme, the results are reasonably expected to hold in more general situations.

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The interplay of reaction and transport processes in physical systems has been the subject of a considerable amount of work in the last years (see Ref. [1] and references therein). This has been motivated by the interest of such phenomena in many applications [2,3], but it is also due to the paradigmatic role that those systems play as models of complex behavior [4]. In this sense, spatially extended reacting systems are also of interest in other areas of science, such as biology [5] or economics [6].

Although most of the research in this field has been focused on the study of reacting and diffusing systems, alternative transport mechanisms—other than diffusion—are worth considering. In fact, turbulent or convective transport could be dominant in many situations [3,5]. In Ref. [7] a model has been introduced to study the mutual effect of reaction and transport in a perfect fluid. Following the lines used to derive reaction-diffusion equations, the proposed model equations for a reacting perfect gas at constant temperature T are a straightforward extension of Euler equations [8],

$$\begin{aligned} &\partial_t n + \nabla \cdot (n\mathbf{u}) = F(n), \\ &\partial_t \mathbf{u} + (\mathbf{u}\cdot\nabla)\mathbf{u} = -\nabla p/\rho - \gamma \mathbf{u} + \mathbf{f}/m, \end{aligned} \tag{1}$$

where $n(\mathbf{r},t)$, $\mathbf{u}(\mathbf{r},t)$, and $p(\mathbf{r},t)$ are the number density, velocity, and pressure fields, respectively. Here $\rho = mn$ is the mass density, with m the mass of a fluid particle, and $\mathbf{f}(\mathbf{r})$ is the external force per particle. It is supposed that this external force can be derived from a potential, $\mathbf{f} = -\nabla V$. The nonlinear source term $F(n)$ in the density equation accounts for reaction processes, which are considered to have no effect on the velocity or the temperature. Generally, these reactions take place between the particles of the fluid and with a particle background, which is not supposed to be affected by the dynamics of the fluid [9]. Finally, the term $-\gamma\mathbf{u}$ in the velocity equation stands for the dissipative effects of the background.

In the absence of reactions, $F(n) \equiv 0$, the stationary solution to Eqs. (1) is the usual hydrostatic state, where the pressure gradient balances the external forces and $\mathbf{u} = \mathbf{0}$. Supposing that the pressure of the perfect fluid is given by the ideal gas equation of state, $p = nkT$ (k being the Boltzmann constant) the hydrostatic density n_H is given by the equilibrium thermodynamics formula

$$n_H(\mathbf{r}) = n_0 \exp\left[-\frac{V(\mathbf{r})}{kT}\right], \tag{2}$$

where n_0 is an arbitrary constant related to the total particle number.

How do reaction processes modify this hydrostatic state? In order to answer this question, which is the main goal of this paper, one must look for the time-independent solutions to the full reaction-convection equations (1). In order to simplify the mathematical problem, one-dimensional problems will be considered as a first step. They correspond to the case when the external force is directed along a single spatial coordinate and depends on that coordinate only—such as the gravitational or the centrifugal forces. Under these conditions, the stationary state satisfies the equations

$$\frac{d}{dx}(nu) = F(n), \tag{3}$$

and

$$\begin{aligned} u\frac{du}{dx} &= -\frac{kT}{mn}\frac{dn}{dx} - \gamma u + \frac{f(x)}{m} \\ &= -\frac{kT}{m}\frac{d}{dx}\left[\ln n + \frac{V(x)}{kT}\right] - \gamma u, \end{aligned} \tag{4}$$

where $u \equiv \mathbf{u}\cdot\hat{\mathbf{x}}$ and $f \equiv \mathbf{f}\cdot\hat{\mathbf{x}}$. From these equations, it is apparent that, under the effect of reactions, the stationary state cannot correspond to the fluid at rest. In fact, if $u = 0$, Eq. (3) would imply that the density should equal one of the roots of $F(n)$ and, in particular, n should not depend on x. This is in contradiction with Eq. (4) which, for vanishing velocity, has Eq. (2) as its only solution. Hence, if a solution to Eqs. (3) and (4) does exist, it would imply that the interplay of reactions and external forces determine a dynamical stationary state with the fluid in motion.

Due to their nonlinear character, solving Eqs. (3) and (4) is not trivial at all. In general, it would be necessary to resort to the use of numerical techniques. However, much insight on the physical processes that determine the stationary states can be obtained in an analytical way

by partially linearizing the equations as follows. Consider that both n and u differ from their respective hydrostatic values, $n_H(x)$ and $u_H = 0$, in vanishingly small quantities. Then linearize the equations in the usual way, keeping, however, the full nonlinear character of the reaction function $F(n)$. This partial linearization—which simplifies the hydrodynamical problem but preserves all the complexity involved in the reaction processes—produces, from Eq. (4), an explicit expression for the velocity,

$$u = -\frac{kT}{\gamma m}\frac{d}{dx}\left(\frac{n}{n_H}\right). \tag{5}$$

Replacing this in the partially linearized version of Eq. (3) and taking into account Eq. (2), one obtains for the density

$$-\frac{kT}{\gamma m}\frac{d^2n}{dx^2} + \frac{d}{dx}\left[\frac{f(x)}{\gamma m}n\right] = F(n). \tag{6}$$

This is a stationary inhomogeneous Fokker-Planck equation [10]. It is interesting to note that if the left-hand side of the first of Eqs. (1) had contained an additional term describing diffusion, $-D_0\nabla^2 n$, it would be simply added to the left-hand side of Eq. (6), producing an effective diffusivity $D = D_0 + kT/\gamma m$. Therefore, the analysis can be trivially extended to consider an additional diffusion effect. In view of this fact, the coefficient multiplying the second derivative of n in Eq. (6) will be henceforth denoted by D.

Remarkably enough—although a linearization has been performed—the solution to Eq. (6) in the absence of reactions is again Eq. (2). For nonvanishing $F(n)$, instead, solving the stationary Fokker-Planck equation is practically impossible. Even for a linear reaction function and simple forms of $f(x)$, the solution contains typically very involved special functions [11]. The analysis will be, therefore, restricted to the case of a constant external force that, for the sake of concreteness, is taken as the gravity force near the Earth surface, $f = -mg$. Physically meaningful solutions to the hydrodynamical problem under the action of a constant force are obtained by imposing a boundary, the Earth surface, able to compete with the drifting effect of the force. At that boundary, situated at $x = 0$, a condition of total reflexion or zero net flux, $u = 0$, will be imposed. As for the reaction function, the Ballast model [12],

$$F(n) = -\frac{1}{\tau}\left[n - n_3\theta(n - n_2)\right], \tag{7}$$

is considered. Here, θ is the Heaviside step function. The Ballast model is a bistable reaction model with stationary stable states at $n_1 = 0$ and n_3, separated by the unstable state n_2 ($0 < n_2 < n_3$). It mimics more complex bistable models such as Schlögl's [1]. Its piecewise-linear character preserves the main ingredients of nonlinearity and makes possible an analytical treatment. In Eq. (7), τ is a typical relaxation time associated to reaction events, which measures the efficiency of reactions.

Under the above mentioned conditions, the solution to Eq. (6) reads

$$n(x) = \begin{cases} n_3 + A\exp(x/\lambda_+) + B\exp(x/\lambda_-) & \text{for } x < x_2 \\ C\ \exp(x/\lambda_-) & \text{for } x > x_2, \end{cases} \tag{8}$$

where

$$\frac{1}{\lambda_\pm} = -\frac{1}{\lambda_0} \pm \sqrt{\frac{1}{\lambda_0^2} + \frac{1}{\tau D}}, \tag{9}$$

($\lambda_- < 0 < \lambda_+$) and $\lambda_0 = 2\gamma D/g$. In Eq. (8), x_2 is the point at which n equals the unstable state n_2. The coefficients A, B, C, and x_2 itself must be obtained from continuity conditions of n and its first derivative at x_2, along with the boundary conditions $n(x_2) = n_2$ and $u(0) = 0$.

Figure 1 shows the normalized density $n(x)/n_3$ as a function of the scaled coordinate x/λ_0, for $\tau D = 0.5\lambda_0^2$, and $n_2 = 0.22n_3$. The dotted curve stands for the hydrostatic density (2). One can observe that $n(x)$ differs from $n_H(x)$ as expected according to the form of the reaction function $F(n)$. In the region where $n_2 < n < n_3$ ($x_3 < x < x_2$), $F(n)$ is positive and particles are created due to reactions. Approximately in this zone, the stationary density is overpopulated with respect to the hydrostatic state. The opposite effect is seen in the zone where $n > n_3$, which implies $F(n) < 0$. There the density is relatively depleted. Note that this is not observed in the region where $n < n_2$ ($x > x_2$), certainly due, as explained later, by the positive contribution of particles transported from the left.

The velocity field $u(x)$ is also shown in Fig. 1, measured in arbitrary units. Note the discontinuity in its derivative at x_2, a consequence of the singularity of $F(n)$, Eq. (7), at n_2. In the region of particle creation, $x_3 < x < x_2$, the velocity changes its sign passing from negative values to the left to positive values to the right. Particles situated just above x_3 are, therefore, transported towards the lower region of particle annihilation, $x < x_3$. In an analogous way, particles in the

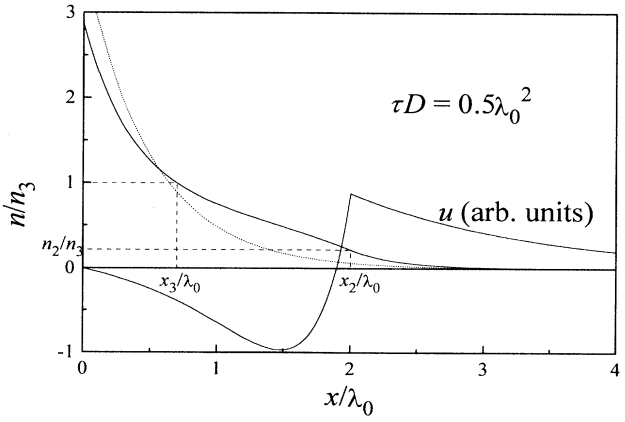

FIG. 1. The normalized nonequilibrium density n/n_3 as a function of the scaled coordinate x/λ_0, for $\tau D = 0.5\lambda_0^2$ and $n_2/n_3 = 0.22$. The dotted curve corresponds to the hydrostatic density. The velocity $u(x)$, measured in arbitrary units, is also shown.

creation zone just below x_2 move toward the upper annihilation zone, $x > x_2$. Hence, there is a continuous flow of particles from the zone where they are created to the regions in which they are annihilated.

This provides an explanation for the mechanism which sustains the stationary nonequilibrium density profile in the presence of reaction processes: the hydrostatic state breaks up, and a net circulation of fluid arises, driving the particles created by reactions to the zones where they disappear. Of course, this line of reasoning can be inverted to consider the effect of external forces on reacting fluids. In the absence of forces, the stationary state of the fluid is given by $u = 0$ and $F(n) = 0$, i.e., $n = 0$ or $n = n_3$, a spatially homogeneous density. The presence of a force, then, generates a velocity field that determines the density to exhibit a nontrivial profile.

Although these results have been derived under very particular conditions, one can reasonably conjecture that the interplay mechanism of reactions and forces explained above holds in more general situations. Another important point that deserves further consideration is the stability of the nonequilibrium states generated by that interplay. These aspects are the subject of work in progress.

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