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## THERMAL FLUCTUATIONS IN A KNUDSEN FLOW SYSTEM

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Spatial correlations of thermal fluctuations in a model system are examined. The system is a one-dimensional chain of cells containing a dilute gas and connected by Knudsen apertures. A Monte Carlo simulation is described in which the long range correlations observed agree quantitatively with a general master equation formulation.

Interesting modifications to the dynamic correlations of thermodynamic fluctuations in fluids subjected to a thermal gradient has been predicted by both fluctuating hydrodynamics [1] and kinetic theory [2]. This phenomenon is observed experimentally in the linear dependence on temperature gradient of the asymmetry in Brillouin light scattering [3,4]. The master equation formalism which has been so successful with composition fluctuations in chemical systems [5] has recently been applied to the study of thermal problems [6,7]. Nicolis and Malek Mansour [8] have examined a one-dimensional system of  $M$  identical, thermally coupled cells (of volume  $\Delta V$  and heat capacity  $C_v$ ) connected to two reservoirs held at different temperatures. The instantaneous state of the system is described by the vector  $E$  where  $E_k$  is the energy of cell  $k$ , the energy of the boundary cells,  $E_0$  and  $E_{M+1}$ , being held fixed. The transition rate  $W$  for a cell to transfer an amount of energy  $\epsilon$  to a neighbor cell is assumed to be independent of the state of the neighboring cells; i.e.

$$W(E_k + \epsilon, E_{k+1} - \epsilon \rightarrow E_k, E_{k+1}) = W(E_k, \epsilon). \quad (1)$$

One obtains an expression for this quantity from the condition of detailed balance at equilibrium,

$$W(E_k, \epsilon) = \varphi(\epsilon) \exp(-\epsilon/k_B T_k), \quad (2)$$

where  $T_k$  is the instantaneous temperature,  $k_B$  is the Boltzmann constant and  $\varphi(\epsilon)$  is an unspecified function. By definition, the quantity  $W$  is related to the drift  $f$  and the diffusion  $Q$  of the Fokker-Planck

equation as follows [9]:

$$f(E_k) = \frac{1}{\Delta V} \int d\epsilon \epsilon W(E_k, \epsilon), \quad (3a)$$

$$Q(E_k) = \frac{1}{\Delta V} \int d\epsilon \epsilon^2 W(E_k, \epsilon). \quad (3b)$$

From eq. (2), a further relation is

$$Q(E_k) = -\frac{\partial}{\partial(1/k_B T_k)} f(E_k). \quad (4)$$

One imposes that the average temperature  $\langle T_k \rangle$  obey the heat equation (with constant thermal diffusivity  $\kappa$ ), written in discrete form as

$$\frac{\partial}{\partial t} \langle T_k \rangle = \kappa (\langle T_{k+1} \rangle + \langle T_{k-1} \rangle - 2\langle T_k \rangle). \quad (5)$$

Identifying this macroscopic law with the drift component of the Fokker-Planck equation, the evolution is entirely specified by eqs. (4) and (5). From (5), the temperature gradient is linear and its amplitude is  $\gamma \equiv (T_{M+1} - T_0)/(M+1)$ . After some algebra, one arrives at the static correlation function for the temperature fluctuations,

$$\begin{aligned} \langle \delta T_k \delta T_m \rangle &= \frac{k_B \delta_{km} \langle T_k \rangle^2}{C_v \Delta V} + \frac{k_B \gamma^2}{C_v (M+1)} \\ &\times k(M+1-m), \quad k < m, \\ &\times m(M+1-k), \quad m < k. \end{aligned} \quad (6)$$

The first term is the local equilibrium part of the static correlation function; the second is the nonequilibrium contribution. Note the presence of long range linear correlations with an amplitude proportional to the square of the temperature gradient.

As discussed by Nicolis and Malek Mansour, from eqs. (2)–(5), one could solve explicitly for  $W(E_k, \epsilon)$ . However, the condition that  $\kappa$  be a constant independent of temperature leads to the unphysical result  $\varphi(\epsilon) \sim 1/\epsilon$ . This prevents one from constructing a numerical Monte Carlo simulation for this model since the transition rate implies an infinitely frequent exchange of an infinitesimal amount of energy.

A markovian system for which the transport mechanism is exactly derivable from microscopic arguments was numerically simulated in order to see if the results predicted by the above theory could be reproduced. Consider a chain of cells filled with a

dilute gas and connected by very small apertures. If the size of the apertures is on the order of a mean free path, the flow between the cells will be Knudsen. We make the strong assumption that the form of the correlation function is invariant for different systems with only a model dependent scale factor. The single cell ( $M=1$ ) result for the Knudsen system [6] fixes this scale factor. If  $N_k$  is the number of particles in cell  $k$  then the static correlation function for a Knudsen system is

$$\langle \delta T_k \delta T_m \rangle = \frac{\delta_{km} \langle T_k \rangle^2}{\frac{1}{2} \langle N_k \rangle} + \frac{\frac{1}{3} \gamma^2}{\frac{1}{2} \langle N_k \rangle (M+1)}$$

$$\times k(M+1-m), \quad k < m,$$

$$\times m(M+1-k), \quad m < k. \tag{7}$$

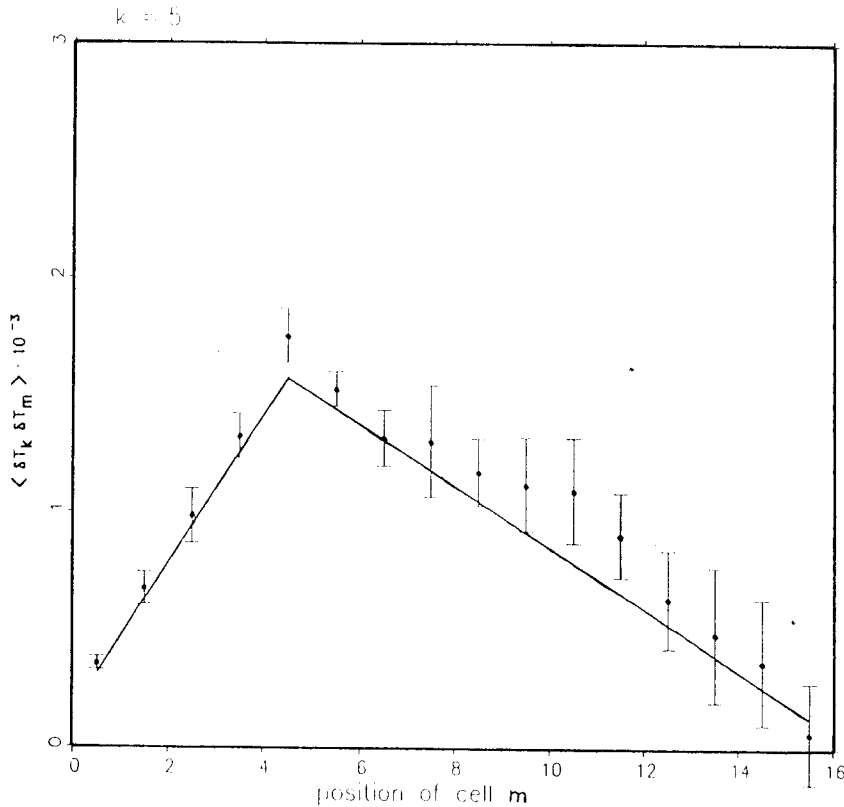


Fig. 1. Plot of the nonequilibrium contribution to the temperature fluctuation correlation of cell  $k=5$  with cell  $m$  versus the position of cell  $m$ . The solid line is obtained from the second term in eq. (7). The points are data from the Monte Carlo simulation; the error bars are 90% confidence intervals.

Note that this implies relations induced by a [10].

For Knudsen flow, energy  $\epsilon$  leave cell  $k$  is

$$W(E_k, \epsilon) = DN_k T_k^{1/2} \times \exp(-\epsilon/k_B T_k) (1$$

where  $D$  is a geometric that  $W(E_k, \epsilon)$  is only cell  $k$ , which is consist

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$$\Omega_k = DN_k T_k^{1/2} (1 - \frac{1}{2} \delta_k$$

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Note that this implies that there are no spatial correlations induced by a particle gradient (see also ref. [10]).

For Knudsen flow, the rate at which particles of energy  $\epsilon$  leave cell  $k$  is

$$W(E_k, \epsilon) = DN_k T_k^{1/2} [\epsilon / (k_B T_k)^2] \times \exp(-\epsilon / k_B T_k) (1 - \frac{1}{2} \delta_{k,0} - \frac{1}{2} \delta_{k,M+1}), \quad (8)$$

where  $D$  is a geometrical factor for the aperture. Note that  $W(E_k, \epsilon)$  is only a function of the state of the cell  $k$ , which is consistent with the above discussion. If the states of the boundary cells are chosen such that  $N_0 \sqrt{T_0} = N_{M+1} \sqrt{T_{M+1}}$  these boundary conditions imply that the temperature gradient is linear and the mass flux through the system is zero. Integrating eq. (8), the total rate at which particles leave cell  $k$  is

$$\Omega_k = DN_k T_k^{1/2} (1 - \frac{1}{2} \delta_{k,0} - \frac{1}{2} \delta_{k,M+1}). \quad (9)$$

The formulation of the Monte Carlo simulation is as follows [11]: from  $\{\Omega_k\}$ , choose the cell  $i$  in which the next event will take place as

$$\sum_{k=-1}^{i-1} \Omega_k < R \Omega_{\Sigma} < \sum_{k=-1}^i \Omega_k, \quad (10)$$

where  $R$  is a uniformly distributed random number in  $(0, 1]$ ,  $\Omega_{-1} \equiv 0$  and

$$\Omega_{\Sigma} \equiv \sum_{k=0}^{M+1} \Omega_k. \quad (11)$$

Since the process is markovian, the time between events is an exponentially distributed random number; the time until the next event  $\tau$  is chosen as

$$\tau = -\ln(R') / \Omega_{\Sigma}. \quad (12)$$

The probability that a particle leaving a cell has energy  $\epsilon$  is

(7)

(7)

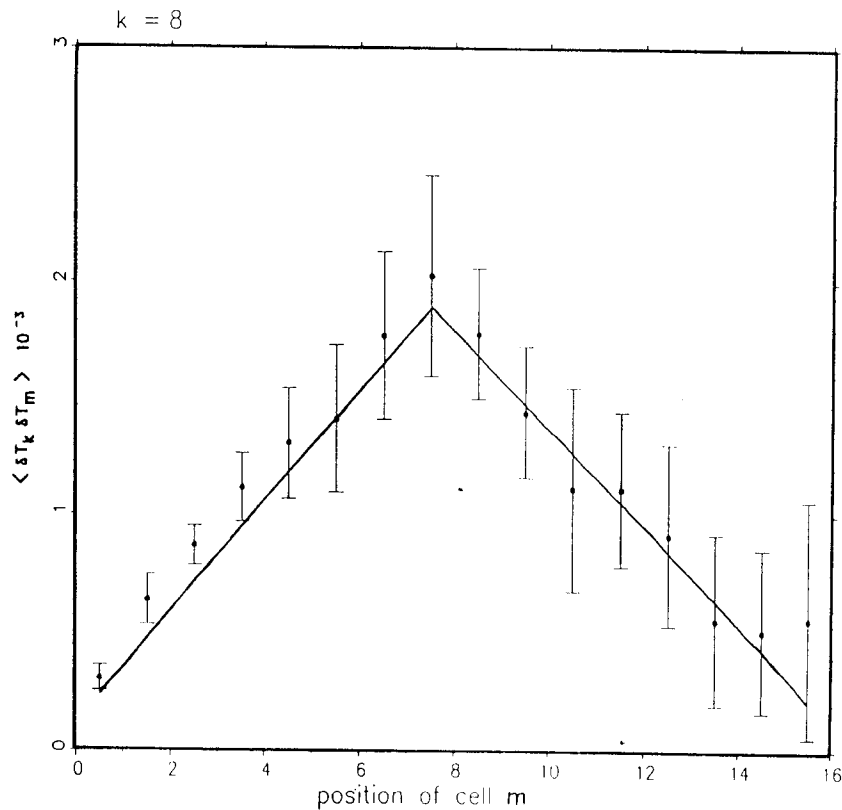


Fig. 2. Same as fig. 1 for cell  $k=8$ .

us the position of on; the error bars

$$\pi(E_k, \epsilon) = [\epsilon / (k_B T_k)^2] \exp(-\epsilon / k_B T_k), \quad (13)$$

the energy of the particle which leaves cell  $i$  is chosen as [12]

$$\epsilon = k_B T_i \ln(RR'). \quad (14)$$

The neighboring cell to which the particle moves is taken at random and the two cells are updated. The state of the boundary cells is always held fixed.

A one-dimensional system of sixteen cells between two reservoirs was simulated numerically. An equilibrium run ( $N_0 = N_{17}$ ,  $T_0 = T_{17}$ ) was made to verify that the program yielded the correct equilibrium correlations. For the nonequilibrium run, the reservoirs were set to  $N_0 = 900$ ,  $T_0 = 400$ ,  $N_{17} = 246.2$ ,  $T_{17} = 5345$ , giving a linear temperature gradient of about 300 degrees per cell. The simulation was run for over 200 million total events. The measured temperature correlations are shown in figs. 1 and 2 along with the theoretical prediction from eq. (7). Note that there are no free parameters; the good agreement is truly quantitative. Similar results were also obtained for a smaller (10 cell) system. Molecular dynamics simulations [13] and an approximate simulation of the Kac model for a dilute gas [14] have yielded qualitatively similar results [15].

From the single cell calculation for the Knudsen system, we know that the nonequilibrium contributions to the number and number-temperature correlations ( $\langle \delta N_k \delta N_m \rangle$  and  $\langle \delta N_k \delta T_m \rangle$ ) are about an order of magnitude smaller than the contribution to  $\langle \delta T_k \delta T_m \rangle$ . These other correlation functions are of interest as they are directly related to the amplitude of the Rayleigh line but unfortunately their nonequilibrium components are difficult to observe. We do expect all the correlations to be similar in form, that is, long ranged with an amplitude proportional to the square of the temperature gradient. This is in agreement with theoretical predictions of the modifications to the Rayleigh line in light scattering [4,16].

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EXACT EVALUATION OF CORRELATIONS ASSOCIATED WITH A ONE-DIMENSIONAL SYSTEM OF PARTICLES

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We give an exact evaluation of the correlation function  $\int_0^t ds f(t,s)x(s)f(t,s)$

In connection with the exact evaluation of the correlation functions for the description of the surface growth, the following result arises:

$$I(t) = \int Dx(t) \delta \left[ \dot{x}(t) \right]$$

(see also ref. [3]), where the integration is carried out over all  $x(t)$ , and  $\delta$  is the delta functional. The result can be written out explicitly,

$$I(T) = \lim_{N \rightarrow \infty, \epsilon \rightarrow 0} \frac{1}{N\epsilon} \int \dots \int \times \delta \left( \frac{x_{j+1} + x_{j-1} - 2x_j}{\epsilon^2} \right)$$

with  $x_0 = 0$ ,  $x_{N+1} = 0$  and  $\epsilon$  is to be taken as the time step in eq. (1). The integration is carried out to obtain  $I(T)$  to be equal to the inverse of a Fredholm determinant

$$I(T) = \lim_{N \rightarrow \infty} \epsilon^{-1} \|\mathbf{A}_N\|^{-1}$$

where  $(\mathbf{A})_{jk} = -2\delta_{jk}$  and that  $I(T)$  is nothing but

<sup>1</sup> Permanent address.