Journal of Statistical Physics, Vol. 90, Nos. 5/6, 1998

Long-Ranged Correlations in Bounded Nonequilibrium Fluids

Alejandro L. Garcia,[1,3] G. Sonnino,[2] and M. Malek Mansour[2]

Received June 30,† 1997; final August 21, 1997

In a recent paper, Liu and Oppenheim [*J. Stat. Phys.* **86**:179 (1997)] solve the fluctuating heat diffusion equation for a bounded system with a temperature gradient. This note demonstrates that, contrary to their claims, their solution for the temperature correlation function is indeed long-ranged and reduces to that of Garcia *et al.* [*J. Stat. Phys.* **47**:209 (1987)].

KEY WORDS: Fluctuating hydrodynamics; long-ranged correlations; direct simulation Monte Carlo; molecular dynamics.

The theoretical analysis of long-ranged spatial correlations of fluctuations in a simple fluid was recently reexamined by Liu and Oppenheim.[1] Specifically, they consider the fluctuating heat diffusion (Fourier) equation for a system confined between heat reservoirs at $z=0$ and L_z held at fixed temperatures T_0 and T_L, respectively, and subjected to periodic boundary conditions in the other directions. If one neglects the state dependence of the thermal conductivity coefficient λ, then the stationary temperature profile is a linear function of the z coordinates:

$$T_s(\mathbf{r}) = T_0 + z\gamma \qquad (1)$$

where $\gamma \equiv (T_L - T_0)/L_z$ stands for the imposed temperature gradient.

---

[1] Center for Computational Sciences and Engineering, Lawrence Berkeley National Laboratory, Berkeley, California 94720.

[2] Centre for Nonlinear Phenomena and Complex Systems, Université Libre de Bruxelles, Campus Plaine, C.P. 231, B-1050 Brussels, Belgium.

[3] Permanent address: Physics Department, San Jose State University, San Jose, California 95192.

0022-4715/98/0300-1489$15.00/0

Consider now the temperature fluctuations around the stationary state, defined as $\delta T(\mathbf{r}, t) \equiv T(\mathbf{r}, t) - T_s(\mathbf{r})$. Using the Landau Lifshitz fluctuating hydrodynamics formalism, one finds[2]

$$\frac{\partial}{\partial t} \delta T(\mathbf{r}, t) = \frac{\lambda}{\rho C_p} \nabla^2 \delta T(\mathbf{r}, t) - \frac{1}{\rho C_p} \nabla \cdot \mathbf{J}(\mathbf{r}, t) \tag{2}$$

where ρ and C_p represent the mass density and the heat capacity per unit mass at constant pressure, respectively. The fluctuating heat flux $\mathbf{J}(\mathbf{r}, t)$ has zero mean and variance given by

$$\langle J_i(\mathbf{r}, t)\, J_j(\mathbf{r}', t') \rangle = 2k_B \lambda T_s^2(\mathbf{r})\, \delta(\mathbf{r} - \mathbf{r}')\, \delta(t - t')\, \delta_{i,j}^{\mathrm{Kr}} \tag{3}$$

where k_B is Boltzmann's constant. Being a hydrodynamic formulation, (2) is accurate down to the transport scale (e.g., mean free path) but not to the molecular scale (i.e., atomic diameter).[3] In what follows, we shall be mainly interested in the spatial correlation of temperature fluctuations $\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle$ which can be decomposed as

$$\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle = \frac{k_B T_s^2(\mathbf{r})}{\rho C_p} \delta(\mathbf{r} - \mathbf{r}') + \overline{\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle} \tag{4}$$

where the first term on the right hand side represents the local-equilibrium contribution.

We first consider the "reduced" temperature correlation, defined as the spatial average over the L_x and L_y directions,

$$\overline{\langle \delta T(z)\, \delta T(z') \rangle} \equiv \frac{1}{L_x^2 L_y^2} \int_0^{L_x} dx \int_0^{L_y} dy \int_0^{L_x} dx' \int_0^{L_y} dy'\, \overline{\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle} \tag{5}$$

Combining the equations for $\delta T(\mathbf{r}, t)$ and $\delta T(\mathbf{r}', t)$ and using the definition (5), one gets a *closed* equation for $\overline{\langle \delta T(z)\, \delta T(z') \rangle}$:

$$\left(\frac{\partial^2}{\partial z^2} + \frac{\partial^2}{\partial z'^2} \right) \overline{\langle \delta T(z)\, \delta T(z') \rangle} + 2 \frac{\gamma^2 k_B}{\rho C_p} \frac{\delta(z - z')}{L_x L_y} = 0 \tag{6}$$

The solution of this equation is readily found to be[4, 5]

$$\overline{\langle \delta T(z)\, \delta T(z') \rangle} = \frac{\gamma^2 k_B}{\rho C_p L_x L_y L_z} \begin{cases} z(L_z - z'), & z < z' \\ z'(L_z - z), & z > z' \end{cases} \tag{7}$$

which is clearly long-ranged and does not involve any intrinsic length scale, i.e., the correlation encompasses the entire system. The validity of this solution has been questioned by Liu and Oppenheim on precisely these grounds.

Assuming the system is infinite in the x and y directions and using the method of images, Liu and Oppenheim obtain the three dimensional spatial correlation of temperature fluctuations to be

$$\overline{\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle} = \frac{\gamma^2 k_B}{4\pi\rho C_p} \left(\sum_{n=-\infty}^{\infty} \frac{1}{|\mathbf{r}-\mathbf{r}'_n|} - \sum_{n=-\infty}^{\infty} \frac{1}{|\mathbf{r}-\mathbf{r}''_n|} \right) \tag{8}$$

with $\mathbf{r}'_n = \mathbf{r}' + 2nL_z\hat{\mathbf{z}}$ and $\mathbf{r}''_n = \mathbf{r}' + 2(nL_z - z')\,\hat{\mathbf{z}}$. They then claim that this result demonstrates the absence of long-ranged correlations of temperature fluctuations since $\overline{\langle \delta T(\mathbf{r})\, \delta T(\mathbf{r}') \rangle} \propto |\mathbf{r}-\mathbf{r}'|^{-1}$ when $L_z \gg |\mathbf{r}-\mathbf{r}'|$.

On the other hand, one may compute the reduced temperature correlation function by inserting (8) in (5) and evaluating the integrals and sums numerically. Taking 100 points per spatial direction and 20 images on each side of the boundaries we obtain the result shown in Fig. 1 (filled circles). Comparing with the result given by (7) (solid line) shows that the two are equivalent, to within the accuracy of the numerical calculation. This equivalence was expected since both (7) and (8) come from the Greens' function solution of the Poisson equation. Clearly, neither solution has an intrinsic length scale.

Finally, we stress that reduced variables are not introduced merely to simplify the theoretical analysis. Besides light scattering experiments,[6] nonequilibrium fluctuations are studied by time-integrating the stochastic hydrodynamic equations and by simulations of a fluid at the microscopic level (e.g., molecular dynamics, direct simulation Monte Carlo). For these numerical experiments, spatial averaging is routinely applied to improve

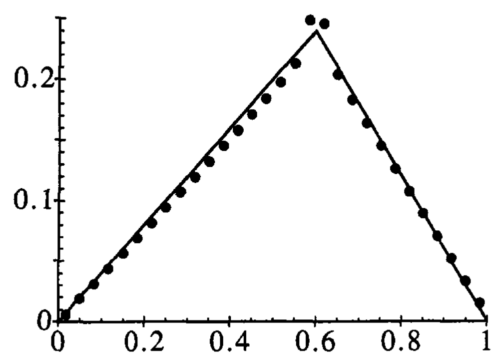

Fig. 1. Correlation of reduced temperature fluctuations $\overline{\langle \delta T(z)\, \delta T(z') \rangle}$ versus z'/L as given by Eq. (8) (filled circles) and by Eq. (7) (solid line) for $z/L = 0.6$. The parameters are set so that $(T_L - T_0)^2 k_B/(\rho C_p L_x L_y L_z) = 1$.

the statistics and long-ranged correlations of the form given by (7) have been observed in numerous nonequilibrium scenarios.[5, 7, 8]

ACKNOWLEDGMENTS

The authors wish to thank Professor G. Nicolis for helpful discussions. The work was supported in part by National Science Foundation, contract 21-1507-7543.

REFERENCES

1. C. Z. W. Liu and I. Oppenheim, *J. Stat. Phys.* **86**:179 (1997).
2. L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon Press, Oxford, 1984).
3. J. P. Boon and S. Yip, *Molecular Hydrodynamics* (Dover, New York, 1991).
4. G. Nicolis and M. Malek Mansour, *Phys. Rev. A* **29**:2845 (1984).
5. A. L. Garcia, M. Malek Mansour, G. C. Lie, and E. Clementi, *J. Stat. Phys.* **47**:209 (1987).
6. Some recent experimental results concerning systems under temperature gradient can be found in: B. M. Law and J. V. Sengers, *J. Stat. Phys.* **57**:531 (1989); B. M. Law, P. N. Segrè, R. W. Gammon, and J. V. Sengers, *Phys. Rev. A* **41**:816 (1990); W. B. Li, J. V. Sengers, R. W. Gammon, and P. N. Segrè, *Int. J. Thermophysics* **16**:23 (1995).
7. M. Malek Mansour, A. L. Garcia, G. Lie, and E. Clementi, *Phys. Rev. Lett.* **58**:874 (1987); A. L. Garcia, M. Malek Mansour, G. Lie, M. Mareschal, and E. Clementi, *Phys. Rev. A* **36**:4348 (1987); M. Mareschal, M. Malek Mansour, G. Sonnino, and E. Kestemont, *Phys. Rev. A* **45**:7180 (1992).
8. A. Suárez, J. P. Boon and P. Grosfils, *Phys. Rev. E* **54**:1208 (1997).