

# Lattice Boltzmann Simulation of Non-Ideal Fluids

Michael R. Swift, W. R. Osborn and J. M. Yeomans

Theoretical Physics, 1 Keble Road, Oxford OX1 3NP, U.K.

## Abstract

A lattice Boltzmann scheme able to model the hydrodynamics of phase separation and two-phase flow is described. Thermodynamic consistency is ensured by introducing a non-ideal pressure tensor directly into the collision operator. We also show how an external chemical potential can be used to supplement standard boundary conditions in order to investigate the effect of wetting on phase separation and fluid flow in confined geometries. The approach has the additional advantage of reducing many of the unphysical discretisation problems common to previous lattice Boltzmann methods.

PACS numbers: 02.70Ns, 05.70Fh, 47.11+j

The hydrodynamics and kinetics of two-component fluids present a wealth of physical problems of both fundamental and technological importance [1]. There is much current interest in the relevance of hydrodynamics to spinodal decomposition [2] and the effects of substrates with different wetting properties on phase separation and domain growth [3]. In addition, the flow properties of multicomponent systems, particularly in porous media, have been intensively studied and are of great relevance to oil recovery [4].

Conventional methods for simulating two-phase flow include numerical integration of the Navier-Stokes equations and molecular dynamics simulations [5]. These techniques are extremely computationally intensive and particularly difficult to implement in random geometries. A newer approach, the lattice Boltzmann method, has recently proved competitive [6]. Here a set of distribution functions defined on a lattice is allowed to relax to equilibrium via a Boltzmann equation, discrete in both space and time. The correct choice of equilibrium distribution ensures that in the long wavelength limit the Navier-Stokes equations are recovered.

Several authors have set up lattice Boltzmann schemes for two-phase systems. In most approaches interface formation has been introduced phenomenologically by modifying the Boltzmann collision operator to impose phase separation [7]. Recent work by Shan and Chen [8] has attempted to relate phase separation to microscopic interactions by redefining the equilibrium velocity distribution so as to simulate a fluid with a non-ideal equation of state. However, their approach leads to inconsistent thermodynamics unless a particular equation of state is chosen. In addition, all current schemes reach equilibrium distributions which have unphysical velocity fluctuations within the interfacial region [9].

In this letter we show for the first time that it is possible to set up a lattice Boltzmann scheme modelling isothermal hydrodynamics for two-phase systems. This is achieved by introducing directly into the collision operator the equilibrium pressure tensor for a non-ideal fluid. The resulting phase transition is pressure driven, as pertinent to a liquid-vapour system quenched to well below the critical point [10]. The fluid reaches the correct thermodynamic equilibrium as determined by the equation of state and a Maxwell construction.

We first summarise the relevant results from the Van-der-Waals formulation of quasi-local thermodynamics for a two-component fluid in thermodynamic equilibrium at a fixed temperature [11]. The free energy functional is taken to be

$$\Psi(\underline{x}) = \int \left\{ \frac{\kappa}{2} |\underline{\nabla} n(\underline{x})|^2 + \psi(n(\underline{x})) \right\} d\underline{x}, \quad (1)$$

where the first term gives the contribution from any density gradients and the second describes the bulk free energy density. The non-local pressure is defined by

$$p(\underline{x}) = n \frac{\delta \Psi}{\delta n} - \Psi(\underline{x}) = p_0 - \kappa n \nabla^2 n - \frac{\kappa}{2} |\underline{\nabla} n|^2, \quad (2)$$

where  $p_0 = n\psi'(n) - \psi(n)$  is the equation of state of the fluid. To obtain the full pressure tensor in a non-uniform fluid, non-diagonal terms must be added [12]:

$$P_{\alpha\beta}(\underline{x}) = p(\underline{x}) \delta_{\alpha\beta} + \kappa \frac{\partial n}{\partial x_\alpha} : \frac{\partial n}{\partial x_\beta}. \quad (3)$$

In equilibrium the components of the pressure tensor define the surface tension in inhomogeneous regions of the fluid.

We next describe how a lattice Boltzmann simulation can be set up to ensure that these equilibrium conditions are met while incorporating the dynamical behaviour pertinent to fluids, namely, order-parameter conservation and hydrodynamic transport. To illustrate our method, we choose to work in two dimensions on a triangular lattice, the simplest geometry that allows us to reproduce the Navier-Stokes equations. Let  $f_i(\underline{x}, t)$  be a non-negative real number describing the distribution function of the fluid density at site  $\underline{x}$  at time  $t$  moving in direction  $\underline{e}_i, i = 1 \dots 6$ . The unit vectors  $\underline{e}_i = \{\cos(2\pi(i-1)/6), \sin(2\pi(i-1)/6)\}$  are the velocity vectors along the links of the lattice. With each site is also associated a function  $f_0(\underline{x}, t)$  which corresponds to the component of the distribution with zero velocity.

The distribution functions evolve according to a Boltzmann equation which is discrete in both space and time

$$f_i(\underline{x} + \underline{e}_i, t + 1) - f_i(\underline{x}, t) = \Omega_i(\underline{x}, t). \quad (4)$$

The most convenient choice for  $\Omega_i$  is a single relaxation time form [13]

$$\Omega_i = -\frac{1}{\tau}(f_i - f_i^{eq}). \quad (5)$$

The density  $n$  and macroscopic velocity  $\underline{u}$  are defined by

$$n = \sum_i f_i, \quad (6)$$

$$nu_\alpha = \sum_i f_i e_{i\alpha}, \quad (7)$$

and the equilibrium distribution,  $f_i^{eq}$ , is chosen so as to reproduce the correct dynamic equations for  $n$  and  $\underline{u}$ .

For a one-component fluid with an ideal gas equation of state,  $f_i^{eq}$  is expanded as a power series in the local velocity and the coefficients determined by local conservation of mass and momentum and by the constraints of Galilean invariance and isotropy of the pressure tensor. In order to include the correct non-local thermodynamic properties of a non-ideal fluid, additional non-local terms are needed in the expansion for  $f_i^{eq}$ . We thus define

$$f_i^{eq} = A + Be_{i\alpha}u_\alpha + Cu^2 + Du_\alpha u_\beta e_{i\alpha}e_{i\beta} + F_\alpha e_{i\alpha} \\ + G_{\alpha\beta}e_{i\alpha}e_{i\beta}, \quad (8)$$

$$f_0^{eq} = A_0 + C_0u^2, \quad (9)$$

where the coefficients, now functions of  $n$  and its derivatives, can be determined by three macroscopic constraints.

The first two are, as for the non-interacting case, local conservation of mass and momentum

$$\sum_i f_i^{eq} = n, \quad (10)$$

$$\sum_i f_i^{eq} e_{i\alpha} = nu_\alpha. \quad (11)$$

The third constraint is that the pressure tensor takes the form

$$\sum_i f_i^{eq} e_{i\alpha}e_{i\beta} = P_{\alpha\beta} + nu_\alpha u_\beta. \quad (12)$$

The constraints (10), (11) and (12), together with the equilibrium thermodynamic definitions (1)–(3), are sufficient to determine the coefficients in the expansions (8) and (9)

$$\begin{aligned}
A_0 &= n - 2(p_0 - \kappa n \nabla^2 n) \\
A &= (p_0 - \kappa n \nabla^2 n)/3 \\
B &= n/3 & C_0 &= -n \\
C &= -n/6 & D &= 2n/3 \\
F_\alpha &= 0 \\
G_{xx} &= -G_{yy} = \frac{\kappa}{3} \left\{ \left( \frac{\partial n}{\partial x} \right)^2 - \left( \frac{\partial n}{\partial y} \right)^2 \right\} \\
G_{xy} &= 2 \frac{\kappa}{3} \frac{\partial n}{\partial x} \frac{\partial n}{\partial y}
\end{aligned} \tag{13}$$

where, on the lattice, derivatives are simply expressed by finite difference approximations.

The continuum hydrodynamic equations modelled by this dynamic scheme can be determined by performing a Chapman-Enskog expansion on the Boltzmann equation (4). To second order, the usual continuity and Navier-Stokes equations result

$$\frac{\partial n}{\partial t} + \frac{\partial(nu_\alpha)}{\partial x_\alpha} = 0, \tag{14}$$

$$\frac{\partial nu_\alpha}{\partial t} + \frac{\partial nu_\beta u_\alpha}{\partial x_\beta} = -\frac{\partial p_0}{\partial x_\alpha} + \nu \nabla^2 nu_\alpha + \frac{\partial}{\partial x_\alpha} (\lambda(n) \underline{\nabla} \cdot n \underline{u}), \tag{15}$$

where  $\nu = (2\tau - 1)/8$  and

$$\lambda(n) = (\tau - \frac{1}{2}) \left( \frac{1}{2} - \frac{\partial p_0}{\partial n} \right). \tag{16}$$

Note that to this order, the only difference between equation (15) and the Navier-Stokes equation for an ideal fluid is the appearance of the non-ideal pressure  $p_0$ .

To test the correctness and applicability of the approach described above we performed simulations on a Van-der-Waals fluid for which

$$\psi = nT \ln \left( \frac{n}{1 - nb} \right) - an^2. \tag{17}$$

We first consider the equilibrium configuration for a system with periodic boundary conditions and zero net flow velocity. The inset of Figure 1 shows the coexistence curve as a function of temperature  $T$ , calculated from equation (2) using a Maxwell construction. The points show simulation data obtained from lattices of size  $256 \times 256$ , equilibrated for 10,000 time steps. Because the correct equilibrium thermodynamics is inherent in the model, the bulk phases reached in the simulations obey the Maxwell construction. Note the wide range of coexisting densities that can be reached by the simulation before significant finite difference errors come into play as the temperature is lowered.

Figure 1 also shows the equilibrium interface density profiles which are seen to depend, as expected, on  $\kappa$  and the temperature. Again the agreement with a direct integration of the continuum thermodynamic equations is excellent. The interfacial width can be varied, typically between  $\sim 2 - 30$  lattice sites. This ensures that lattice anisotropy effects can be made unimportant. Our mechanism for interface formation also reduces the magnitude of the microscopic velocities in the interfacial region by a factor of  $\sim 10^3$  relative to other non-local models. This is because in equilibrium, the effective pressure force goes to zero and thus does not need to be balanced by momentum transfer on the scale of the lattice discretisation.

In Figure 2 we emphasise the consistency between the mechanical definition of surface tension

$$\sigma = R\Delta P, \quad (18)$$

where  $\Delta P$  is the pressure difference between the inside and outside of a spherical domain of radius  $R$ , and the thermodynamic definition

$$\sigma = \kappa \int \left( \frac{\partial n}{\partial z} \right)^2 dz \quad (19)$$

for a flat interface. Agreement is seen as  $R \rightarrow \infty$  with the expected curvature correction to equation (18) appearing for  $R \lesssim 10$  lattice units.

Far from the interface, the fluid obeys the usual Navier-Stokes equation common to other lattice Boltzmann schemes. However, the dynamical behaviour of the interface itself is of

importance if domain growth is to be correctly described by the model. In Figure 3 the dispersion relation for capillary waves is displayed [14] giving a best fit of  $\omega \sim k^{1.6}$ . We attribute the slight discrepancy from the expected dispersion relation  $\omega^2 \sim k^3$  to curvature corrections to Laplace’s Law. These results were obtained by imposing a sine-wave of given wavevector on an interface that had reached equilibrium in a  $128 \times 128$  system and observing the period of the subsequent oscillations for, typically, 500 timesteps.

Finally, we demonstrate how the addition of an external chemical potential at the surfaces of a confined system can be used to supplement the usual bounce-back boundary conditions, allowing us to change the substrate properties and hence study wetting. Gradients in the chemical potential  $\mu_{ex}(\underline{r})$  act as a thermodynamic force on the fluid and can be included within the lattice Boltzmann framework by modifying equation(11)

$$\sum_i f_i^{eq} e_{i\alpha} = nu_\alpha - \tau n \frac{\partial \mu_{ex}}{\partial x_\alpha} \quad (20)$$

which, in turn, introduces a force into the collision operator, equation(8),

$$F_\alpha = -\frac{\tau n}{3} \frac{\partial \mu_{ex}}{\partial x_\alpha}. \quad (21)$$

As an illustrative example, by letting  $\mu_{ex}(\underline{r})$  differ from zero only at the boundary sites, the affinity of the boundaries for each of the phases can be tuned in a simple, physically appealing way. Results in Figure 4 show how the fluid configuration develops in a one-dimensional pore for a situation when the black, dense, fluid (a) wets and (b) does not wet the walls. By modifying the functional form of  $\mu_{ex}(\underline{r})$ , for example by introducing long range interactions, equilibrium phases comprising of plugs, tubes and capsules discussed by Liu et. al. [15] can be reached. In addition, an important consequence of this formalism is that the fluid–boundary interface is diffuse. This allows for a reduction of many of the problems associated with surface orientation and standard bounce-back boundary conditions [16].

To summarise, we have described a lattice Boltzmann scheme, the main new features of which are the direct introduction of a non-ideal pressure tensor and an external chemical potential. This enables us to obtain an isothermal model of phase separation which correctly

describes bulk and interfacial dynamics at low temperatures. The method also provides a convenient, physically motivated way of tuning boundary conditions, giving a new approach to situations when flow and phase separation are affected by fluid-substrate interactions. Moreover, unphysical velocity oscillations at surfaces and interfaces are substantially reduced.

The simplicity of the method and the ease of implementation suggests that our approach may be a valuable tool in the study of multi-phase hydrodynamical systems. Furthermore, the introduction of a temperature parameter in our mechanism for interface formation should allow the formalism to be extended to non-isothermal situations where heat transfer is important [17]. We are currently in the process of investigating such a model.

We are indebted to Peter Coveney, Bruce Boghosian and Tim Newman for many stimulating discussions.



## REFERENCES

- [1] For a recent review see M. Sahimi, Rev. Mod. Phys. **65**,1393 (1993).
- [2] H. Furukawa, Adv. in Physics, **34**, 703 (1985); F.J. Alexander, S. Chen and D.W. Grunau, Phys. Rev. **B48**, R634 (1993).
- [3] H. Tanaka, Phys. Rev. Lett. **70**, 2770 (1993); D.W. Grunau, T. Lookman, S.Y. Chen and A.S. Lapedes, Phys. Rev. Lett. **71**, 4198 (1993).
- [4] A.K. Gunstensen and D.H. Rothman, J. Geophys. Res. **98**, 6431 (1993); S. Chen, K. Diemer, G. D. Doolen, K. Eggert, C. Fu, S. Gutman, and B. J. Travis, Physica **D47**, 72 (1991).
- [5] W.J. Ma, A Maritan, J.R. Banavar and J. Koplik, Phys. Rev. **A45**, R5347 (1992) and references therein.
- [6] R. Benzi, S. Succi and M. Vergassola, Phys. Rep. **222**, 145 (1992); Y. H. Qian, D. d’Humières and P. Lallemand, Europhys. Lett. **17**, 479 (1992).
- [7] A.K. Gunstensen, D.H. Rothman, S. Zaleski and G. Zannetti, Phys. Rev. **A43**, 4320 (1991); U. D’Ortona, D. Salin, M. Cieplak and J.R. Banavar, preprint (1994).
- [8] X. Shan and H. Chen, Phys. Rev. **E47**, 1815 (1993); X. Shan and H. Chen, preprint (1994). See also C. Appert and S. Zaleski, Phys. Rev. Lett. **64**,1 (1990).
- [9] A. K. Gunstensen, Ph. D. thesis, MIT (1992).
- [10] A.J. Bray, Adv. in Physics, in press.
- [11] J.S. Rowlinson and B. Widom, “Molecular Theory of Capillarity”, Clarendon Press, Oxford (1982).
- [12] R. Evans, Advances in Physics, **28**, 143 (1979).
- [13] P.L. Bhatnagar, E.P. Gross and M. Krook, Phys. Rev. **94**, 511 (1954); H. Chen, S. Chen

- and W.H. Matthaeus, Phys. Rev. **A45**, R5339, (1992).
- [14] L.D. Landau and E.M. Lifshitz, “Fluid Mechanics”, Pergamon Press (1959).
- [15] A.J. Liu, D.J. Durian, E. Herbolzheimer and S.A. Safran, Phys. Rev. Lett. **65**, 1897 (1990); A.J. Liu and G.S. Grest, Phys. Rev. **A44**, R7894 (1991).
- [16] R. Cornubert, D. d’Humières and D. Levermore, Physica **D47**, 241 (1991).
- [17] F. J. Alexander, S. Chen and J. D. Sterling, Phys. Rev. **E47**, R2249 (1993).

## FIGURE CAPTIONS

**Fig. 1** Equilibrium density profiles normal to a flat interface for a Van der Waals fluid for the three highest values of  $T$  shown on the coexistence curve (inset). The solid lines are numerical solutions of the continuum thermodynamic equations while the points are from the lattice Boltzmann simulations. The parameter values are  $a = 9/49$  and  $b = 2/21$ , while  $\kappa = 0.01$  for the bold curves and 0.02 for the dashed curve.

**Fig. 2**  $\Delta P$  plotted vs.  $1/R$  as a test of Laplace's Law. The solid line has a gradient calculated using equation (19).

**Fig. 3** The dispersion curve for capillary waves on an interface, plotted on a log-log scale. The best fit line has gradient  $1.6 \pm 0.05$ .

**Fig. 4** Time evolution (vertically downwards) of phase separation in a narrow capillary. In (a) the dense (dark) fluid wets the surfaces while in (b) non-wetting is illustrated. The simulations were performed on a  $128 \times 32$  lattice at  $T = 0.56$  with (a)  $\mu_{ex} = -0.1$  and (b)  $\mu_{ex} = 0.5$ .

Figure 1

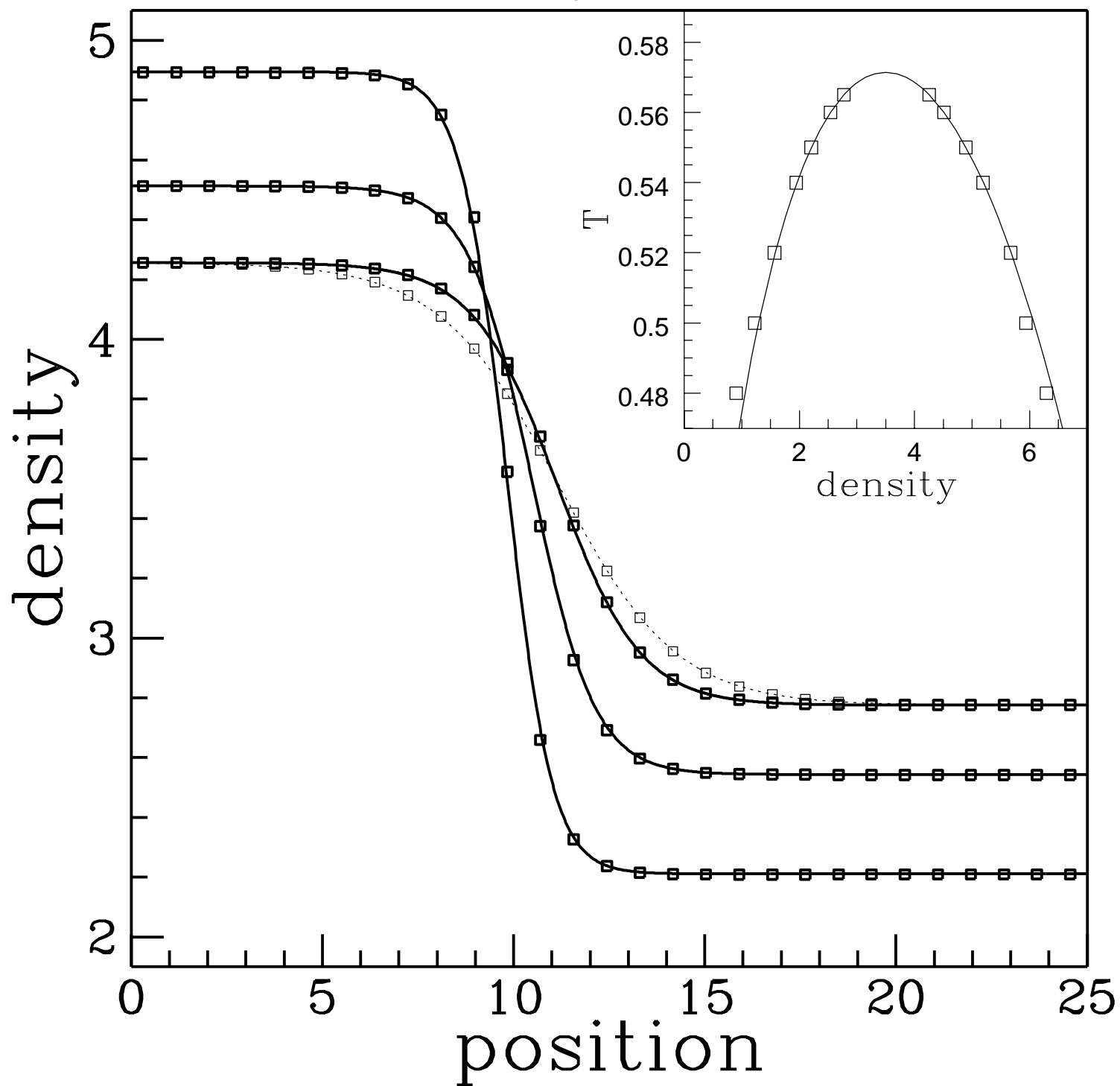


Figure 2

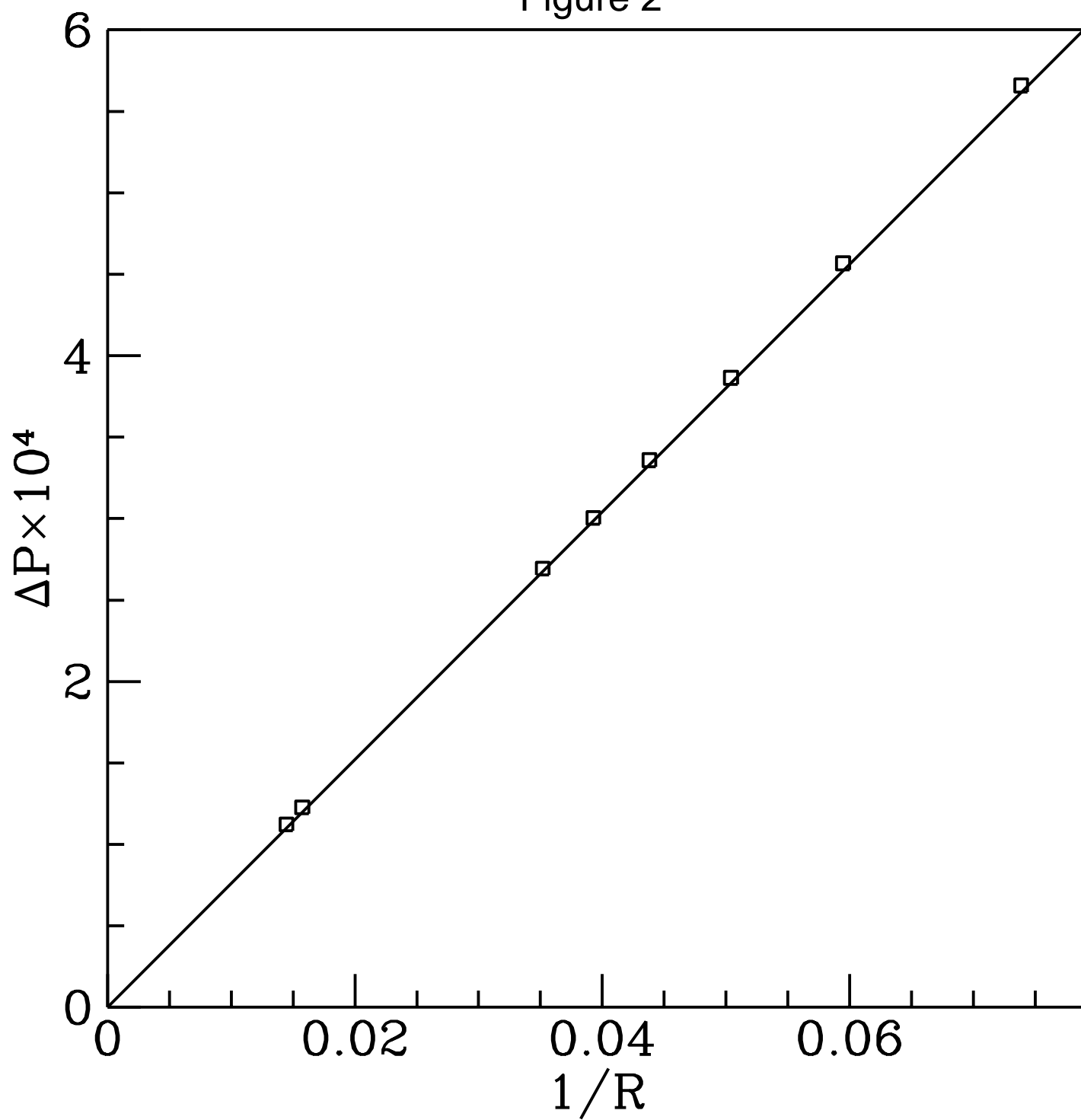


Figure 3

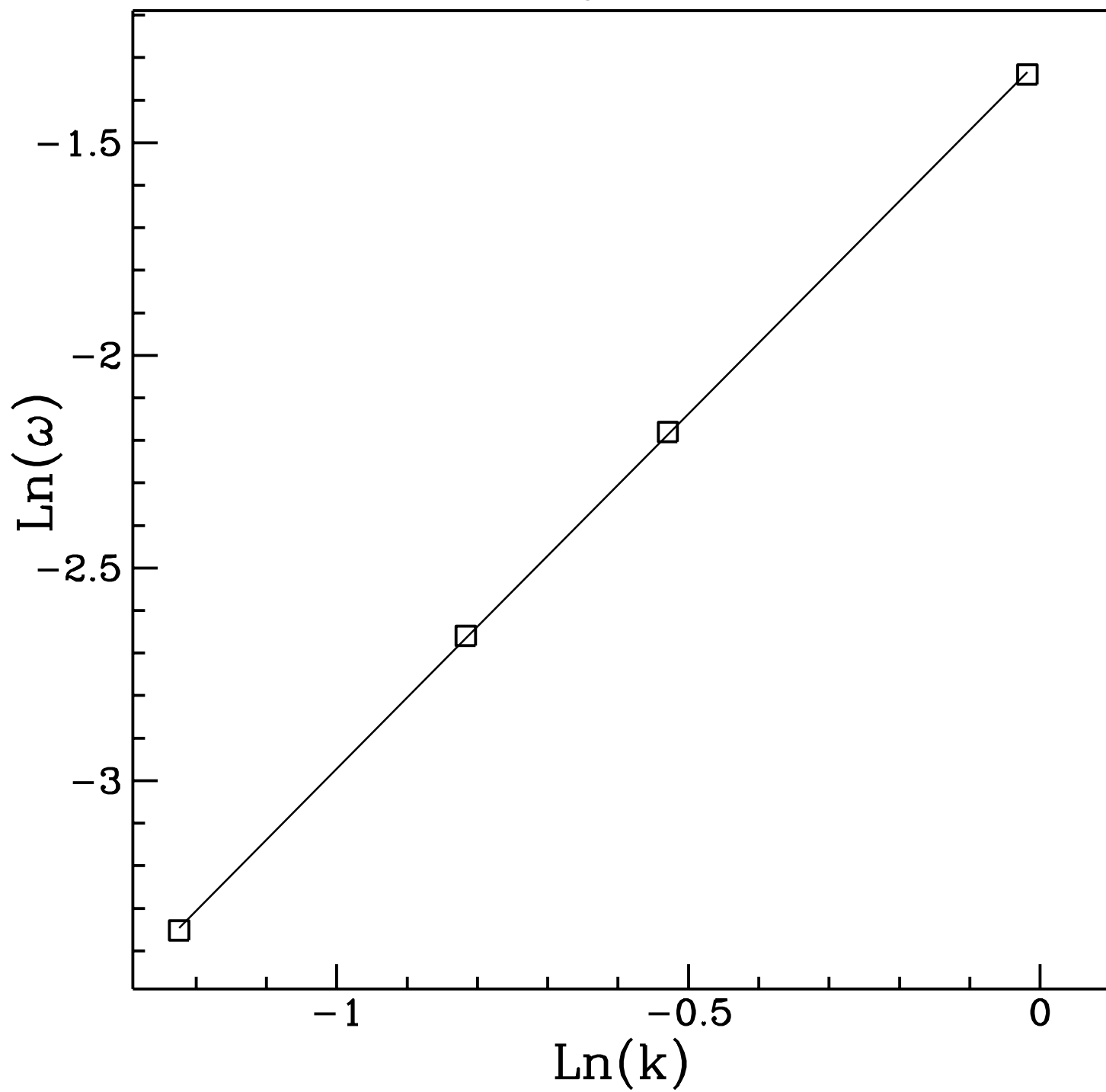
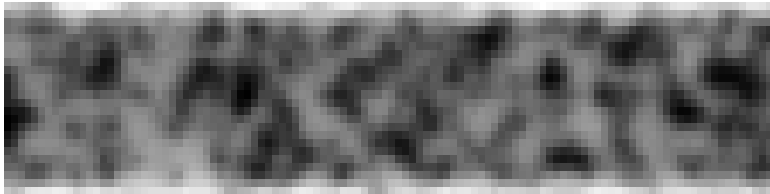
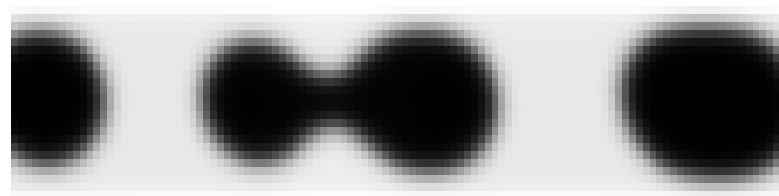
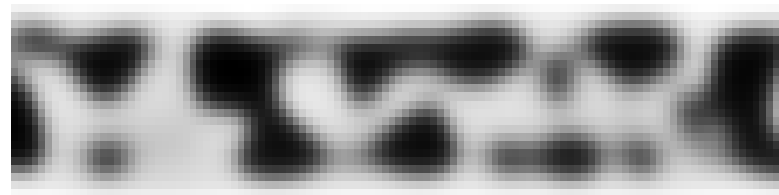
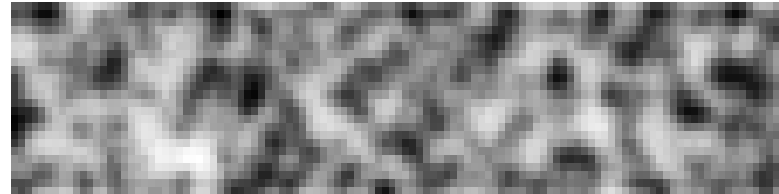


Figure 4



(a)



(b)