

# Two-dimensional computer studies of crystal stability and fluid viscosity\*

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The mathematical analogy between the elastic stress due to particle displacements in Hooke's law solids and the viscous stress due to velocity gradients in incompressible fluids correlates two interesting phenomena. In a two-dimensional crystal the elastic restoring force opposing particle displacements approaches zero with increasing crystal size, leading to a logarithmically diverging rms displacement in the large-system limit. The vanishing of the solid-phase force is mathematically analogous to the lack of viscous damping for a particle moving slowly through a two-dimensional incompressible fluid. These two *continuum* results are compared with *discrete-particle* computer simulations of two-dimensional solids and fluids. The divergence predicted by macroscopic elasticity theory agrees quantitatively with computer results for two-dimensional harmonic crystals. These same results can also be correlated with White's experimental study of the viscous resistance to a cylinder (a falling wire) moving slowly through a viscous fluid. The agreement is good. Finally, we carried out a computer study of a two-dimensional fluid confined between two moving walls (plane Couette flow). Despite theoretical predictions that transport coefficients in two-dimensional systems diverge, no viscosity anomalies were observed under the conditions of the computer simulation.

## I. INTRODUCTION

Surprising things can happen when three-dimensional physical ideas are applied to hypothetical two-dimensional systems. Here we consider a closely related pair of these surprises or paradoxes:

- (i) the rms displacement of a typical particle in a two-dimensional crystal diverges<sup>1</sup> as  $[\ln N]^{1/2}$  as the size of the  $N$ -particle crystal increases;
- (ii) the viscous resistance to a typical particle being slowly pushed through a two-dimensional viscous fluid is also anomalous,<sup>2,3</sup> approaching zero as  $1/\ln N$  as the size of the system increases.

These surprises contrast qualitatively with three-dimensional behavior. In three-dimensional solids the rms displacement of an atom approaches a finite limit (a fraction of a lattice spacing) in the large crystal limit, and the viscous resistance of a three-dimensional fluid to a sphere of diameter  $\sigma$  is given by Stokes' drag formula:

$$F = -3\pi\eta\sigma v, \tag{1}$$

where  $\eta$  is the shear viscosity and  $v$  is the (slow) velocity of the sphere.

In Sec. II we point out that the macroscopic mathematics of these two problems, elastic and viscous, is the same. In Sec. III we study the diverging rms displacement in the crystal from two different points of view: the microscopic view of lattice dynamics and the macroscopic view of continuum elasticity. We show that these

two crystal methods agree quantitatively on the precise nature of the divergence for large crystals. The crystal calculations can also be used to predict the frictional force on a cylinder drawn through a viscous fluid. We show that the force according to this viscous analogy is consistent with White's experimental data. The mathematical analogy between elasticity and viscosity suggests, in accord with theoretical predictions,<sup>4</sup> that two-dimensional viscous fluids might exhibit corresponding viscosity anomalies. In Section IV we use molecular dynamics to study such a fluid in order to investigate the possibility of anomalous behavior.

## II. LINEAR ELASTICITY AND SLOW VISCOUS FLOW

In this section we recall the "well-known" mathematical (as opposed to physical) similarity of two different physical systems: the idealized isotropic Hookean solid with reversible elastic forces proportional to displacements, and the idealized Newtonian fluid with irreversible viscous damping forces proportional to velocity gradients. The similarity breaks down if displacements in the solid are large enough for Hooke's law to fail or if the velocities in the fluid are large enough for inertial terms of order  $v^2$  to be significant.

The elastic solid is described by the phenomenological pressure tensor

$$\mathbf{P} = [P_0 - \lambda(\nabla \cdot \mathbf{v})] \mathbf{I} - \eta(\nabla \mathbf{v} + \nabla \mathbf{v}^T), \tag{2}$$

where  $P_0$  is the pressure when the elastic displacement  $\mathbf{v}$  is zero,  $\lambda$  and  $\eta$  are the Lamé elastic constants, and  $\mathbf{I}$  is the diagonal unit tensor. The transpose of the dis-

placement gradient tensor  $\nabla v$  is written  $\nabla v^T$ . The minimum energy principle<sup>5</sup> asserts that the displacement field actually found as a response to a set of specified displacements (boundary conditions) is that which minimizes the stored elastic energy:

$$\Phi \equiv \int \phi \, dr \quad ,$$

where the energy density  $\phi$  is quadratic in the displacement gradients:

$$\phi = \frac{1}{2} \lambda (\nabla \cdot v)^2 + \frac{1}{4} \eta (\nabla v + \nabla v^T) : (\nabla v + \nabla v^T) \quad . \quad (3)$$

The Newtonian viscous fluid is described by the same phenomenological pressure tensor

$$\mathbf{P} = [P_0 - \lambda (\nabla \cdot v)] \mathbf{I} - \eta (\nabla v + \nabla v^T) \quad , \quad (2')$$

but now  $\lambda$  and  $\eta$  are viscosity coefficients ( $\lambda = \eta_v - \frac{2}{3} \eta$  in three dimensions and  $\lambda = \eta_v - \eta$  in two dimensions where  $\eta_v$  is the "bulk" viscosity), and  $v$  is the hydrodynamic velocity. The minimum entropy production principle<sup>6</sup> asserts that the velocity field actually found subject to certain specified velocities (boundary conditions) is that which minimizes the production of entropy in the fluid:

$$\dot{S} \equiv \int \dot{s} \, dr \quad ,$$

where the density of entropy production  $\dot{s}$  is quadratic in the velocity gradients:

$$\dot{s} = (\lambda/T) (\nabla \cdot v)^2 + \frac{1}{2} (\eta/T) (\nabla v + \nabla v^T) : (\nabla v + \nabla v^T) \quad . \quad (3')$$

$T$  is the temperature.

The similarity of the two sets of equations is noteworthy, but also a little deceptive. In the elastic case the response of stress to volume and shape changes is described by the Lamé constants and  $P_0$  is constant (usually zero). In the viscous case  $P_0$  is interpreted differently. It varies with  $r$  and is the local equilibrium pressure, a function of density and temperature. The compressibility of the fluid is not explicitly displayed. In the special case of an *incompressible* fluid the elastic and viscous formulations coincide. Then the solution of an elastic (viscous) problem can be used to generate the corresponding viscous (elastic) solution. As an example, consider the Stokes drag exerted by a viscous incompressible fluid on a slowly moving sphere of diameter  $\sigma$ :

$$\mathbf{F} = -2\pi\eta\sigma v \quad (\text{slipping boundary condition}); \quad (1')$$

$$\mathbf{F} = -3\pi\eta\sigma v \quad (\text{sticking boundary condition}). \quad (1)$$

The velocity field set up by the moving sphere corresponds exactly to the displacement field obtained in an elastic solid by displacing a rigid sphere of material a distance  $v$ .

In the following section we exploit the analogy between the viscous and elastic problems in two rather than three dimensions in order to demonstrate that two apparently dissimilar and interesting phenomena are mathematically one and the same.

### III. ELASTICITY AND VISCOUS FLOW IN TWO DIMENSIONS

Consider the slow steady motion of a two-dimensional disk through a two-dimensional incompressible viscous

fluid. Over 100 years ago, Stokes found paradoxical results ("Stokes' Paradox") for this system.<sup>7</sup> We will see that, with a particular choice of boundary conditions, the larger the system in which the disk moves, the smaller the restoring force, with the force disappearing in the large-system limit. This surprising two-dimensional result has a simple three-dimensional analog—the slow steady motion of a very long cylinder through a three-dimensional viscous fluid. The motion is perpendicular to the axis of the cylinder.

Long after Stokes' work on viscous drag, Peierls, Frenkel, and others pointed out that the restoring force on a displaced particle in a two-dimensional elastic solid disappears in the large-system limit, with the formal result that the rms displacement diverges. The lack of dependence of the restoring force on details of order the atomic size is common to both the elastic and viscous problems. As we will see, the macroscopic nature of these two-dimensional displacement and velocity fields furnishes an exact connection between the macroscopic elastic constants and the microscopic rms displacement.

The displacement created by a force  $F$  applied at the origin along the  $x$  axis can be used to express the stored elastic energy in a two-dimensional continuum. If we consider a system of size  $R$  (i.e., with the volume proportional to  $R^2$  with proportionality constant determined by the crystal's shape), with interatomic spacing  $r_0$ , the stored energy turns out to be (see Appendix A)

$$\Phi = \{(\lambda + 3\eta)/[(\lambda + 2\eta)(8\pi\eta)]\} F^2 [\ln(R/r_0) + O(1)] \quad , \quad (4)$$

proportional to  $\ln N$ , with  $N$  the number of particles in the crystal. Because the crystal obeys Hooke's law, the energy is of the form  $\Phi = \frac{1}{2} k x^2 = \frac{1}{2} F^2/k$ , where  $k$  is the force constant for the motion. The large-system value of the force constant  $k$  then follows from (4):

$$k = (8\pi\eta/\ln N) (\lambda + 2\eta)/(\lambda + 3\eta) \quad . \quad (5)$$

Thus the finite-crystal force constant is given in terms of crystal size and the macroscopic elastic constants. We can verify the expression (5) in two ways: by a comparison with exact results for small crystals calculated with lattice dynamics, and (for the special case  $\lambda \gg \eta$ ) by comparison with the analogous hydrodynamic problem of the Stokes drag on a cylinder.

The lattice dynamics results for small two-dimensional crystals were generated several years ago in a study of the thermodynamic properties of one-, two-, and three-dimensional crystals.<sup>8</sup> The two-dimensional crystal is periodic, composed of  $N$  particles linked to their six nearest neighbors (hexagonal lattice) by Hookean springs of energy  $\frac{1}{2} \kappa (\delta r)^2$ . For such a crystal we can compare the actual mean squared displacement to that calculated in the Einstein approximation in which a single particle moves in the field of its fixed neighbors. The results from the lattice-dynamics calculations are given in Table I and plotted in Fig. 1. The results can be fitted by the following empirical relation:

$$\langle r^2 \rangle / \langle r^2 \rangle_{\text{Einstein}} \doteq 0.28 + 0.368 \ln N \quad . \quad (6)$$

If we *calculate* the elastic constants for the same har-

TABLE I. Mean squared displacement relative to the Einstein approximation for  $N$ -particle harmonic crystals with nearest-neighbor interactions. These crystals are periodic, with fixed center of mass, and rectangular, with height/width =  $\sqrt{3}$ , as shown in Fig. 1.

$N$	Ratio	$N$	Ratio	$N$	Ratio
2	0.50000	72	1.86604	242	2.31056
8	1.07500	98	1.97888	288	2.37447
18	1.36438	128	2.07679	338	2.43327
32	1.57104	162	2.16321	392	2.48772
50	1.73296	200	2.24056	450	2.53841

monic crystal (see Appendix B) we find

$$\lambda = \eta = \frac{1}{4}\sqrt{3}\kappa \quad (7)$$

which provides a theoretical estimate for the coefficient of  $\ln N$ , namely  $2/(\sqrt{3}\pi) = 0.367553$ , reproducing the results from lattice dynamics. Thus the lattice dynamics results agree *exactly* (for large  $N$ ) with the results of the continuum theory. This unexpected agreement occurs because the displacements are large (of order  $[\ln N]^{1/2}$ ) with respect to the interparticle spacing so that the discrete nature of the lattice can be ignored. In a three-dimensional crystal one expects that lattice dynamics and elasticity theory will differ by a factor of order 1. The agreement found in the two-dimensional case is an indirect check of both the elastic and the lattice-dynamics calculations.

As a second verification of Eq. (5) for the effective force constant, we exploit the analogy with incompressible hydrodynamics. Consider the drag force on an infinitely long cylinder moving perpendicular to its axis through an incompressible viscous fluid. The force per unit length of cylinder corresponds to the drag force for a two-dimensional disk moving through a two-dimensional fluid. It is a "well-known" result, "Stokes' Paradox", that Eqs. (2') and (3') have no solution satisfying the boundary conditions of velocity  $\mathbf{v}$  at the disk surface and velocity 0 at infinity. Physically, as Stokes pointed out,<sup>7</sup> the difficulty is that a slowly moving cylinder in an infinite fluid gradually carries more and more fluid with it as time goes on so that a steady state is never reached. Actually, neglected nonlinear terms proportional to  $v^2$  eventually (at a distance  $> \eta/\rho_0 v$ , where  $\rho_0$  is the mass density) dominate the terms retained and, as Lamb showed, the resistance on a disk is finite, proportional to  $v/\ln(1/v)$  for small  $v$ .

Our own result (5) predicts a force varying as  $\mathbf{v}/\ln N$  instead. The crucial difference between Lamb's result and ours is the extent of the flow region. The linearized equation of Stokes is only appropriate for small distances ( $\ll \eta/\rho_0 v$ ). Lamb's calculation is correct for a fluid infinite in extent. Because Lamb included nonlinear inertial terms, his resistance calculation has no simple elastic analog.

Our two-dimensional results should be approximated by the resistance to a cylinder moving between two channel walls separated by a distance of order  $N^{1/2}$ . White<sup>3</sup> has carried out such an experiment, dropping wires of

diameter  $\sigma$  both in cylinders of diameter  $D$  and between planar walls a distance  $D$  apart. White found a boundary influence upon fluid drag for  $\text{Re}D/\sigma < \sim 40$ , where the Reynolds number  $\text{Re}$  is  $\rho_0 v \sigma / \eta$ . For larger  $D$  Lamb's formula applied. For finite flow regions with  $\text{Re}D/\sigma < \sim 3$ , White's results are described by the equation

$$F = -4\pi\eta\mathbf{v}/[\ln(D/\sigma) + O(1)], \quad (8)$$

where the term of order 1 takes the flow region shape into account. White actually found the form (8) empirically, and with a slightly different value for the multiplicative constant  $4\pi$ . Taking into account the scatter of White's data leads to an experimental determination of the coefficient in (8),  $4\pi \pm 15\%$ . Our own elasticity equation, when specialized to the case of an incompressible material ( $\lambda \gg \eta$ ) shows that the large- $N$  force constant is indeed  $8\pi\eta/\ln N \sim 4\pi\eta/\ln(D/\sigma)$ , consistent with White's experimental results for both cylindrical and planar geometries.

The corresponding elastic calculation in three dimensions does not seem to be well known. Although the Stokes drag on a sphere appears in several texts, with both slipping and sticking boundary conditions, the corresponding elasticity problem has not been studied explicitly. If one simply calculates the displacement of a spherical surface in an elastic continuum responding to a force applied within it, the force constant found is too small. The discrepancy is due to the deformation of the spherical region. It would be an interesting exercise to verify that the constraint of constant shape on a displaced sphere in an elastic continuum does result in the force constants calculated for the viscous Stokes drag problem, Eqs. (1) and (1').

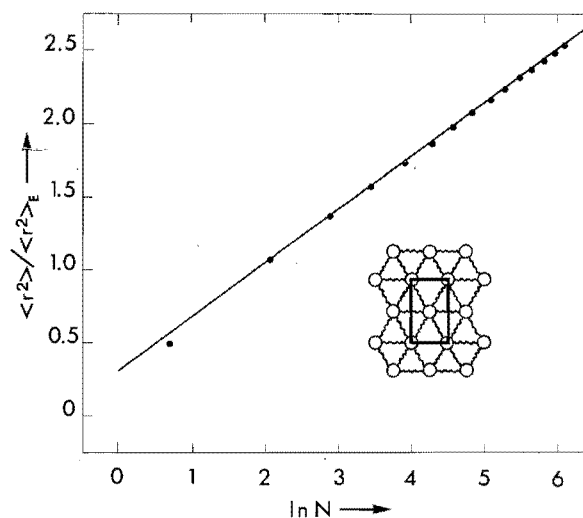


FIG. 1. Ratio of the mean squared displacement to the Einstein approximation,  $\langle r^2 \rangle_E = 2k_B T / 3\kappa$ , where  $k_B$  is Boltzmann's constant and  $\kappa$  is the nearest-neighbor spring constant. The empirical slope of the curve is 0.368, in good agreement with the value derived in the text,  $(4/3)^{1/2}/\pi = 0.367553$ . The inset shows the crystal structure. The crystals were built up using an  $n$  by  $n$  array of rectangles like that heavily outlined in the figure. The data cover values of  $N = 2n^2$  from 2 to 450.

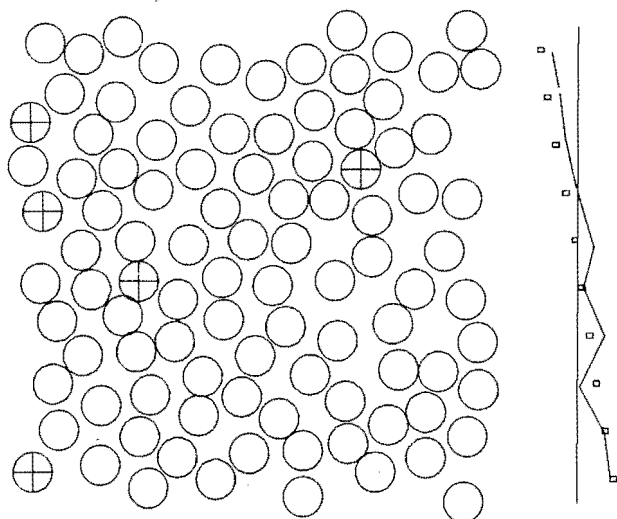


FIG. 2. Typical movie frame showing 98 soft disks undergoing plane Couette flow. Five particles are marked with crosses to make the motion easier to follow in the movie. 28 particles, not shown in the figure, drive those shown and are in turn interacting collectively with external momentum and heat reservoirs. At the right side of the figure the instantaneous velocity profile (solid line) and its long-time average (small squares) [for a time  $190\sigma(m/\epsilon)^{1/2}$ ] are shown. The particles have mass  $m$  and interact with a pair potential  $\epsilon(\sigma/r)^{12}$ . The circles are drawn with diameter  $\sigma$ , and the boundary temperature is  $\epsilon/k$ . The upward flow of horizontal momentum is responsible for the preponderance of near-particle pairs in the direction  $x=y$  over the number of pairs in the direction  $x=-y$ . The viscosity is determined from the cumulative average shear stress and the measured velocity gradient,  $\eta \equiv -P_{xy}/u_{x,y}$ .

#### IV. COUETTE FLOW IN TWO DIMENSIONS

The vanishing of the viscous resistance on a moving disk is reminiscent of paradoxical results obtained from Kubo's theory of transport when that theory is applied to two-dimensional transport coefficients. In Kubo's theory the coefficients are expressed as time integrals of correlation functions involving particle coordinates and velocities. These integrals diverge in the two-dimensional case,<sup>4</sup> suggesting that the transport coefficients themselves are in some sense divergent (that is, that the linear transport equations are invalid for infinite two-dimensional systems). The physical significance of this divergence is difficult to assess. If it is related to the divergence of the rms displacement in two-dimensional crystals, then it is certainly possible that transport coefficients are both useful and well-defined in *finite*, as opposed to infinite, systems. To indicate that there is a very significant difference between large and infinite systems, consider a large two-dimensional crystal of 3 Å diameter particles. The Einstein prediction for the rms displacement would be small, of order 0.5 Å at the melting temperature. The actual rms displacement would be larger by a factor of only  $(\ln N)^{1/2}$ ; that is, about 13, if the crystal were the size of the known universe ( $\sim 10^{10}$  light years in radius)! Thus a very large two-dimensional crystal would still have an rms displacement less than 10 Å.

We set out to examine viscosity in nonequilibrium two-

dimensional systems to search for anomalous behavior. The only method available for such a study is nonequilibrium molecular dynamics. A system of particles, such as that shown in Fig. 2, is followed for tens of thousands of time steps (a time step corresponding roughly to one tenth of a mean vibration time) by integrating the classical equations of motion. Figure 2 is a single frame from a computer movie of 98 soft-disk particles undergoing planar Couette flow. The 98 particles shown are driven by 28 additional particles (14 above and 14 below the square region shown in the movie frame). External forces and thermostats are used to maintain the average velocity and temperature of particles in the two boundary regions.<sup>9</sup> The upward flux of  $x$  momentum quickly produces a velocity gradient. From the average value of the gradient,  $u_{x,y}$ , the viscosity can be calculated, using  $P_{xy} = -\eta u_{x,y}$ , with  $P_{xy}$  the momentum flux.

In order to decide whether or not such a system is usefully described by linear hydrodynamic equations we must answer the questions:

- (i) is the velocity profile linear?
- (ii) does the viscosity vary with the size of the system?
- (iii) does the viscosity vary with the gradient  $u_{x,y}$ ?

The answers to these questions cannot be derived from numerical data alone. Statistical fluctuations, no matter how small, might mask still smaller effects. The fluctuations found in the two-dimensional systems are in fact rather large, making it impossible to detect anomalous behavior if the anomaly is less than about 10%. The results from a series of molecular dynamics runs with 32, 50, 98, and 392 soft-disk particles [interparticle potential  $= \epsilon(\sigma/r)^{12}$ ] are given in Table II. The viscosities found all have uncertainties of order 10% and show no clearcut dependence on either size or velocity gradient. Note that  $\ln N$  varies from 3.5 to 6.0, a 72% increase, for the range of  $N$  studied.

In order to see whether the viscosity coefficient found

TABLE II. Shear viscosity and compressibility factor of  $N$ -particle soft-disk systems with pair potential  $\epsilon(\sigma/r)^{12}$  at reduced density  $(3/4)^{1/2} N\sigma^2/V = 0.6$ . The notation  $N = 32 + 8$  indicates 32 particles in the central region of the system with 4 particles in each of the two boundary regions. The total length of the calculation is the time  $t$ . Uncertainties in the compressibility factor,  $PV/NkT$ , are 1% or less. For the 392-disk system we estimated the thermal conductivity from the parabolic temperature profile. The result is  $5(k_B/\sigma)(\epsilon/m)^{1/2}$  with an uncertainty of order 50%; the Enskog prediction is about twice as big (see Refs. 9 and 11).  $\rho = .6928$

$N$	$\sigma u_{x,y}(m/\epsilon)^{1/2}$	$kT/\epsilon$	$\eta\sigma/(m\epsilon)^{1/2}$	$t(\epsilon/m)^{1/2}/\sigma$	$PV/NkT$
32+8	0.072	1.06	$1.00 \pm 0.10$	544	5.18
32+16	0.075	1.00	$1.23 \pm 0.08$	544	5.51
50+20	0.043	0.49	$1.12 \pm 0.08$	1539	7.37
50+20	0.033	1.00	$0.92 \pm 0.21$	408	5.54
50+20	0.056	0.98	$1.22 \pm 0.09$	680	5.55
50+20	0.074	1.47	$1.05 \pm 0.11$	888	4.81
98+28	0.041	0.99	$1.14 \pm 0.09$	1522	5.49
98+28	0.181	0.71	$0.94 \pm 0.02$	571	5.41
392+56	0.042	0.94	$1.02 \pm 0.05$	571	5.46

is reasonable we compare the measured result,  $\eta = (m\epsilon)^{1/2}/\sigma \pm 10\%$ , with two simple theoretical predictions. Andrade<sup>10</sup> suggested that viscosity can be estimated by considering a particle to oscillate at the Einstein frequency, transferring transverse momentum to its neighbors at each turning point. This argument, applied to our two-dimensional problem, leads to the result:

$$\eta(\text{Andrade}) = 2m\nu_{\text{Einstein}} = 6.618(m\epsilon)^{1/2} \rho^{7/2}/\sigma, \quad (9)$$

which is  $1.1(m\epsilon)^{1/2}/\sigma$  at the density,  $\rho = (3/4)^{1/2} N\sigma^2/V = 0.6$ , of the computer runs. A second approximate estimate can be obtained from Enskog's theory,<sup>11</sup> using a corrected hard-disk viscosity coefficient to estimate the low-density viscosity coefficient for our soft disks. The Enskog estimate,  $1.0(m\epsilon)^{1/2}/\sigma$  at  $\epsilon = kT$ , is not very different from Andrade's. If Enskog's theory is used to predict the temperature dependence of  $\eta$ , the increase is marginally greater than that consistent with the few computer results in Table II. This same defect of the Enskog theory appears when the theory is used to predict the temperature dependence of viscosities for real three-dimensional fluids.

Although the uncertainty in the computer experiments is relatively large, our results do rule out the possibility that the viscosity varies as strongly with  $N$  as  $\ln N$ . The velocity profiles associated with the calculations are also linear within the statistical fluctuations. We conclude that the average flow patterns found in small two-dimensional systems can be described (with at least 10% accuracy) using the equations of ordinary hydrodynamics. The relation between equilibrium and nonequilibrium molecular dynamics still needs clarification. At equilibrium the correlation functions contributing to Kubo's formulation of the transport coefficients have "long-time tails" so that the coefficients diverge. The nature of this divergence, particularly its dependence on system size and boundary conditions, deserves thorough investigation. (See "Noted added in proof," Ref. 13.)

## APPENDIX A

In this Appendix we outline the derivation of Eq. (4) of the text, the elastic energy stored in a two-dimensional Hookean solid of sidelength  $R$  with a force  $F$  applied at the origin. Timoshenko and Goodier<sup>12</sup> furnish our starting point, the stress field in an infinite plate subject to a force applied at the origin. The stress is converted to strain using the phenomenological pressure tensor of the text. The energy stored is then expressed in polar coordinates. After the angular integration, we find

$$\Phi = [F^2(3 - \nu)/(16\pi\eta)] \int d\ln r, \quad (\text{A1})$$

where  $\nu$  is Poisson's ratio,  $\nu = \lambda/(\lambda + 2\eta)$  in two dimensions. If we arbitrarily cut off the integration, at an atomic diameter for small  $r$ , and at a distance of order  $N^{1/2}$  for large  $r$ , we obtain Eq. (4) of the text. The most direct justification for the cutoffs is the comparison with the lattice dynamics results given in the text.

## APPENDIX B

In this Appendix we calculate the Lamé constants for a two-dimensional harmonic hexagonal crystal in which particles interact with nearest-neighbor Hookean springs.

The spacing between atoms is  $d$ , related to the volume per atom by  $V/N = (3/4)^{1/2} d^2$ . Consider first a uniform expansion of the crystal in which all lengths undergo a relative extension  $\delta L/L = \Delta$ . The macroscopic energy calculated from Eq. (3) of the text is

$$\Phi = [\frac{1}{2}\lambda(2\Delta)^2 + \frac{1}{4}\eta\{(2\Delta)^2 + (2\Delta)^2\}] V = 2\Delta^2(\lambda + \eta) V. \quad (\text{B1})$$

The microscopic approach, in which we notice that each spring is stretched by an amount  $d\Delta$ , gives an alternative expression for the energy:

$$\Phi = (3N)(\frac{1}{2}k d^2 \Delta^2) = \sqrt{3} k \Delta^2 V. \quad (\text{B1}')$$

Combining the two relations (B1) and (B1') gives the result

$$\lambda + \eta = (3/4)^{1/2} k. \quad (\text{B2})$$

We obtain the shear modulus  $\eta$  by considering the deformation

$$\nabla \mathbf{v} = \begin{pmatrix} 0 & \Delta \\ 0 & 0 \end{pmatrix}.$$

The macroscopic energy for this deformation is  $\frac{1}{2}\Delta^2 \eta V$ . The microscopic energy is  $(3/16) N k d^2 \Delta^2$ , leading to the identification

$$\eta = \frac{1}{4} \sqrt{3} k. \quad (\text{B3})$$

Equations (B2) and (B3) establish Eq. (7) of the text:

$$\lambda = \eta = \frac{1}{4} \sqrt{3} k. \quad (7)$$

\*Work performed under the auspices of the U. S. Atomic Energy Commission.

<sup>1</sup>J. Frenkel, *Kinetic Theory of Liquids* (Dover, New York, 1955), p. 119.

<sup>2</sup>H. Lamb, *Hydrodynamics* (Dover, New York, 1945), Chap. XI.

<sup>3</sup>C. M. White, Proc. R. Soc. A 186, 472 (1946).

<sup>4</sup>This divergence should not be confused with the divergence of coefficients in a density expansion of the form  $\eta = \eta_0 + \eta_1 \rho + \eta_2 \rho^2 + \dots$ . The divergence of the Kubo expression for  $\eta$  would imply that the Newtonian relation between the pressure tensor and strain-rate tensor does not hold, even at small rates of strain, so that large-system small strain-rate measurements of  $\eta$  would produce a limiting value of either 0 or  $\infty$  for  $\eta$ . See, for example, W. W. Wood, Acta Phys. Austriaca, Suppl. 10, 451 (1973) and references listed in T. Keyes and I. Oppenheim, Phys. Rev. A 8, 937 (1973).

<sup>5</sup>L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, London, 1959), p. 11.

<sup>6</sup>T. Glandsdorff and I. Prigogine, Physica 30, 351 (1964).

<sup>7</sup>G. G. Stokes, *Mathematical and Physical Papers* (Cambridge U. P., Cambridge, England, 1901), Vol. III, p. 1.

<sup>8</sup>W. G. Hoover, J. Chem. Phys. 49, 1981 (1968).

<sup>9</sup>W. T. Ashurst, Ph. D. thesis, University of California at Davis-Livermore (in preparation); see also W. T. Ashurst and W. G. Hoover, Phys. Rev. Lett. 31, 206 (1973).

<sup>10</sup>E. N. da C. Andrade, *Viscosity and Plasticity* (Chemical Publ. Co., New York, 1951), p. 18.

<sup>11</sup>D. M. Gass, J. Chem. Phys. 54, 1898 (1971).

<sup>12</sup>S. Timoshenko and J. N. Goodier, *Theory of Elasticity* (McGraw-Hill, New York, 1951), p. 114.

<sup>13</sup>Note added in proof: We thank Tom Keyes of MIT for some helpful correspondence. Keyes has considered the size dependence of the shear viscosity in two dimensions using the "mode-mode coupling theory." This approximate theory predicts an increase of about 7% in  $\eta$  as the number of particles increases from 40 to 448. The predicted increase is just too small to be unambiguously detected in the computer experiments.