Hamiltonian dynamics of thermostated systems: Two-temperature heat-conducting ϕ^4 chains

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(Received 31 January 2007; accepted 7 March 2007)

We consider and compare four Hamiltonian formulations of thermostated mechanics, three of them kinetic, and the other one configurational. Though all four approaches "work" at equilibrium, their application to many-body nonequilibrium simulations can fail to provide a proper flow of heat. All the Hamiltonian formulations considered here are applied to the same prototypical two-temperature "ϕ^4" model of a heat-conducting chain. This model incorporates nearest-neighbor Hooke's-Law interactions plus a quartic tethering potential. Physically correct results, obtained with the isokinetic Gaussian and Nosé-Hoover thermostats, are compared with two other Hamiltonian results. The latter results, based on constrained Hamiltonian thermostats, fail to model correctly the flow of heat. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2720839]

I. INTRODUCTION

Computational "thermostats" arose as a means for controlling numerical simulations of both equilibrium and nonequilibrium stationary states. *Without* thermostats systems driven away from equilibrium heat up. *With* thermostats the heat generated by irreversible processes can be steadily removed, making it possible to simulate nonequilibrium steady states.[1,2] Because most Hamiltonian-based mechanics problems conserve energy, novel *nonHamiltonian* ideas are typically required when thermostats are to be included. Nevertheless, several approaches to Hamiltonian thermostats have been developed. Here we consider various approaches pioneered by Ashurst, Dettmann, Evans, Hoover, Leete, Morriss, Nosé, and Woodcock over a span of about 25 years.[1–10]

The simplest thermostat type maintains a (nearly) constant kinetic energy by "rescaling" the velocities at the end of each computational time step. For a more elegant, but equivalent, *continuous* approach to rescaling, see the "Gaussian isokinetic" method described in Sec. II B and illustrated in Sec. VII. For # Cartesian degrees of freedom the kinetic-theory relation

$$K = \sum^{\#} \frac{m\dot{q}^2}{2} = \sum^{\#} \frac{p^2}{2m} = \# \frac{kT_{\rm kin}}{2},$$

defines the kinetic temperature $T_{\rm kin}$. Thermostats based on $T_{\rm kin}$ can be applied to an entire system, or separately to two or more subsystems. With this *ad hoc* rescaling approach

$$p \to p_0 \sqrt{(K/K_0)},$$

there is no difficulty in maintaining several temperatures in specified parts of a single nonequilibrium system, as did Ashurst in his Ph.D. thesis research.[11]

The kinetic theory and the Gibbs-Boltzmann development of thermodynamics based on ideal-gas thermometry certainly suggest that the kinetic temperature, $T_{\rm kin}$, is both the simplest and the most fundamental of the many possible "temperature" types. There are alternatives. Among the many possible temperatures based on particle coordinates, rather than velocities, the simplest—on which we focus here—is based on the mean-squared forces, $\langle F^2 \rangle$. This definition of a configurational temperature is derived directly from the potential energy Φ and its space derivatives

$$kT = \langle (\nabla\Phi)^2 \rangle / \langle \nabla^2 \Phi \rangle = - \langle F^2 \rangle / \langle \nabla F \rangle .$$

This somewhat cumbersome relation appears as an aside in Landau and Lifshitz' classic 1958 text.[12] It was also recently rediscovered by Rugh, and has subsequently been much discussed.[13–16] This configurational representation of temperature follows most simply from an integration by parts, in the canonical ensemble:

$$\int dq \nabla^2 \Phi = \nabla\Phi ; \; (-d/dq) e^{-\Phi/kT}$$

$$= (\nabla\Phi/kT) e^{-\Phi/kT} \to \int dq \nabla^2 \Phi e^{-\Phi/kT}$$

$$= (1/kT) \int dq (\nabla\Phi)^2 e^{-\Phi/kT}.$$

About ten years after Woodcock and Ashurst's introduction of the rescaling isokinetic thermostat in the early 1970s, Hoover and Evans discovered that the isokinetic velocity-rescaling equations of motion can be derived from Gauss' Principle of Least Constraint.[5] Fifteen years after this discovery, Dettmann and Morriss[4] found a straightforward Hamiltonian approach to these same motion equations. Previously, Leete and Hoover had derived a Hamiltonian formulation which likewise maintained the velocity-based kinetic energy $K(\{\dot{q}\})$ constant.[6,8]

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A Hamiltonian generating the Gauss' principle isokinetic equations of motion was not found until 1996.[4] In the meantime Nosé had discovered his canonical-ensemble thermostated dynamics, which is related to two rather different Hamiltonians.[7,9] The purpose of the present work is twofold. First, we explore the relation of the Hoover-Leete kinetic thermostat work to its successors. A 2007 literature search shows only a single reference to it.[6] We also explore the consequences of a "Landau-Lifshitz" configurational thermostat based on the canonical-ensemble definition rediscovered by Rugh with a relatively complicated measure-based approach using the *microcanonical* ensemble. Recent pedagogical reviews of this configurational-temperature work can be found in Refs. 13–16.

II. THE NOSÉ-HOOVER THERMOSTAT

The Nosé-Hoover equations of motion[1,9] are a standard approach to carrying out canonical-ensemble dynamics at a particular temperature T_0. This approach uses *feedback*, controlling the fluctuating kinetic energy with a "friction coefficient" or "control variable" ζ. The frictional feedback force is $-\zeta p$. The time rate of change of the friction coefficient is based on the current value of the kinetic energy $K(t)$ relative to the desired mean value $K_0(T_0)$. The equations of motion also include a characteristic time τ which determines the time-range over which the feedback acts

$$\{\dot q = p/m; \quad \dot p = F - \zeta p\}; \quad \dot\zeta = [(K/K_0) - 1]/\tau^2.$$

It is quite interesting to see that a canonical-ensemble approach[1] to a Nosé-Hoover-type *configurational* thermostat led Braga and Travis[14] to the motion equations (for a harmonic oscillator with unit mass, force constant, and temperature)

$$\dot q = p - \zeta q; \quad \dot p = -q; \quad \dot\zeta = [q^2 - 1]/\tau^2.$$

The substitution $(+q, +p, +\zeta, +t) \rightarrow (-p, -q, -\zeta, -t)$ gives

$$\dot p = -q - \zeta p; \quad \dot q = +p; \quad \dot\zeta = [p^2 - 1]/\tau^2,$$

exactly the same as the Nosé-Hoover equations for the *kinetic* thermostating of an oscillator!

In most cases, including our ϕ^4 model studies described in detail in Secs. V–IX, a reasonable choice of τ corresponds to a typical collision time or vibration time. Such a choice typically provides the Gibbs' equilibrium canonical phase-space distribution with a Gaussian distribution for the friction coefficient ζ

$$f(q,p,\zeta) = e^{-\Phi/kT} e^{-K/kT} e^{-\#\zeta^2\tau^2/2}.$$

Evidently the amplitude of the fluctuations of ζ varies as $\sqrt{1/\#\tau^2}$, where # is the number of degrees of freedom. Thus its effect on the dynamics vanishes in the large-system limit.

Nonequilibrium situations are quite different. By using two or more different friction coefficients (or "thermostat variables") temperature differences can be established, leading to heat flow. Then the corresponding phase-space distributions are no longer smooth and Gibbsian, but instead become fractal, with the underlying phase-space trajectories satisfying the second law of thermodynamics infinitely more probable than those violating the law.[2] In the equilibrium case there is a close connection of Nosé-Hoover mechanics to Hamiltonian mechanics. We describe this connection next.

A. The Dettmann-Nosé Hamiltonian

In 1984 Nosé discovered a Hamiltonian consistent with Gibbs' constant-temperature canonical ensemble

$$f(q,p) \propto \exp(-\mathcal{H}_{\text{Nosé}}/kT).$$

He was able to derive the Nosé-Hoover equations of motion described above from his Hamiltonian by an artificial "time scaling."

In July of 1996 Dettmann[7] discovered a simpler approach to the Nosé-Hoover equations of motion. Dettmann's *vanishing* Hamiltonian is

$$\mathcal{H}_{\text{Dettmann}} \equiv s\mathcal{H}_{\text{Nosé}} = \sum \frac{p^2}{2ms} + s\left[\Phi + \frac{p_s^2}{2M} + \#kT\ln s\right] \equiv 0.$$

Provided that Dettmann's Hamiltonian is set equal to the special value, zero, Nosé's time-scaling variable s can be eliminated. Then the Nosé-Hoover equations of motion result *without the need for any time scaling*. It needs to be emphasized that this $\mathcal{H}=0$ trick does *not* work for the prototypical *nonequilibrium* case of a system with two different temperatures. In a two-temperature system the two different variables $s_{\text{Hot/Cold}}$ are necessarily coupled and cannot both be eliminated, so that Dettmann's $\mathcal{H}=0$ trick does not work.

There is a special case of Nosé-Hoover dynamics that deserves special mention, the "isokinetic" case in which the temperature is constant. This corresponds to the choice $\tau \rightarrow 0$. For isokinetic mechanics Dettmann and Morris discovered the special Hamiltonian detailed next.

B. The Dettmann-Morriss isokinetic Hamiltonian

Soon after Dettmann discovered a Hamiltonian route to the Nosé-Hoover motion equations, Dettmann and Morriss[4] discovered a related approach to isokinetic (constant temperature, provided that temperature is defined by the kinetic energy) Hamiltonian dynamics

$$\mathcal{H}_{\text{DM}} = Ke^{+\Phi/2K_0} - K_0 e^{-\Phi/2K_0} \equiv 0; \quad K \equiv \sum \frac{p^2}{2m}.$$

As usual, the equations of motion follow by differentiation

$$\{\dot q = \partial\mathcal{H}/\partial p = pe^{+\Phi/2K_0}; \quad \dot p = -\partial\mathcal{H}/\partial q = Fe^{-\Phi/2K_0}\}.$$

The accelerations which follow, using the identity

$$K/K_0 = e^{-\Phi/K_0},$$

are then exactly the same as those from the isokinetic equations of motion given above

$$\{\ddot{q} = (F/m) - p\sum (F\cdot\dot{q}/2K_0)e^{+\Phi/2K_0} = (F/m) - \zeta\dot{q}\};$$

$$\zeta = \sum F\cdot\dot{q}/(2K_0).$$

III. THE HOOVER-LEETE ISOKINETIC THERMOSTAT IN ITS LAGRANGIAN AND HAMILTONIAN FORMS

The Hamiltonians corresponding to the isokinetic Gauss'-Principle and Nosé-Hoover velocity-based approaches are not the only such means of thermostating equilibrium systems. A straightforward application of nonholonomic Lagrangian mechanics, as well as the familiar Hamiltonian mechanics outlined in Leete's Master's thesis,[6] lead to another type of isokinetic mechanics. These two equivalent forms of mechanics, both of which can be used to keep the *velocity*-based kinetic energy $K(\dot{q})$ constant, while allowing the momentum-based kinetic energy $K(p)$ to vary, proceed by modifying the $\{\dot{q}\}$, rather than the $\{\dot{p}\}$, equations.

To distinguish the two different kinetic energies we use the notation

$$K(\dot{q}) = \sum \frac{m\dot{q}^2}{2}; \qquad K(p) = \sum \frac{p^2}{2m}.$$

Let us begin with the Lagrangian case

$$\mathcal{L}_{\rm HL} \equiv K(\dot{q}) - \Phi + \lambda[K(\dot{q}) - K_0],$$

where $K(\dot{q})$ is the *velocity*-based kinetic energy, $\Sigma(m/2)\dot{q}^2$, and the Lagrange multiplier λ has the task of maintaining $K(\dot{q})$ at its initial value, K_0, as the motion proceeds. Lagrange's equations of motion follow from the usual textbook differentiations of the Lagrangian with respect to the velocities and coordinates:

$$\left\{ p = \frac{\partial \mathcal{L}_{\rm HL}}{\partial \dot{q}} = m\dot{q}(1+\lambda); \right.$$

$$\left. \dot{p} = m\ddot{q}(1+\lambda) + m\dot{q}\dot{\lambda} = \frac{\partial \mathcal{L}_{\rm HL}}{\partial q} \equiv F \right\}.$$

Now multiply the $\dot{p}$ equation by $\dot{q}$ and sum

$$\sum m\ddot{q}(1+\lambda)\dot{q} + \sum m\dot{q}\dot{\lambda}\dot{q} = 0 + 2K_0\dot{\lambda}$$

$$= \sum F\cdot\dot{q} \equiv -\dot{\Phi}.$$

The value of the Lagrange multiplier λ follows:

$$\lambda = \frac{(\Phi_0 - \Phi)}{2K_0} = \frac{\sqrt{4K(p)K_0} - K_0}{2K_0} = \frac{2K_0(1+\lambda) - 2K_0}{2K_0} = \lambda,$$

where the last expression, which completes the identity, follows from the Hamiltonian given just below.

With Leete's help, Hoover discovered, in 1979, that the *velocity*-based kinetic energy $K(\dot{q})$ can alternatively be kept constant by using the (constant in time) Hoover-Leete Hamiltonian

$$\mathcal{H}_{\rm HL} \equiv \sum \dot{q}p - \mathcal{L}_{\rm HL} = \sqrt{4K_0K(p)} + \Phi - K_0$$

$$= \mathcal{H}_0 = \Phi_0 + K_0,$$

where Φ is again the usual potential energy and Φ_0 is its initial value.

The constancy of the kinetic energy is easy to see. The equations of motion

$$\left\{ \dot{q} = \frac{\partial \mathcal{H}}{\partial p}; \quad \dot{p} = -\frac{\partial \mathcal{H}}{\partial q} \right\} \rightarrow \left\{ \dot{q} = \frac{p}{m}\sqrt{\frac{K_0}{K(p)}}; \quad \dot{p} = F(q) \right\},$$

imply that the velocity-based kinetic energy does not vary

$$K(\dot{q}) = \frac{m}{2}\sum \dot{q}^2 = \sum \frac{p^2}{2m}\left[\frac{K_0}{K(p)}\right] \equiv K_0.$$

At the same time, except in the equilibrium case with a large number of degrees of freedom, there is no guarantee that the momentum-based kinetic energy $K(p)$ is similar in magnitude to K_0. But, provided that $\{m\dot{q}=p\}\rightarrow K(p)=K(\dot{q})=K_0$ initially, Leete's equations of motion do match the Gaussian isokinetic ones to second order

$$\left\{ \ddot{q} = \frac{F}{m} - \zeta\dot{q} \right\}; \quad \zeta = \frac{\sum F\cdot\dot{q}}{2K_0},$$

through the second derivatives, $\{\ddot{q}\}$, but differ in the *third* derivatives, where the mean-squared constraint force in the isokinetic case is less than the Hoover-Leete analog, in accordance with Gauss' Principle of Least Constraint.[1,5]

The constraint of constant temperature in Hamiltonian mechanics contradicts the thermodynamic notion that the energy and temperature cannot be varied independently in a system of fixed composition and volume. Both E and T are *constants* of the motion, using Leete's approach. Provided that the initial conditions are wisely chosen, with E and T corresponding to the *same* thermodynamic state, this approach *can* certainly be used to determine equilibrium properties. We will see, in Sec. VIII, what the (rather strange) consequences of this thermostat are away from equilibrium.

IV. LANDAU-LIFSHITZ' CONFIGURATIONAL THERMOSTAT

A (much) more complicated Hamiltonian thermostat, conserving the force-based *configurational* temperature $T_{\rm con}$, can be based on straightforward (though quite tedious) holonomic Hamiltonian mechanics. A proper configurationally thermostated Hamiltonian conserves not only the Hamiltonian but also the configurational temperature

$$kT_{\rm con} \equiv \sum_N F_i^2 / \sum_N \nabla_i^2 \mathcal{H}.$$

Here k is Boltzmann's constant, which we set equal to unity in the numerical work. A molecular dynamics simulation based on this definition of temperature follows standard Hamiltonian mechanics, as the temperature constraint is just a (complicated) holonomic (coordinates only) constraint. The simplest procedure begins with the system Lagrangian, augmented with a Lagrange multiplier λ which constrains

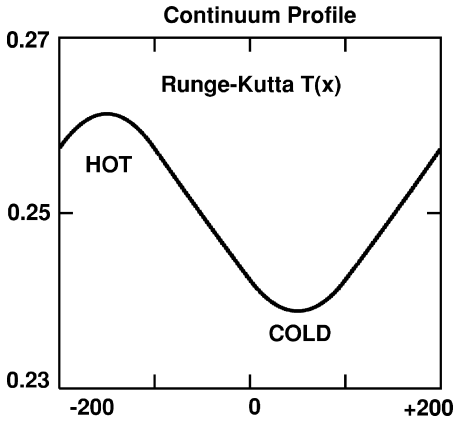

FIG. 1. Temperature profile according to Fourier's law, with mean reservoir temperatures of $0.26(-200<x<-100)$ and $0.24(0<x<100)$.

$\{T_{\rm con}, \dot{T}_{\rm con}, \ddot{T}_{\rm con}\}$. The corresponding Hamiltonian has the form

$$\mathcal{H} = \Phi(\{q\}) + K(\{p\}) + \lambda(T_{\rm con} - T_0).$$

For success, the initial conditions have to be chosen to correspond to both the desired temperature, $T_{\rm con}=T_0$, and to the condition $\dot{T}_{\rm con}=0$. Then, to begin the analytic work, differentiate the temperature equation, $T_{\rm con}(\{q\})=T_0$, twice with respect to time. The two time differentiations, using the chain rule, give first $\{\dot{q}\}$ and then $\{\ddot{q}\}$. By substituting the constrained equations of motion

$$\{m\ddot{q}_i = \dot{p}_i = F_i(\{q\}) - \lambda\nabla_i T_{\rm con}\},$$

for the $\{\ddot{q}\}$, we obtain the Lagrange multiplier λ. For further details of this calculation, see Sec. IX. The numerical work can be checked by noting that both $T=T_{\rm con}$ and $\mathcal{H}$ are constants of the motion when the calculation is error free.

V. AOKI AND KUSNEZOV'S φ^4 MODEL FOR HEAT CONDUCTION

The ϕ^4 model gets its name from the functional form of a quartic "tethering potential," $\Phi_{\rm Teth}$, which links each particle to a fixed lattice site with a cubic restoring force

$$\Phi_{\rm Teth} \equiv \sum_N \left(\frac{\kappa\delta^4}{4}\right) \rightarrow F_{\rm Teth} = -\kappa\delta^3.$$

In the pedagogical simulations which follow we will choose the tethering force constant equal to unity, $\kappa=1$. Nearest-neighbor particle pair interactions in the ϕ^4 model are governed by Hooke's Law

$$\Phi_{\rm Pair} = \sum_{\rm NN} (\mu/2)(|r| - d)^2.$$

Aoki and Kusnezov have carried out a variety of pedagogical heat flow simulations for this model in both one and three space dimensions.[17,18] The ϕ^4 model has a finite Fourier conductivity in one dimension. The numerical work carried out by Aoki and Kusnezov established the temperature dependence of the thermal conductivity, $3/T^{4/3}$.

For simplicity in the one-dimensional work which we carry out here, we always choose the nearest-neighbor separation d equal to unity. We also choose the strength of the Hooke's-Law interaction μ and the particle mass equal to unity. As was abundantly demonstrated by Aoki and Kusnezov, the combination of a site-based tethering potential with a Hooke's-Law pair potential provides the usual Fourier conductivity, with the heat flux proportional to the (sufficiently small) temperature gradient. In what follows we apply four different thermostat constraints to this potential model, using four different dynamical approaches: Nosé-Hoover, Gaussian isokinetic, Hoover-Leete isokinetic, and the Landau-Lifshitz isoconfigurational.

A numerical solution of the heat flow equation for the one-dimensional chain

$$\dot{T} = \nabla(3T^{-4/3}\nabla T) \pm \alpha T,$$

can be obtained by Runge-Kutta integration, with a rescaling of the temperatures within the reservoir regions at the end of each time step accounting for the source and sink terms $\pm\alpha T$ in the flow equation. Figure 1 shows a fully converged temperature profile obtained in this way, using hot and cold temperatures of 0.26 and 0.24, with 400 mesh points. The spatial gradient operations were replaced by finite-difference approximations

$$(\nabla T)_i = (T_{i+1} - T_{i-1})/2,$$

with appropriate subscript changes accounting for the periodic boundary conditions.

VI. RESULTS WITH THE NOSÉ-HOOVER THERMOSTAT

With a periodic system composed of four parts, "Hot," "Newton$_1$," "Cold," and "Newton$_2$," each part containing $N/4$ particles, the equations of motion are

$$\{m\ddot{q} = F - \zeta_{\rm Hot} m\dot{q}\}; \quad \dot{\zeta}_{\rm Hot} = [(K/K_{\rm Hot}) - 1]/\tau^2,$$

$$\{m\ddot{q} = F - \zeta_{\rm Cold} m\dot{q}\}; \quad \dot{\zeta}_{\rm Cold} = [(K/K_{\rm Cold}) - 1]/\tau^2,$$

in the hot and cold regions, and

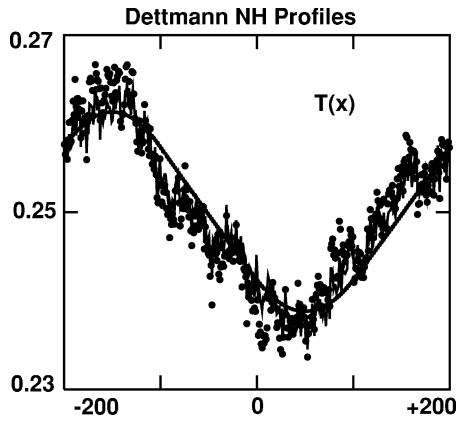

FIG. 2. Temperature profile for a conventional two-temperature Nosé-Hoover simulation, as described in the text. Here, and in the following figures, the kinetic temperature is indicated by a (quite wiggly) line, while the configurational temperature is indicated by filled circles.

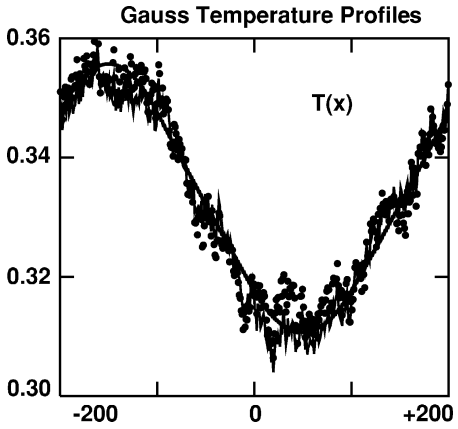

FIG. 3. Temperature profile for a conventional two-temperature Gaussian isokinetic simulation, as described in the text. The kinetic temperature corresponds to the wiggly line; the configurational temperature to the filled circles.

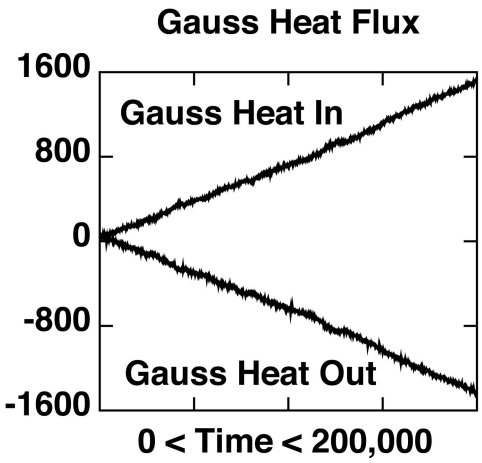

FIG. 4. Summed-up values of the heat transferred to the hot and cold Gaussian isokinetic reservoirs for the simulation of Fig. 3.

$$\{m\ddot{q} = F\},$$

in the two Newtonian regions. The ϕ^4 force F_i depends upon the three coordinates $\{x_{i-1}, x_i, x_{i+1}\}$ and also includes the tethering force, $F_{\text{Teth}} = -\kappa(x_i - x_0)^3$, where x_0 is the lattice site for the ith particle in the perfect lattice. The initial velocities and displacements were selected randomly within ranges

$$-\Delta_v < \{v\} < +\Delta_v; \quad -\Delta_q < \{\delta q\} < +\Delta_q,$$

with Δ_v or Δ_q chosen to reproduce the desired stationary reservoir temperatures, 0.26 and 0.24. The *average* temperatures, both kinetic and configurational, from the last half of a 400-particle, 40-million time step simulation,

$$0 < t < 100\,000 \text{ (equilibration)};$$

$$100\,000 < t < 200\,000 \text{(averaging)},$$

are shown in Fig. 2, along with a comparison solution of the macroscopic heat flow equation. The agreement is quite good, as would be expected from Aoki and Kusnezov's work.

VII. RESULTS WITH THE GAUSSIAN ISOKINETIC THERMOSTAT

Simulations with two *Gaussian isokinetic thermostats*, with the same time step, $dt = 0.005$, and the same equilibration and averaging intervals, produced similar results, maintaining the initial kinetic temperatures throughout. The averaged profiles, along with a corresponding continuum solution, are shown in Fig. 3. Again the agreement, for both the kinetic and the configurational temperatures, is perfectly satisfactory.

The instantaneous (at time t) values of the total heat added (from time 0 to time t) to the "hot" reservoir as well as that removed from the "cold" reservoir are shown in Fig. 4. The sum of the two necessarily oscillates about zero as t increases. Straight lines drawn through the data provide an estimate for the Fourier conductivity in agreement with the results of Aoki and Kusnezov's work. The two conventional thermostat types, Nosé-Hoover and Gaussian isokinetic, show that either method can be used to simulate the simple two-temperature heat flow problem.

VIII. RESULTS WITH THE HOOVER-LEETE HAMILTONIAN ISOKINETIC THERMOSTAT

Results with the Hamiltonian isokinetic thermostats

$$\left\{ \dot{q}_{\text{Hot/Cold}} = \frac{p}{m}\sqrt{\frac{K_{\text{Hot/Cold}}}{K(p)}}; \quad \dot{p} = F(q) \right\},$$

where the fixed hot and cold kinetic temperatures $\langle m\dot{q}^2/k\rangle_{\text{Hot/Cold}}$ are fixed by the initial conditions, but come to differ from the unconstrained momentum-based temperatures $\langle p^2/mk\rangle_{\text{Hot/Cold}}$. Unlike the Nosé-Hoover and Gaussian isokinetic profiles, the Hamiltonian-based profiles show *no temperature gradients at all* (see Fig. 5). Instead there are discontinuities between the fixed kinetic temperatures and the adjacent constant values of the unconstrained Newtonian regions. In the latter regions the kinetic and configurational temperatures equilibrate, and match the unconstrained configurational temperature of the thermostated regions. In the

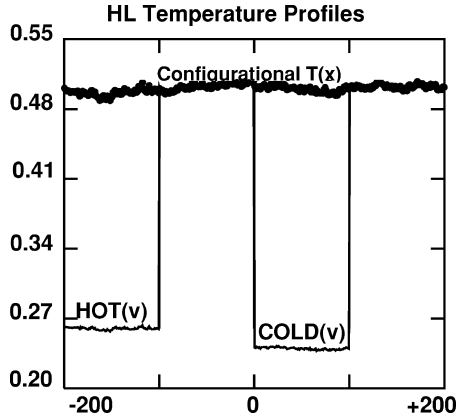

FIG. 5. Temperature profiles (kinetic and configurational) for a simulation using constrained Hoover-Leete isokinetic reservoirs. The time step is $dt = 0.005$, with a total elapsed time of 10^6 divided equally between equilibration and averaging portions. The kinetic temperature corresponds to the wiggly line; the configurational temperature to the filled circles.

thermostated regions there is no such equilibration.

Evidently there are no heat fluxes in the system either. By constraining not only the energy (through the constant Hamiltonian driving the motion) but also the kinetic temperatures, the system is evidently prevented from supporting heat flow. The Hamiltonian nature of the motion equations also prevents the formation of the multifractal phase-space distributions associated with nonequilibrium stationary states.[1,2] A more detailed analysis of the reasons for the failure of this Hamiltonian approach could perhaps be based on a detailed analysis of the microscopic equations describing heat flux.[1]

IX. RESULTS WITH THE LANDAU-LIFSHITZ HAMILTONIAN ISOCONFIGURATIONAL THERMOSTAT

For the special ϕ^4 model considered here, with $d=1$, the force F_i is

$$F_i = -\kappa\delta_i^3 + \mu[\delta_{i+1} - 2\delta_i + \delta_{1-1}].$$

The *pair-potential* contribution to $\nabla_i^2\mathcal{H}$ is 2μ, for the two nearest-neighbor interactions

$$\frac{\partial^2}{\partial x_i^2}(\mu/2)(|x_i - x_{i\pm1}| - 1)^2 = \mu.$$

The *tethering* potential contribution is $3\kappa\delta_i^2 = \nabla_i^2(\kappa\delta_i^4/4)$

$$\sum_N \nabla^2\mathcal{H} = 2N\mu + \sum_N 3\kappa\delta^2.$$

The time derivative of the configurational temperature is a quotient of products of single-particle sums

$$\dot{T} = \frac{2\sum F\dot{F}}{\sum(2\mu + 3\kappa\delta^2)} - \frac{6\sum F^2 \sum \kappa v\delta}{\left[\sum(2\mu + 3\kappa\delta^2)\right]^2},$$

where

$$\dot{F}_i = \mu[v_{i+1} - 2v_i + v_{i-1}] - 3\kappa\delta_i^2 v_i.$$

Another time differentiation gives the second derivative

$$\ddot{T}_{\text{Newton}} = \frac{2\sum F\ddot{F} + 2\sum \dot{F}^2}{\sum(2\mu + 3\kappa\delta^2)} + \frac{72\sum F^2\left[\sum \kappa v\delta\right]^2}{\left[\sum(2\mu + 3\kappa\delta^2)\right]^3} - \frac{24\sum F\dot{F}\sum \kappa v\delta + 6\sum F^2\left(\kappa\sum[a\delta + \sum v^2]\right)}{\left[\sum(2\mu + 3\kappa\delta^2)\right]^2},$$

where

$$\ddot{F}_i = \mu[a_{i+1} - 2a_i + a_{i-1}] - \kappa[3\delta_i^2 a_i + 6\delta_i v_i^2].$$

The *second* derivative, $\ddot{T}$, introduces the Lagrange multiplier through the equations of motion

$$\{a_i = \dot{v}_i = F_i - \lambda\nabla_i T\}.$$

Though complicated, the resulting equation for λ is linear, and gives an explicit expression for the Lagrange multiplier that stabilizes T

$$\lambda = \frac{\ddot{T}_{\text{Newton}}}{\sum \nabla_i T\left([2\mu(F_{i+1} - 2F_i + F_{i-1}) - 6\kappa\delta_i^2 F_i] - \left[\sum F^2 \sum 6\kappa F_i\delta_i\right]\right)}.$$

Because T cannot change, both $T=T_0$ and $\dot{T}=0$ need to be properly specified in the initial conditions. A convenient way to do this is to choose the velocities equal to zero so that $\dot{T} \equiv 0$.

A typical profile using two configurational thermostats is shown in Fig. 6. Just as in the Hoover-Leete isokinetic case there is no equilibration of the constrained temperature (here configurational). The unconstrained kinetic temperature equilibrates throughout the system to a value dependent upon the initial conditions. Evidently the Landau-Lifshitz constrained configurational thermostat is quite useless for nonequilibrium work.

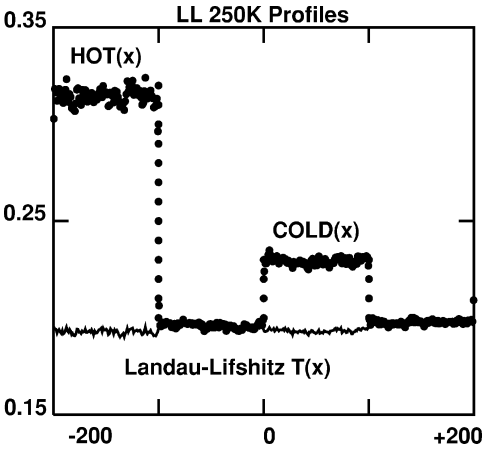

FIG. 6. Temperature profiles (kinetic and configurational) for a simulation using constrained Landau-Lifshitz isoconfigurational reservoirs. The time step is $dt=0.002$, with a total elapsed time of 500 000 divided equally between equilibration and averaging portions. The kinetic temperature corresponds to the wiggly line; the configurational temperature to the filled circles.

X. CONCLUSIONS

The results obtained with the Gaussian isokinetic and Nose-Hoover thermostats are quite consistent with the past history of their use over the last quarter century.[1,2,6,7] For these familiar thermostats there was no problem in reaching

a nonequilibrium steady state with temperature profiles and heat fluxes very similar to those expected from macroscopic hydrodynamics using Fourier's law. Although the simple ϕ^4 model may strike the reader as rather special we believe that the results obtained using it are *typical* of realistic applications. The lack of momentum conservation in this model, due to the tethering potential, though it certainly eliminates the low-frequency divergence of the one-dimensional heat conductivity, actually makes the one- and two-dimensional models more closely resemble their *more physical* three-dimensional cousins. We expect that the configurational analog[14] of the Nosé-Hoover kinetic thermostat, discussed in Sec. II, would provide very similar results. It would be particularly interesting to explore the differences between the tensor forms of the kinetic and configurational temperatures, by applying these ideas to strong shockwaves.

On the other hand, the results obtained with *purely Hamiltonian* mechanics, using either the Hoover-Leete constrained isokinetic or the Landau-Lifshitz constrained configurational thermostats were both unexpected and thought-provoking. The main message of the current work is that unadulterated Hamiltonian mechanics is simply unsuitable to steady-state nonequilibrium simulations. The longtime results, using Hamiltonian mechanics, are tied to both the energy and the phase volume of the initial conditions, in a way simply inconsistent with the known phase-space contraction typical of nonequilibrium systems. From the æsthetic standpoint, we believe that the canonical-ensemble derivations of Landau and Lifshitz[12] and Travis and Braga[14] are not only clearer, but also more useful and stimulating than is Rugh's alternative microcanonical approach.[13]

Both the Hamiltonian approaches stably constrained their target temperatures in the two reservoir regions. Nevertheless temperature differences led to no net heat flow and the computed temperature profiles bore no resemblance to the predictions of hydrodynamics. The kinetic and configurational temperatures, though *unequal* in the constrained reservoirs

$$T_{\mathrm{kin}} \neq T_{\mathrm{con}}\ [\mathrm{reservoirs}],$$

were equal (to yet a third nearly constant temperature) in the unconstrained bulk Newtonian regions

$$T_{\mathrm{kin}} = T_{\mathrm{con}}\ [\mathrm{bulk}].$$

These results emphasize the fact that *nonHamiltonian* dynamics is absolutely necessary to the realistic modeling of nonequilibrium systems. A direct proof/understanding of the failure of the Hamiltonian systems to show a nonvanishing heat flux would be most welcome.

ACKNOWLEDGMENTS

This work was first presented at a Mexico City workshop organized by Francisco Uribe at Universidad Autónoma Metropolitana-Iztapalapa in January, 2007. Subsequently, a shorter version was presented at the Schrödinger Institute workshop, in Wien, 18–20 April 2007, honoring the 65th birthday of Herr Professor Doktor Harald Posch. We are especially grateful to Professors Uribe, Posch, and Dellago for making this work possible. Brad Holian and Janka Petravic kindly commented on an earlier version of this paper. Karl Travis provided us with copies of Refs. 14–16.

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