Dynamical gauge invariance of statistical mechanics

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We investigate gauge invariance against phase space shifting in nonequilibrium systems, as represented by time-dependent many-body Hamiltonians that drive an initial ensemble out of thermal equilibrium. The theory gives rise to gauge correlation functions that characterize spatial and temporal inhomogeneity with microscopic resolution on the one-body level. Analyzing the dynamical gauge invariance allows one to identify a specific localized shift gauge current as a fundamental nonequilibrium observable that characterizes particle-based dynamics. When averaged over the nonequilibrium ensemble, the shift current vanishes identically, which constitutes an exact nonequilibrium conservation law that generalizes the Yvon-Born-Green equilibrium balance of the vanishing sum of ideal, interparticle, and external forces. Any given observable is associated with a corresponding dynamical hyperforce density and hypercurrent correlation function. An exact nonequilibrium sum rule interrelates these one-body functions, in generalization of the recent hyperforce balance for equilibrium systems. We demonstrate the physical consequences of the dynamical gauge invariance using both harmonically confined ideal gas setups, for which we present analytical solutions, and molecular dynamics simulations of interacting systems, for which we demonstrate the shift current and hypercurrent correlation functions to be accessible both via finite-difference methods and via trajectory-based automatic differentiation. We show that the theory constitutes a starting point for developing nonequilibrium reduced-variance sampling algorithms and for investigating thermally-activated barrier crossing.

I. INTRODUCTION

Gauge invariance is one of the arguably most powerful construction principles of theoretical physics, with profound consequences that range from classical electrodynamics to the standard model of particle physics and beyond. The gauge freedom in a given theory is intimately linked to the validity of exact relationships, which typically possess the form of conservation laws, such as those for electrical and more general charges in the above fundamental theories. The link between the gauge freedom and the associated conservation laws is provided by Noether's theorem of invariant variations [1–4]. In a typical setting, one applies the theorem to an action functional, which serves the dual purpose of generating the dynamics from an associated variational principle and providing the conservation laws via the gauge freedom.

For the deterministic dynamics of particle-based systems a range of variational approaches exists, perhaps most notably in the form of Hamilton's principle of stationary action [5]. A statistical mechanical description of the dynamics is then typically based on the Liouville equation for the time evolution of the many-body probability distribution on phase space [6, 7]. While direct numerical solution of the Liouville equation for realistic systems comes at prohibitive cost, trajectory-based molecular dynamics simulations provide a powerful alternative [7–9]. Corresponding theoretical developments in nonequilibrium statistical mechanics [6, 7] are based on a range of techniques and coarse-graining strategies [10], including mode-coupling theory of the glass transition [11, 12], stochastic thermodynamics and its associated fluctuation theorems [13], as well as dynamical density [14–17] and power functional theory [18–21].

The dynamical behaviour and nonequilibrium phenomena that occur in soft matter systems encompass a broad spectrum of physical systems and effects [7, 22]. Representative topical examples include the emergence of solitons under overdamped Brownian dynamics [23], the physics of dynamical exclusion processes [24], the dynamics of confined electrolytes [25], the non-trivial properties of the electrical noise at the nanoscale [26, 27], the aging of glasses [28], and features of memory in generalised Langevin dynamics [29].

Owing to the significant increase of the degree of complexity of the nonequilibrium problem over the equilibrium physics, the body of available *exact* dynamical results is arguably less well developed than in equilibrium, where a breadth of statistical mechanical sum rules is available [7, 14, 30–34]. Notable fundamental relations that are relevant in nonequilibrium include the Yvon theorem [7, 11, 35], as is based on partial integration on phase space, and Hirschfelder's hypervirial theorem [36] which generalizes the standard virial theorm [7] to arbitrary observables. Furthermore, stochastic thermodynamics provides a significant body of fluctuation theorems and further results [13].

Noether's theorem [1–4] has only recently been applied to statistical physics as a systematic construction principle within a variety of theoretical approaches [37–46]. In particular, based on a specific 'shifting' operation on phase space, a broad range of equilibrium statistical mechanical sum rules could both be reproduced and extended [47–56]. The underlying localized shifting operation on phase space was identified subsequently as a gauge transformation of equilibrium statistical mechanics [55, 56]; see Refs. [57, 58] for recent popular accounts. In equilibrium, making use of the nontrivial Lie algebra

structure gives a promising perspective on the future development of novel equilibrium sampling methods and exact sum rule construction [55, 56].

Here we investigate the consequences of the statistical mechanical gauge invariance for the dynamics of particle-based systems in general *nonequilibrium* setups, as described by the Liouville equation. We introduce dynamical versions of the static differential shifting operators [55, 56]. These allow one, upon systematic analysis of the effects of dynamic shifting on the temporal propagation of dynamical averages, to derive and to validate very general exact nonequilibrium sum rules. These identities interrelate specific gauge correlation functions that possess microscopically sharp dependence on position and on time. In particular the shift current, the dynamical hyperforce density, and the hypercurrent correlation function emerge naturally in the theory and they are interrelated by exact nonequilibrium sum rules. The limit of equilibrium dynamics constitutes a special, yet nontrivial case, as we demonstrate. A central mechanism of the framework is a specific differential operation induced by the time evolution of the initial equilibrium ensemble, see Fig. 1 for an illustration.

As a simple yet useful and analytically tractable toy model, we apply the dynamical gauge framework to the harmonically confined ideal gas where switching at an initial time generates a nonequilibrium situation. Analyzing the gauge correlation functions provides deep insight into the motion of the ensemble. Even when reduced to equilibrium, the theory yields nontrivial insight into the thermal dynamics. To address the behaviour of systems of mutually interacting particles, we demonstrate that results for the relevant shift and hypercurrent correlation functions are accessible in molecular dynamics.

The access to the gauge correlation functions in molecular simulations is provided by implementing differentiation with respect to the initial state. Automatic differentiation [59] is a natural choice for realizing this operation, and it has gained much recent popularity, as it allows one to access with great ease derivatives that are otherwise out of practical reach of symbolic differentiation, whether via pencil and paper or algorithmically assisted. The method does not suffer from the drawbacks of numerical finite-difference schemes and it has been used to great effect in molecular dynamics simulations in recent work addressing design and optimal control tasks [60–63], as well as in the implementation of functional calculus based on analytical [64, 65] and machine-learned neural density functionals [65–76]. Here we exploit that the dynamical gauge invariance is inherently linked to a specific initial state derivative of the nonequilibrium dynamics. We also demonstrate the alternative accessibility of all gauge correlation functions via finite-difference differentiation, which works universally with no need for specialized compute environment.

The paper is organized as follows. In Sec. II we describe the particle-based classical statistical mechanics, including the setup for the Hamiltonian in Sec. II A and

the Liouvillian formulation of the dynamics in Sec. II B. We lay out the microscopically resolved one-body equation of motion in Sec. II C and describe the standard realization of the dynamics via many-body trajectories in Sec. II D.

The dynamical gauge invariance theory is developed in Sec. III, starting with an elementary derivation of an exact dynamical shift current sum rule in Sec. III A. We then address the deeper mechanism underlying this derivation by formalizing the phase space shifting on the basis of Poisson brackets in Sec. III B. The Poisson bracket formulation is then used to generalize to dynamical phase space shifting and thus to formulate dynamical gauge invariance in Sec. III C, which allows one to derive exact hypercurrent sum rules that apply to general observables under nonequilibrium dynamics, as described in Sec. III D. The connection to a specific initial-state time derivative is presented in Sec. III E. The relevance for trajectory-based simulations is described in Sec. III F. We illustrate the general theory by making specific choices for relevant hyperobservables in Sec. III G.

We present applications of the general gauge theory in Sec. IV. We first demonstrate that in equilibrium the formalism recovers correctly the static hyperforce theory [54–56], while providing additional nontrivial temporal insight, as presented in Sec. IV A. As an initial concrete toy model that permits analytical solution, we consider harmonically confined noninteracting particles in Sec. IV B. To address mutually interacting systems, we turn to simulations and use molecular dynamics together with initial state differentiation implemented via automatic or finite-difference derivatives to access the shift current and hypercurrent correlation functions in Sec. IV C. The framework allows one to address the construction of nonequilibrium reduced-variance estimators and to shed new light on the classical barrier crossing problem. We give conclusions in Sec. V.

II. STATISTICAL MECHANICS

A. Microscopic many-body model

We consider classical systems of N particles with position coordinates $\mathbf{r}_1, \ldots, \mathbf{r}_N \equiv \mathbf{r}^N$ and momentum variables $\mathbf{p}_1, \ldots, \mathbf{p}_N \equiv \mathbf{p}^N$, where $\mathbf{r}^N$ and $\mathbf{p}^N$ are shorthand notations. The Hamiltonian is taken to possess the standard form

$$H = \sum_i \frac{\mathbf{p}_i^2}{2m} + u(\mathbf{r}^N) + \sum_i V_{\text{ext}}(\mathbf{r}_i), \qquad (1)$$

where the summation index $i = 1, \ldots, N$ runs over all N particles, m denotes the particle mass, $u(\mathbf{r}^N)$ is the interparticle interaction potential, and $V_{\text{ext}}(\mathbf{r})$ is an external potential, here expressed as a function of the generic position variable $\mathbf{r}$. The initial state at times $t < 0$ is characterized by a stationary Hamiltonian H_0, where the

FIG. 1. Illustration of the temporal structure of the nonequilibrium gauge theory. The initial state is in thermal equilibrium at inverse temperature β up to time 0, with the dynamics being governed by the stationary Liouvillian L_0 (orange vertical arrow). The time-dependent Liouvillian $L(t)$ then creates nonequilibrium dynamics (magenta horizontal arrows) at times $t \geq 0$.

mass m_0, interparticle potential $u_0(\mathbf{r}^N)$, and external potential $V_{\text{ext},0}(\mathbf{r})$ are all explicitly time-independent, as is indicated by the subscript 0. Per construction the initial system possesses a well-defined thermal equilibrium, as described below. For times $t \geq 0$, general explicit time-dependence in Eq. (1) can occur. This includes temporal variation of the mass m, of the interparticle interaction potential $u(\mathbf{r}^N)$, and of the external potential $V_{\text{ext}}(\mathbf{r})$; here and throughout we suppress mere *parametric* dependence on time t in the notation and reserve all time arguments to denote *dynamical* dependence, as it is induced by the particle motion.

The switching at $t = 0$ need not be smooth and hence a discontinuity may occur such that in general we allow for: $\lim_{t \to 0^+} H \neq H_0$. That the initial state Hamiltonian H_0 and the nonequilibrium time evolution generator H can in general differ from each other allows for flexible modelling of a broad range of situations. Thereby the initial many-body phase space probability distribution $f_0(\mathbf{r}^N, \mathbf{p}^N)$ is taken to be in thermal equilibrium, as characterized by the canonical ensemble for the given form of H_0. Hence as a function of the phase space point $\mathbf{r}^N, \mathbf{p}^N$ we have:

$$f_0(\mathbf{r}^N, \mathbf{p}^N) = \frac{e^{-\beta H_0}}{Z_0}, \qquad (2)$$

where $\beta = 1/(k_B T)$, with Boltzmann constant k_B, absolute temperature T of the initial state, and the (scaled) canonical partition sum $Z_0 = \text{Tr}\, e^{-\beta H_0}$. The normalization is such that $\text{Tr}\, f_0 = 1$, where the canonical 'trace' is the phase space integral over all degrees of freedom: $\text{Tr} \cdot = \int d\mathbf{r}_1 \ldots d\mathbf{r}_N d\mathbf{p}_1 \ldots d\mathbf{p}_N \cdot$. The partition sum in standard form is then $Z_0/(h^{dN} N!)$ with Planck constant h and dimensionality d.

As a special case, the present dynamical setup includes the switching at time $t = 0$ from an initial Hamiltonian H_0 to a different Hamiltonian $H \neq H_0$, where H however is also taken to be stationary. Then the system characterized by H is driven out of *its* associated equilibrium already at the initial time, which forms a common situation, e.g. when applying linear response theory [6].

B. Phase space time evolution and dynamical averages

The time evolution of the dynamical probability distribution function $f(\mathbf{r}^N, \mathbf{p}^N, t)$ is governed by the Liouville equation $\partial f(t)/\partial t = -Lf(t)$, where we have left away the dependence on the phase space variables $\mathbf{r}^N, \mathbf{p}^N$ in the notation. The Liouvillian L is the phase space differential operator given via Poisson brackets by:

$$L = \{\,\cdot\,, H\}. \qquad (3)$$

The Poisson bracket $\{\,\cdot\,,\,\cdot\,\}$ of two general phase space functions g and h is thereby defined [5] as:

$$\{g, h\} = \sum_i \left(\frac{\partial g}{\partial \mathbf{r}_i} \cdot \frac{\partial h}{\partial \mathbf{p}_i} - \frac{\partial g}{\partial \mathbf{p}_i} \cdot \frac{\partial h}{\partial \mathbf{r}_i} \right). \qquad (4)$$

In our chosen setup, the time evolution for $t < 0$ is governed by the stationary Liouvillian $L_0 = \{\,\cdot\,, H_0\}$, where we recall that H_0 carries no explicit time dependence and that in general $H_0 \neq H(0^+)$. At later times $t \geq 0$ any potentially occurring parametric dependence of H on time t is passed on to L in Eq. (3); as noted above we suppress the parametric dependence on time throughout in the notation, such that time arguments denote exclusively the dynamical dependence.

Due to the specific additive structure of the Hamiltonian (1), the Liouvillian L consists correspondingly of a sum of ideal (kinetic), interparticle, and external contributions according to

$$L = L_{\text{id}} + L_{\text{int}} + L_{\text{ext}}. \qquad (5)$$

Inserting the general form of the Hamiltonian (1) into the generic Poisson bracket form of the Liouvillian (3) gives explicit expressions for the three contributions in the Liouvillian splitting (5). Thereby the ideal part is $L_{\text{id}} = \sum_i (\mathbf{p}_i/m) \cdot \nabla_i$ as is generated by the kinetic energy in Eq. (1), the interparticle part is $L_{\text{int}} = -\sum_i [\nabla_i u(\mathbf{r}^N)] \cdot \nabla_{\mathbf{p}_i}$, and the external contribution is $L_{\text{ext}} = -\sum_i [\nabla_i V_{\text{ext}}(\mathbf{r}_i)] \cdot \nabla_{\mathbf{p}_i}$, and the latter two forms arise, respectively, from the interparticle potential $u(\mathbf{r}^N)$ and external potential energy $\sum_i V_{\text{ext}}(\mathbf{r}_i)$ in Eq. (1). Forces are generated from position gradients, where $\nabla_i = \partial/\partial \mathbf{r}_i$ denotes the derivative with respect to position $\mathbf{r}_i$ and $\nabla_{\mathbf{p}_i} = \partial/\partial \mathbf{p}_i$ is the partial derivative with respect to the momentum of particle i.

The Liouvillian splitting (5) into its constituent parts follows a standard scheme. A prominent example is the derivation of the celebrated velocity Verlet algorithm [8, 77], starting from the Trotter expansion of the propagator [7], which we briefly relate to. In the notation of Ref. [7], the Liouvillian is decomposed as $L = \mathrm{i}\mathcal{L}_\mathbf{r} + \mathrm{i}\mathcal{L}_\mathbf{p}$, with the position contribution $\mathrm{i}\mathcal{L}_\mathbf{r} = L_\mathrm{id}$ and the momentum part $\mathrm{i}\mathcal{L}_\mathbf{p} = L_\mathrm{int} + L_\mathrm{ext}$, where by convention the imaginary unit i is included [10]. This further splitting of $\mathrm{i}\mathcal{L}_\mathbf{p}$ into interparticle (L_int) and external contributions (L_ext) follows naturally from the additive form of the Hamiltonian (1).

Formally, the many-body probability distribution $f(t)$ that solves the Liouville equation can be expressed as

$$f(t) = \mathcal{U}(t,0)f_0, \qquad (6)$$

where $\mathcal{U}(t,0)$ denotes the propagator that performs the time evolution from time 0 to time t. The propagator can be expressed as $\mathcal{U}(t,0) = \mathrm{e}_+^{\int_0^t dt'\, L'}$, where e_+ denotes the positively time-ordered exponential [78] and L' denotes the Liouvillian (3), which is *parametrically* evaluated at time t'. Per construction $\mathcal{U}(0,0) = 1$ such that $f(0) = \mathcal{U}(0,0)f_0$. Working with the propagator $\mathcal{U}(t,0)$ is a powerful formal method that we rely on in the following. The equivalent and arguably more intuitive trajectory-based picture of the Hamiltonian dynamics is laid out below in Sec. II D.

Any general phase space function $\hat{A}(\mathbf{r}^N, \mathbf{p}^N)$ that acts as an observable of interest acquires dynamical time dependence, as generated from the motion in the system, via

$$\hat{A}(t) = \mathcal{U}^\dagger(t,0)\hat{A}, \qquad (7)$$

where the dagger indicates the adjoint; here the adjoint of an operator $\mathcal{O}$ is defined in the standard way as: $\mathrm{Tr}\, g\mathcal{O}h = \mathrm{Tr}\, h\mathcal{O}^\dagger g$, where we recall g and h being two phase space functions and Tr indicating the full phase space integral. The propagator $\mathcal{U}(t,0)$ and its adjoint $\mathcal{U}^\dagger(t,0)$ are inverse of each other, such that $\mathcal{U}(t,0)\mathcal{U}^\dagger(t,0) = 1$ and $\mathcal{U}^\dagger(t,0)\mathcal{U}(t,0) = 1$, as is characteristic of unitary time evolution.

We have left away the phase space arguments for brevity of notation. More explicitly, the variable $\hat{A}(\mathbf{r}^N, \mathbf{p}^N)$ on the right hand side of Eq. (7) represents both the observable itself as well as its value at the initial microstate $\mathbf{r}^N, \mathbf{p}^N$ [6]. Using a quantum analogy, $\hat{A}$ is a Schrödinger observable. Then the corresponding dynamical Heisenberg observable $\hat{A}(\mathbf{r}^N, \mathbf{p}^N, t)$ is given via Eq. (7). In the present classical context $\hat{A}(\mathbf{r}^N, \mathbf{p}^N, t)$ constitutes the value that the observable attains at time t, given the specific trajectory that started at $\mathbf{r}^N, \mathbf{p}^N$ at the initial time [6], as is laid out further in Sec. II D. Hence $\hat{A}(t) = \hat{A}(\mathbf{r}^N, \mathbf{p}^N, t)$ remains dependent on the initial state, which will be important in the development of the dynamical gauge theory in Sec. III.

The Liouville equation for general Heisenberg observables is $\partial\hat{A}(t)/\partial t = L(t)\hat{A}(t)$, where the time-evolved Liouvillian $L(t)$ is obtained from L, as defined via Eq. (3), by:

$$L(t) = \mathcal{U}^\dagger(t,0)L\mathcal{U}(t,0). \qquad (8)$$

An alternative form to Eq. (8) is obtained by using Poisson brackets: $L(t) = \{\,\cdot\,, H(t)\}$, where the temporally evolved form of the Hamiltonian is obtained according to the standard form of a Heisenberg observable as $H(t) = \mathcal{U}^\dagger(t,0)H$, with all parametric time dependences being evaluated at time t and suppressed in the notation. Time-resolved averages are then obtained as $A(t) = \langle\hat{A}(t)\rangle = \mathrm{Tr}\, f_0\hat{A}(t)$, which can equivalently be written as $A(t) = \mathrm{Tr}\, f(t)\hat{A}$.

The present formalism for the dynamics on the full many-body level requires one to choose a coarse-graining strategy to proceed in formulating a statistical mechanical description.

C. One-body level of correlation functions

We work with microscopic resolution, using the classical operators (phase space functions) for the one-body density, $\hat{\rho}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)$, and for the one-body current,

$$\hat{\mathbf{J}}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\frac{\mathbf{p}_i}{m}, \qquad (9)$$

where $\delta(\cdot)$ denotes the Dirac distribution in d dimensions. We first lay out the standard derivation of the dynamical one-body force density balance, which constitutes Newtons' second law in the present context [18]. The Heisenberg observable for the current (momentum density) is obtained as $m\hat{\mathbf{J}}(\mathbf{r}, t) = \mathcal{U}^\dagger(t,0)m\hat{\mathbf{J}}(\mathbf{r})$ and it satisfies the Liouville equation of motion $\partial m\hat{\mathbf{J}}(\mathbf{r}, t)/\partial t = L(t)m\hat{\mathbf{J}}(\mathbf{r}, t)$. Explicit calculation of the latter right hand side yields the Heisenberg force density observable:

$$\hat{\mathbf{F}}(\mathbf{r}, t) = L(t)m\hat{\mathbf{J}}(\mathbf{r}, t). \qquad (10)$$

where $\hat{\mathbf{F}}(\mathbf{r}, t) = \mathcal{U}^\dagger(t,0)\hat{\mathbf{F}}(\mathbf{r})$. Using the splitting (5) of the Liouvillian, the force density $\hat{\mathbf{F}}(\mathbf{r})$ can be decomposed correspondingly into ideal (or kinetic), interparticle, and external parts: $\hat{\mathbf{F}}(\mathbf{r}) = \nabla\cdot\hat{\boldsymbol{\tau}}(\mathbf{r}) + \hat{\mathbf{F}}_\mathrm{int}(\mathbf{r}) - \hat{\rho}(\mathbf{r})\nabla V_\mathrm{ext}(\mathbf{r})$, with the kinetic stress operator $\hat{\boldsymbol{\tau}}(\mathbf{r}) = -\sum_i \delta(\mathbf{r} - \mathbf{r}_i)\mathbf{p}_i\mathbf{p}_i/m$, the interparticle force density operator $\hat{\mathbf{F}}_\mathrm{int}(\mathbf{r}) = -\sum_i \delta(\mathbf{r} - \mathbf{r}_i)\nabla_i u(\mathbf{r}^N)$, and the external force field $-\nabla V_\mathrm{ext}(\mathbf{r})$ and we recall that $\hat{\rho}(\mathbf{r})$ denotes the microsopically-resolved density operator defined inline above Eq. (9). As before, any parametric dependence of $m, u(\mathbf{r}^N)$, and $V_\mathrm{ext}(\mathbf{r})$ on time t remains suppressed here and throughout in the notation.

We address the dynamics by considering $\partial m\hat{\mathbf{J}}(\mathbf{r}, t)/\partial t = L(t)m\hat{\mathbf{J}}(\mathbf{r}, t) = \mathcal{U}^\dagger(t,0)Lm\hat{\mathbf{J}}(\mathbf{r}) = \mathcal{U}^\dagger(t,0)\hat{\mathbf{F}}(\mathbf{r})$, where we have first written out $L(t)$ via Eq. (8), then used the identity $\mathcal{U}(t,0)\mathcal{U}^\dagger(t,0) = 1$ and

have identified $\hat{\mathbf{F}}(\mathbf{r}) = Lm\hat{\mathbf{J}}(\mathbf{r})$. Averaging over the initial distribution f_0 and using the decomposition (5) then yields the one-body equation of motion [18]:

$$\mathbf{F}_{\text{id}}(\mathbf{r}, t) + \mathbf{F}_{\text{int}}(\mathbf{r}, t) + \mathbf{F}_{\text{ext}}(\mathbf{r}, t) = \frac{\partial m\mathbf{J}(\mathbf{r}, t)}{\partial t}, \quad (11)$$

where the ideal (kinetic) force density is $\mathbf{F}_{\text{id}}(\mathbf{r}, t) = \nabla \cdot \langle \hat{\boldsymbol{\tau}}(\mathbf{r}, t) \rangle$, the interparticle force density is $\mathbf{F}_{\text{int}}(\mathbf{r}, t) = \langle \hat{\mathbf{F}}_{\text{int}}(\mathbf{r}, t) \rangle$, and the external force density is $\mathbf{F}_{\text{ext}}(\mathbf{r}, t) = -\rho(\mathbf{r}, t)\nabla V_{\text{ext}}(\mathbf{r}, t)$. Here the dynamical density profile is $\rho(\mathbf{r}, t) = \langle \hat{\rho}(\mathbf{r}, t) \rangle$ and the mean one-body current on the right hand side of Eq. (11) is $\mathbf{J}(\mathbf{r}, t) = \langle \hat{\mathbf{J}}(\mathbf{r}, t) \rangle$ with the Heisenberg current observable given below Eq. (9). We recall that averages are built in the standard way as $\langle \cdot \rangle = \text{Tr} f_0 \cdot$, see Sec. II B.

Turning to the initial system, which is in thermal equilibrium according to f_0 given by Eq. (2), the general dynamical force density balance (11) reduces to the static force density relationship $\mathbf{F}_0(\mathbf{r}) = 0$, which can be decomposed as

$$\mathbf{F}_{\text{id},0}(\mathbf{r}) + \mathbf{F}_{\text{int},0}(\mathbf{r}) + \mathbf{F}_{\text{ext},0}(\mathbf{r}) = 0, \quad (12)$$

where explicitly the three terms are: $\mathbf{F}_{\text{id},0}(\mathbf{r}) = \nabla \cdot \text{Tr} f_0\hat{\boldsymbol{\tau}}(\mathbf{r})$, $\mathbf{F}_{\text{int},0}(\mathbf{r}) = -\text{Tr} f_0 \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\nabla_i u_0(\mathbf{r}^N)$, and $\mathbf{F}_{\text{ext},0}(\mathbf{r}) = -\rho_0(\mathbf{r})\nabla V_{\text{ext},0}(\mathbf{r})$, with the initial state density profile $\rho_0(\mathbf{r}) = \text{Tr} f_0\hat{\rho}(\mathbf{r})$. The static force density balance (12) is often refered to as Yvon-Born-Green equation [35, 79], in particular for systems that interact by pairwise interparticle force only, such that $\mathbf{F}_{\text{int},0}(\mathbf{r})$ can be written as an integral over the two-body density distribution [7, 14].

D. Trajectories and initial-state differentiation

The Liouvillian dynamics described in Sec. II B can be complemented usefully by a formulation that is more explicitly based on trajectories. This setup can both guide intuition and provide a practical route to constructing analytical solutions and performing simulation work, as will prove useful below. When working with individual trajectories, the two relevant tasks are to realize initial states according to the distribution function $f_0(\mathbf{r}^N, \mathbf{p}^N)$ and further to obtain a representation of the trajectories $\hat{\mathbf{r}}_i(t), \hat{\mathbf{p}}_i(t)$ for all particles $i = 1, \ldots, N$. While for very simple systems analytical work is possible (an example is presented in Sec. IV B), in general and for realistic systems of interest one needs to rely on simulations.

Generating the initial distribution of microstates $f_0(\mathbf{r}^N, \mathbf{p}^N)$ according to the canonical ensemble form (2) is a standard task, which can be efficiently performed via Monte Carlo simulations for the given choice of the initial Hamiltonian H_0 and prescribed value of the inverse temperature β. We recall that the choice of initial Hamiltonian is independent of the subsequent time evolution based on H, where in general $H \neq H_0$. Then, to address the dynamics

the verlocity Verlet algorithm [7, 8] is an apt choice to integrate Hamilton's equations of motion and hence to obtain trajectories $\hat{\mathbf{r}}_i(\mathbf{r}^N, \mathbf{p}^N, t)$ and $\hat{\mathbf{p}}_i(\mathbf{r}^N, \mathbf{p}^N, t)$ for all particles $i = 1, \ldots, N$, starting from the initial state $\mathbf{r}^N, \mathbf{p}^N$.

General Heisenberg observables $\hat{A}(t)$ can then be represented by $\hat{A}(\mathbf{r}^N, \mathbf{p}^N, t) = \hat{A}(\hat{\mathbf{r}}^N(t), \hat{\mathbf{p}}^N(t))$, where we recall that the phase space point $\mathbf{r}^N, \mathbf{p}^N$ on the left hand side is the initial state. On the right hand side the Schrödinger observable $\hat{A}$ is evaluated at the phase space point $\hat{\mathbf{r}}^N(t), \hat{\mathbf{p}}^N(t)$, which is the configuration that the system has reached at time t.

As an important computational point, we demonstrate below that this interpretation gives immediate and practical access to derivatives with respect to the initial state. This will be key for evaluating the arising gauge correlation functions, as laid out in Sec. III, in simulation work. In particular, the task within the gauge correlation framework is to implement a specific initial-state time derivative according to the action of the initial-state Liouvillian L_0, see below for further details. In practice, the differentiation can be performed via a finite-difference scheme by altering the state according to the initial state dynamics and rerunning the simulation. As a numerically robust alternative, the powerful methodology of automatic differentiation can be leveraged. This technique gives access to the $(2Nd) \times (2Nd)$ Jacobian matrix $\partial(\hat{\mathbf{r}}^N(t), \hat{\mathbf{p}}^N(t))/\partial(\mathbf{r}^N, \mathbf{p}^N)$, from which the initial-state time derivative follows naturally as further laid out in Sec. III E. An illustration of the underlying concepts is shown in Fig. 2.

III. DYNAMICAL GAUGE INVARIANCE

A. Exact shift current sum rule as a prototype

We complement the standard dynamical one-body picture described in Sec. II C by rather letting the *initial time* evolution act on the current operator at time t. Hence we apply the thermally scaled initial state Liouvillian βL_0 to the nonequilibrium phase space current:

$$\hat{\mathbf{C}}(\mathbf{r}, t) = \beta L_0 m\hat{\mathbf{J}}(\mathbf{r}, t). \quad (13)$$

We thereby recall the Schrödinger definition (9) of the current observable $\hat{\mathbf{J}}(\mathbf{r})$ and that the dependence on the phase space point, suppressed in the above notation of the Heisenberg current $\hat{\mathbf{J}}(\mathbf{r}, t)$, describes the initial microstate $\mathbf{r}^N, \mathbf{p}^N$ on which βL_0 acts in Eq. (13).

As we demonstrate, the present strategy is both conceptually and in practice distinct from the conventional time evolution generated by $L(t)$, thus leading to new insight that emerges from introducing the classical 'shift current' observable $\hat{\mathbf{C}}(\mathbf{r}, t)$ via Eq. (13). As an illustration, we spell this out more explicitly, using $\hat{\mathbf{J}}(\mathbf{r}, t) = \mathcal{U}^\dagger(t, 0)\hat{\mathbf{J}}(\mathbf{r})$ as the temporally evolved current (9), which upon insertion into Eq. (13) yields

FIG. 2. Illustration of the different types of time evolution as represented by N-body trajectories on phase space. The initial state dynamics are governed by the initial state Liouvillian L_0, as represented by one specific trajectory (dark blue curve on the left). Each point on this trajectory serves as an initial state for the nonequilibrium time evolution (magenta arrows in the middle). These dynamics are transported by the adjoint propagator $\mathcal{U}^\dagger(t,0)$ forward in time and $\mathcal{U}(t,0)$ performs the inverse operation (light blue dashed arrow to the left). The initial state trajectory is mapped via $\mathcal{U}^\dagger(t,0)$ onto a specific trajectory (orange curve on the right) along which dynamical gauge invariance applies.

$\hat{\mathbf{C}}(\mathbf{r},t) = \beta L_0 \mathcal{U}^\dagger(t,0) \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\mathbf{p}_i$. The latter form makes explicit: i) that the mass m cancels out from the definition of $\hat{\mathbf{C}}(\mathbf{r},t)$; ii) that the initial state Liouvillian L_0 acts *after* the adjoint propagator $\mathcal{U}^\dagger(t,0)$ does – we return to the significance of this temporal structure when discussing the trajectory level in Sec. III F; and iii) that at the initial time $t=0$ the shift current reduces to the scaled force density, $\hat{\mathbf{C}}(\mathbf{r},0) = \beta \hat{\mathbf{F}}_0(\mathbf{r})$, as follows from Eq. (10) and where again the subscript 0 indicates the application to the initial state.

We turn to the statistical mechanical consequences that are implied by the shift current observable (13). The mean one-body shift current $\mathbf{C}(\mathbf{r},t)$ follows via averaging Eq. (13) according to $\mathbf{C}(\mathbf{r},t) = \langle \hat{\mathbf{C}}(\mathbf{r},t)\rangle$ $= \mathrm{Tr}\, f_0 \beta L_0 m \hat{\mathbf{J}}(\mathbf{r},t)$. Spelling out the Poisson bracket form of the initial state Liouvillian $L_0 = \{\,\cdot\,, H_0\}$ gives $\mathbf{C}(\mathbf{r},t) = \mathrm{Tr}\, f_0 \{m\hat{\mathbf{J}}(\mathbf{r},t), \beta H_0\}$. Applying the chain rule to the exponential form of the initial state equilibrium distribution function $f_0(\mathbf{r}^N, \mathbf{p}^N)$ leads to $\mathbf{C}(\mathbf{r},t) = -\mathrm{Tr}\, \{m\hat{\mathbf{J}}(\mathbf{r},t), f_0\} = 0$. That the result vanishes follows from the general property of the phase space integral of any Poisson bracket vanishing, $\mathrm{Tr}\,\{g,h\} = 0$, as is readily seen via integration by parts on phase space (for g and h being well behaved).

In summary, we have shown that the average of the shift current (13) vanishes,

$$\mathbf{C}(\mathbf{r},t) = 0, \qquad (14)$$

which is an exact nonequilibrium sum rule. We express

this identity by splitting the left hand side into three contributions:

$$\mathbf{C}_{\mathrm{id}}(\mathbf{r},t) + \mathbf{C}_{\mathrm{int}}(\mathbf{r},t) + \mathbf{C}_{\mathrm{ext}}(\mathbf{r},t) = 0, \qquad (15)$$

where the decomposition into the three terms follows from the corresponding splitting (5) of the initial state Liouvillian L_0; we recall the presence of L_0 in the definition (13) of the shift current observable. As the splitting is crucial in what follows we spell out the details, first recalling that the initial Hamiltonian H_0 consists of kinetic, interparticle and external parts according Eq. (1).

The splitting (15) arises then from decomposing the initial state Liouvillian L_0 according to (5), such that $L_0 = L_{\mathrm{id},0} + L_{\mathrm{int},0} + L_{\mathrm{ext},0}$ in Eq. (13), thereby using the initial particle mass m_0, interparticle potential $u_0(\mathbf{r}^N)$ and external potential $V_{\mathrm{ext},0}(\mathbf{r})$. The corresponding ideal, interparticle, and external shift currents are $\mathbf{C}_\alpha(\mathbf{r},t) = \langle \hat{\mathbf{C}}_\alpha(\mathbf{r},t)\rangle = \langle \beta L_{\alpha,0} m \hat{\mathbf{J}}(\mathbf{r},t)\rangle$, where $\alpha = $ 'id', 'int', and 'ext', respectively. At the initial time, $t=0$, the shift current balance (15) reduces to the equilibrium force density balance (12); we give an extended account of this limit below in Sec. IV A.

It is revealing to compare the structure of the exact dynamical sum rule (15) with that of the one-body equation of motion (11). Both equations are vectorial, and their left hand sides share an analogous splitting into ideal, interparticle, and external contributions. However, the right hand side of the equation of motion (11) is the time derivative of the current, which in general is very different from the universal zero on the right hand side of the nonequilibrium sum rule (15). Before demonstrating the validity of the shift current balance (15) below, we first aim to uncover the fundamental mechanism that lies behind the above derivation.

B. Static phase space shifting via Poisson brackets

The derivation of the shift current balance (14) and thus of its split form (15) in Sec. III A relies on both the one-body localization and on the properties of the Poisson brackets; the latter occur in the initial state Liouvillian L_0; recall Eq. (3). It is hence natural to use the Poisson brackets as a foundation for defining the following spatially localized differential operators:

$$\boldsymbol{\sigma}(\mathbf{r}) = \{\,\cdot\,, m\hat{\mathbf{J}}(\mathbf{r})\}, \qquad (16)$$

where we recall that $\hat{\mathbf{J}}(\mathbf{r})$ is the microscopically given current phase space function (9) and thus $m\hat{\mathbf{J}}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\mathbf{p}_i$. Applying the differential operator (16) to the (negative) Hamiltonian H yields the classical force density operator $\hat{\mathbf{F}}(\mathbf{r})$ according to

$$-\boldsymbol{\sigma}(\mathbf{r})H = \{m\hat{\mathbf{J}}(\mathbf{r}), H\} = L m \hat{\mathbf{J}}(\mathbf{r}) = \hat{\mathbf{F}}(\mathbf{r}), \qquad (17)$$

where we have first written out the Poisson bracket (16) and then identified both the Liouvillian (3) and the classical force density operator $\hat{\mathbf{F}}(\mathbf{r})$ according to Eq. (10).

As an aside we emphasize that no dynamical dependence is implied yet in Eq. (17). The present Schrödinger force density $\hat{\mathbf{F}}(\mathbf{r})$ is related to the corresponding dynamical Heisenberg observable via $\hat{\mathbf{F}}(\mathbf{r}, t) = \mathcal{U}^{\dagger}(t, 0)\hat{\mathbf{F}}(\mathbf{r})$, see Eq. (10).

The properties of the Poisson bracket render the differential operators $\boldsymbol{\sigma}(\mathbf{r})$ anti-self-adjoint on phase space, $\boldsymbol{\sigma}(\mathbf{r}) = -\boldsymbol{\sigma}^{\dagger}(\mathbf{r})$, as follows via integration by parts on phase space [55]. The explicit form of $\boldsymbol{\sigma}(\mathbf{r})$ is obtained straightforwardly by inserting the definition (9) of the classical current operator $\hat{\mathbf{J}}(\mathbf{r})$ into the Poisson bracket (16), which yields $\boldsymbol{\sigma}(\mathbf{r}) = \{\,\cdot\,, \sum_i \mathbf{p}_i \delta(\mathbf{r} - \mathbf{r}_i)\}$. Simplifying gives the explicit form:

$$\boldsymbol{\sigma}(\mathbf{r}) = \sum_i [\delta(\mathbf{r} - \mathbf{r}_i)\nabla_i + \mathbf{p}_i \nabla \delta(\mathbf{r} - \mathbf{r}_i) \cdot \nabla_{\mathbf{p}_i}], \quad (18)$$

which reveals $\boldsymbol{\sigma}(\mathbf{r})$ to be *identical* to the localized differential operators that represent the static 'shifting' gauge invariance of *equilibrium* statistical mechanics [55, 56].

The immediate implications are: i) the validity of non-trivial commutator structure of $\boldsymbol{\sigma}(\mathbf{r})$ and $\boldsymbol{\sigma}(\mathbf{r}')$ at positions $\mathbf{r}, \mathbf{r}'$ [55, 56] and ii) the geometric interpretation via phase space shifting according to particle position displacement and corresponding momentum transform. These general properties continue to hold for the Poisson bracket form (16) as they follow directly from the structure of phase space [55, 56], irrespective of whether a static [55, 56] or the present dynamical statistical mechanical setup is considered. We recall the pertinence of the infinitesimal shifting operators for generating equilibrium sum rules involving forces and hyperforce correlation functions [54–56] according to the mechanism in Eq. (17).

C. Dynamical phase space shifting operators

We generalize the static shifting operators (16) to dynamical counterparts $\boldsymbol{\sigma}(\mathbf{r}, t)$ according to the standard mechanism to transform differential operators:

$$\boldsymbol{\sigma}(\mathbf{r}, t) = \mathcal{U}^{\dagger}(t, 0)\boldsymbol{\sigma}(\mathbf{r})\mathcal{U}(t, 0), \quad (19)$$

where we recall that $\mathcal{U}(t, 0)$ is the (phase space) propagator from the initial time 0 to time t and $\mathcal{U}^{\dagger}(t, 0)$ denotes its (phase space) adjoint. Mirroring the properties of the static version $\boldsymbol{\sigma}(\mathbf{r})$, the dynamical shifting operators are anti-self-adjoint on phase space:

$$\boldsymbol{\sigma}(\mathbf{r}, t) = -\boldsymbol{\sigma}^{\dagger}(\mathbf{r}, t). \quad (20)$$

The proof of Eq. (20) follows straightforwardly via explicit calculation: $[\mathcal{U}^{\dagger}(t, 0)\boldsymbol{\sigma}(\mathbf{r})\mathcal{U}(t, 0)]^{\dagger} = \mathcal{U}^{\dagger}(t, 0)\boldsymbol{\sigma}^{\dagger}(\mathbf{r})\mathcal{U}^{\dagger\dagger}(t, 0) = -\boldsymbol{\sigma}(\mathbf{r}, t)$, where we have first built the overall adjoint by reversing the factors and then have used both $\boldsymbol{\sigma}^{\dagger}(\mathbf{r}) = -\boldsymbol{\sigma}(\mathbf{r})$ and $\mathcal{U}^{\dagger\dagger}(t, 0) = \mathcal{U}(t, 0)$, which allows one to identify the final result via Eq. (19). As an aside, multiplying Eq. (19) by $\mathcal{U}^{\dagger}(t, 0)$ from the

right yields the identity $\boldsymbol{\sigma}(\mathbf{r}, t)\mathcal{U}^{\dagger}(t, 0) = \mathcal{U}^{\dagger}(t, 0)\boldsymbol{\sigma}(\mathbf{r})$ and corresponding multiplication by $\mathcal{U}(t, 0)$ from the left yields $\mathcal{U}(t, 0)\boldsymbol{\sigma}(\mathbf{r}, t) = \boldsymbol{\sigma}(\mathbf{r})\mathcal{U}(t, 0)$.

As an alternative to the propagator 'sandwich' form (19), the dynamical shifting operators can be expressed equivalently as

$$\boldsymbol{\sigma}(\mathbf{r}, t) = \{\,\cdot\,, m\hat{\mathbf{J}}(\mathbf{r}, t)\}. \quad (21)$$

which mirrors the Poisson bracket form of the static shifting operators $\boldsymbol{\sigma}(\mathbf{r})$ given in Eq. (16). The equivalence of Eqs. (19) and (21) is proven by starting from the right hand side of the latter and re-writing as follows: $\{\,\cdot\,, m\hat{\mathbf{J}}(\mathbf{r}, t)\} = \{\mathcal{U}^{\dagger}(t, 0)\mathcal{U}(t, 0)\cdot, \mathcal{U}^{\dagger}(t, 0)m\hat{\mathbf{J}}(\mathbf{r})\} = \mathcal{U}^{\dagger}(t, 0)\{\mathcal{U}(t, 0)\cdot, m\hat{\mathbf{J}}(\mathbf{r})\} = \mathcal{U}^{\dagger}(t, 0)\{\,\cdot\,, m\hat{\mathbf{J}}(\mathbf{r})\}\mathcal{U}(t, 0) = \mathcal{U}^{\dagger}(t, 0)\boldsymbol{\sigma}(\mathbf{r})\mathcal{U}(t, 0)$, where we have first inserted an identity operator $\mathcal{U}^{\dagger}(t, 0)\mathcal{U}(t, 0) = 1$ and then exploited that the propagator and the Poisson bracket commute, due to the latter being a canonical invariant under the Hamiltonian time evolution.

Besides their anti-self-adjointness (20), the dynamical shifting operators $\boldsymbol{\sigma}(\mathbf{r}, t)$, as expressed in the alternative form (21), possess two further key properties. First, when applied to the (thermally scaled negative) Hamiltonian, one obtains

$$\hat{\mathbf{C}}(\mathbf{r}, t) = -\boldsymbol{\sigma}(\mathbf{r}, t)\beta H_0. \quad (22)$$

where $\hat{\mathbf{C}}(\mathbf{r}, t)$ is the shift current phase space function defined via Eq. (13). The validity of Eq. (22) can be seen by expressing its right hand side via the Poisson bracket (21) as $-\{\beta H_0, m\hat{\mathbf{J}}(\mathbf{r}, t)\} = \beta\{m\hat{\mathbf{J}}(\mathbf{r}, t), H_0\} = \beta L_0 m\hat{\mathbf{J}}(\mathbf{r}, t)$, where identifying the initial state Liouvillian L_0 gives the right hand side of Eq. (13). Secondly, when applying $\boldsymbol{\sigma}(\mathbf{r}, t)$ to the initial state probability distribution f_0, given by the Boltzmann form (2), then

$$\boldsymbol{\sigma}(\mathbf{r}, t)f_0 = \hat{\mathbf{C}}(\mathbf{r}, t)f_0, \quad (23)$$

as follows from the chain rule and identifying $\hat{\mathbf{C}}(\mathbf{r}, t)$ via Eq. (22).

As a demonstration of the power of the present dynamical shifting operator formalism, we first reconsider the derivation of the shift current sum rule (15). Viewing this identity from the perspective of the dynamical shifting gauge invariance allows one to formulate a strictly valid derivation in the following strikingly compact form: $\mathbf{C}(\mathbf{r}, t) = \mathrm{Tr}\,\hat{\mathbf{C}}(\mathbf{r}, t)f_0 = \mathrm{Tr}\,\boldsymbol{\sigma}(\mathbf{r}, t)f_0 = -\mathrm{Tr}\,f_0\boldsymbol{\sigma}(\mathbf{r}, t)1 = 0$, where we have first spelled out the average $\langle\hat{\mathbf{C}}(\mathbf{r}, t)\rangle$, then introduced $\boldsymbol{\sigma}(\mathbf{r}, t)$ via Eq. (23), and reordered terms by using that $\boldsymbol{\sigma}(\mathbf{r}, t)$ is anti-self-adjoint (20). That the resulting average vanishes is trivially due to $\boldsymbol{\sigma}(\mathbf{r}, t)1 = 0$, as follows from Eq. (21) and the vanishing Poisson bracket of any constant.

The connection of the dynamical generalization (21) of equilibrium phase space shifting [55, 56] with the nonequilibrium shift current sum rule (15) reveals the latter to originate from dynamical gauge invariance. As we demonstrate in the following, the dynamical gauge

concept allows one to address a significantly wider class of cases than covered thus far, by incorporating general hyperobservables into the framework.

D. Exact hypercurrent sum rules for general observables

We consider a general phase space function $\hat{A}(\mathbf{r}^N, \mathbf{p}^N)$ to act as a hyperobservable that is of physical interest in the nonequilibrium situation under consideration. We recall both that the corresponding Heisenberg observable $\hat{A}(\mathbf{r}^N, \mathbf{p}^N, t)$ is obtained via application of the adjoint propagator according to $\hat{A}(t) = \mathcal{U}^\dagger(t, 0)\hat{A}$, see Sec. II B, and that trajectory-based methods facilitate alternative access, as described in Sec. II D. We follow Refs. [54–56] in defining the *static* hyperforce density as the phase space function given by

$$\hat{\mathbf{S}}_A(\mathbf{r}) = \boldsymbol{\sigma}(\mathbf{r})\hat{A}, \qquad (24)$$

where we recall the explicit form (18) of the static differential operators $\boldsymbol{\sigma}(\mathbf{r})$. The corresponding Heisenberg observable $\hat{\mathbf{S}}_A(\mathbf{r}, t)$ then follows via the standard procedure as $\hat{\mathbf{S}}_A(\mathbf{r}, t) = \mathcal{U}^\dagger(t, 0)\hat{\mathbf{S}}_A(\mathbf{r})$. Using Eq. (24) and inserting an identity operator $\mathcal{U}(t, 0)\mathcal{U}^\dagger(t, 0) = 1$ leads to $\hat{\mathbf{S}}_A(\mathbf{r}, t) = \mathcal{U}^\dagger(t, 0)\boldsymbol{\sigma}(\mathbf{r})\mathcal{U}(t, 0)\mathcal{U}^\dagger(t, 0)\hat{A}$. The first three factors therein can be identified as the dynamical shifting operator $\boldsymbol{\sigma}(\mathbf{r}, t)$, see Eq. (19), and the remaining two factors constitute the Heisenberg observable $\hat{A}(t)$, see Eq. (7). Hence we formulate the *dynamical* hyperforce density as the following Heisenberg observable:

$$\hat{\mathbf{S}}_A(\mathbf{r}, t) = \boldsymbol{\sigma}(\mathbf{r}, t)\hat{A}(t), \qquad (25)$$

which generalizes the static equivalent $\hat{\mathbf{S}}_A(\mathbf{r})$ given by Eq. (24). Building the dynamical average in the standard way gives the dynamical hyperforce density as $\mathbf{S}_A(\mathbf{r}, t) = \langle\hat{\mathbf{S}}_A(\mathbf{r}, t)\rangle$. As a complement, in trajectory-based work, for a given form of $\hat{\mathbf{S}}_A(\mathbf{r})$, straightforward evaluation gives $\hat{\mathbf{S}}_A(\mathbf{r}, t) = \hat{\mathbf{S}}_A(\mathbf{r}; \hat{\mathbf{r}}^N(t), \hat{\mathbf{p}}^N(t))$, where we have written out the phase space dependence to emphasize that this expression is ready to be averaged over initial states once the trajectory is known for the considered initial microstates; recall that the static hyperforce density $\hat{\mathbf{S}}_A(\mathbf{r}; \mathbf{r}^N, \mathbf{p}^N) = \boldsymbol{\sigma}(\mathbf{r})\hat{A}$ is explicitly available for given form of the hyperobservable $\hat{A}$ [54–56].

Spelling out the average allows one to proceed in explicit form: $\mathbf{S}_A(\mathbf{r}, t) = \text{Tr}\, f_0 \boldsymbol{\sigma}(\mathbf{r}, t)\hat{A}(t) = \text{Tr}\,\hat{A}(t)\boldsymbol{\sigma}^\dagger(\mathbf{r}, t)f_0$, where we have introduced the shifting operator via Eq. (25) and then reordered the integrand via building the adjoint. Using the anti-self-adjoint property (20) of $\boldsymbol{\sigma}(\mathbf{r}, t)$ allows one to rewrite the result as $-\text{Tr}\,\hat{A}(t)\boldsymbol{\sigma}(\mathbf{r}, t)f_0 = -\text{Tr}\,\hat{A}(t)\hat{\mathbf{C}}(\mathbf{r}, t)f_0 = -\langle\hat{A}(t)\hat{\mathbf{C}}(\mathbf{r}, t)\rangle = -\langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle$, where we have used the generation (23) of the shift current Heisenberg observable $\hat{\mathbf{C}}(\mathbf{r}, t)$, then have written the overall result as a nonequilibrium average, and in the last step have interchanged

the two phase space functions. Bringing the result to the initial left hand side yields the following dynamical hypercurrent balance:

$$\mathbf{S}_A(\mathbf{r}, t) + \langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle = 0, \qquad (26)$$

which is exact. Using compact notation for the hypercurrent correlation function, $\mathbf{C}_A(\mathbf{r}, t) = \langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle$, allows one to express the sum rule (26) in the alternative form:

$$\mathbf{S}_A(\mathbf{r}, t) + \mathbf{C}_A(\mathbf{r}, t) = 0, \qquad (27)$$

which expresses the vanishing sum of hyperforce density (first term) and hypercurrent correlation function (second term).

As a simple consistency check, making the choice $\hat{A} = 1$ leads to $\hat{\mathbf{S}}_{\hat{A}=1}(\mathbf{r}, t) = 0$ and hence on average $\mathbf{S}_{\hat{A}=1}(\mathbf{r}, t) = 0$. Thus upon recalling $\langle\hat{\mathbf{C}}(\mathbf{r}, t)\rangle = \mathbf{C}(\mathbf{r}, t) = \mathbf{C}_{\hat{A}=1}(\mathbf{r}, t)$, the hypercurrent identity (26) reduces to $\mathbf{C}(\mathbf{r}, t) = 0$, which constitutes the shift current identity (15). As an aside, the hypercurrent correlation term in Eq. (26) can alternatively be expressed as $\langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle = \text{cov}(\hat{\mathbf{C}}(\mathbf{r}, t), \hat{A}(t))$, where the covariance of two observables is defined in the standard form: $\text{cov}(\hat{\mathbf{C}}(\mathbf{r}, t), \hat{A}(t)) = \langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle - \mathbf{C}(\mathbf{r}, t)\langle\hat{A}(t)\rangle$. The equivalence of correlation and covariance follows from $\mathbf{C}(\mathbf{r}, t) = 0$, see the shift current balance (15). Working with covariances can have practical advantages in simulation work due to systematic subtraction of sampling uncertainties, see e.g. Ref. [54] for such work in equilibrium.

The dynamical correlation function $\langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle$ that features in Eq. (26) can be split into ideal, interparticle, and external contributions, according to $\langle\hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t)\rangle = \langle\hat{\mathbf{C}}_{\text{id}}(\mathbf{r}, t)\hat{A}(t)\rangle + \langle\hat{\mathbf{C}}_{\text{int}}(\mathbf{r}, t)\hat{A}(t)\rangle + \langle\hat{\mathbf{C}}_{\text{ext}}(\mathbf{r}, t)\hat{A}(t)\rangle$, where we recall the definition of the ideal, interparticle, and external particle shift current, as arising from the splitting of the initial state Liouvillian; see the corresponding description given below Eq. (15). The splitting allows one to express the hypercurrent sum rule (26) in equivalent form as:

$$\mathbf{S}_A(\mathbf{r}, t) + \mathbf{C}_A^{\text{id}}(\mathbf{r}, t) + \mathbf{C}_A^{\text{int}}(\mathbf{r}, t) + \mathbf{C}_A^{\text{ext}}(\mathbf{r}, t) = 0, \quad (28)$$

where the three partial hypercurrent correlation functions are defined as $\mathbf{C}_A^{\text{id}}(\mathbf{r}, t) = \langle\hat{\mathbf{C}}_{\text{id}}(\mathbf{r}, t)\hat{A}(t)\rangle$, $\mathbf{C}_A^{\text{int}}(\mathbf{r}, t) = \langle\hat{\mathbf{C}}_{\text{int}}(\mathbf{r}, t)\hat{A}(t)\rangle$, and $\mathbf{C}_A^{\text{ext}}(\mathbf{r}, t) = \langle\hat{\mathbf{C}}_{\text{ext}}(\mathbf{r}, t)\hat{A}(t)\rangle$. We defer a detailed description of the connection of the dynamical identity (28) with the static hyperforce theory [54–56] to Sec. IV A below.

The present hypercurrent formalism rests centrally on the properties of the dynamical shifting operators $\boldsymbol{\sigma}(\mathbf{r}, t)$, as alternatively given by Eqs. (19) and (21). The application of these differential operators generates both the dynamical hyperforce density $\hat{\mathbf{S}}_A(\mathbf{r}, t)$, given by Eq. (25), and the dynamical shift current $\hat{\mathbf{C}}(\mathbf{r}, t)$, which arises from Eq. (22). As $\hat{\mathbf{C}}(\mathbf{r}, t)$ is also given as the phase space function (13), the emerging temporal correlation structure

rests centrally on the relevance of the initial state Liouvillian L_0 for the *nonequilibrium* problem, as we expand on in the following.

E. Initial state time differentiation

Identifying and applying the initial state Liouvillian L_0 to act inside of a nonequilibrium average forms a crucial step in the above laid out hypercurrent theory, see the central definition (13) of $\hat{\mathbf{C}}(\mathbf{r}, t)$. We here explore the implied temporal structure further. For a given Heisenberg hyperobservable $\hat{A}(\mathbf{r}^N, \mathbf{p}^N, t)$ we define its *initial state time derivative* as

$$\hat{A}^\bullet(t) = L_0 \hat{A}(t) \tag{29}$$

$$= \frac{\partial}{\partial s} \mathcal{U}_0^\dagger(s, 0) \hat{A}(t) \Big|_{s=0}, \tag{30}$$

where the superscript bold dot denotes the application of L_0 to $\hat{A}(t)$ in Eq. (29) such that spelling out the initial state Liouvillian in Poisson bracket form (3) implies $\hat{A}^\bullet(t) = \{\hat{A}(t), H_0\}$. In the alternative form (30) the adjoint initial state propagator $\mathcal{U}_0^\dagger(s, 0) = e^{sL_0}$ performs the initial state dynamics from time 0 to time s. We recall that the initial ensemble, as characterized by the Hamiltonian H_0 and corresponding thermal distribution f_0 given by Eq. (2), remains stationary under its inherent time evolution according to its corresponding Liouvillian L_0; see the description of this setup in Sec. II A.

To make the connection of the initial state time differentitation (29) with the shift current observable $\hat{\mathbf{C}}(\mathbf{r}, t)$, as given by Eq. (13), we first define the scaled one-body current $\hat{\mathbf{S}}(\mathbf{r}, t) = \beta m \hat{\mathbf{J}}(\mathbf{r}, t)$, where we recall the definition (9) of the one-body current and that β is the inverse temperature of the initial state. The shift current is then obtained by applying the initial state Liouvillian L_0 according to Eq. (29), which yields

$$\hat{\mathbf{C}}(\mathbf{r}, t) = \hat{\mathbf{S}}^\bullet(\mathbf{r}, t) = L_0 \hat{\mathbf{S}}(\mathbf{r}, t). \tag{31}$$

such that $\hat{\mathbf{C}}(\mathbf{r}, t) = \beta m \hat{\mathbf{J}}^\bullet(\mathbf{r}, t)$, as follows from Eq. (13). As an aside, the explicit dependence on mass m scales out, as described in Sec. III A and further elaborated in Sec. III F below.

Using the bold dot notation (29), the hypercurrent sum rule (26) attains the following form:

$$\mathbf{S}_A(\mathbf{r}, t) + \langle \hat{\mathbf{S}}^\bullet(\mathbf{r}, t) \hat{A}(t) \rangle = 0. \tag{32}$$

We next derive a further nonequilibrium sum rule that is satisfied by the second term on the above left hand side, which is the hypercurrent correlation function $\mathbf{C}_A(\mathbf{r}, t) = \langle \hat{\mathbf{S}}^\bullet(\mathbf{r}, t) \hat{A}(t) \rangle$. Writing out explicitly the implied average allows one to proceed as follows: $\mathrm{Tr}\, f_0 \hat{A}(t) \hat{\mathbf{S}}^\bullet(\mathbf{r}, t) = \mathrm{Tr}\, f_0 \hat{A}(t) L_0 \hat{\mathbf{S}}(\mathbf{r}, t) = \mathrm{Tr}\, \hat{\mathbf{S}}(\mathbf{r}, t) L_0^\dagger \hat{A}(t) f_0$, where we have first used Eq. (31) to reintroduce L_0 and then reordered via building the adjoint. We next use that the initial state

Liouvillian is anti-self-adjoint, $L_0^\dagger = -L_0$, and that the initial state distribution f_0 is stationary under the initial time evolution, $L_0 f_0 = 0$. Upon reordering the result implies the following exact hypercurrent 'swap' identity:

$$\langle \hat{\mathbf{S}}^\bullet(\mathbf{r}, t) \hat{A}(t) \rangle + \langle \hat{\mathbf{S}}(\mathbf{r}, t) \hat{A}^\bullet(t) \rangle = 0, \tag{33}$$

where we recall that according to the notation (29), $\hat{A}^\bullet(t) = L_0 \hat{A}(t) = \{\hat{A}(t), H_0\}$ is the initial state derivative of the Heisenberg observable $\hat{A}(t)$.

We can combine Eqs. (26) and (33) to obtain the following alternative hypercurrent sum rule:

$$\mathbf{S}_A(\mathbf{r}, t) - \langle \hat{\mathbf{S}}(\mathbf{r}, t) \hat{A}^\bullet(t) \rangle = 0. \tag{34}$$

The theory thus far developed applies to general observables $\hat{A}$ and it is formally exact for general nonequilibrium Hamiltonian dynamics, as generated by a time-dependent many-body Hamiltonian H and starting from an initial thermal state with Hamiltonian H_0 and inverse temperature β. We next describe the implications when working with trajectories.

F. Trajectory-based differentiation

As laid out in Sec. II D a trajectory-based picture rests on having access to $\hat{\mathbf{r}}_i(t)$ and $\hat{\mathbf{p}}_i(t)$ for all particles $i = 1, \ldots, N$. We recall that the full phase space dependence, when viewing the particle positions and momenta as Heisenberg observables, is $\hat{\mathbf{r}}_i(\mathbf{r}^N, \mathbf{p}^N, t)$ and $\hat{\mathbf{p}}_i(\mathbf{r}^N, \mathbf{p}^N, t)$, where the argument $\mathbf{r}^N, \mathbf{p}^N$ denotes the phase space point that represents the initial configuration at $t = 0$. One can express this relationship succinctly at the initial time as: $\hat{\mathbf{r}}_i(\mathbf{r}^N, \mathbf{p}^N, 0) = \mathbf{r}_i$ and $\hat{\mathbf{p}}_i(\mathbf{r}^N, \mathbf{p}^N, 0) = \mathbf{p}_i$.

We hence initial-state time differentiate the entire trajectory as follows:

$$\hat{\mathbf{r}}_i^\bullet(t) = L_0 \hat{\mathbf{r}}_i(t) = \{\hat{\mathbf{r}}_i(t), H_0\}$$
$$= \sum_j \left(\mathbf{v}_{j,0} \cdot \nabla_j \hat{\mathbf{r}}_i(t) + \mathbf{f}_{j,0} \cdot \nabla_{\mathbf{p}_j} \hat{\mathbf{r}}_i(t) \right), \tag{35}$$

$$\hat{\mathbf{p}}_i^\bullet(t) = L_0 \hat{\mathbf{p}}_i(t) = \{\hat{\mathbf{p}}_i(t), H_0\}$$
$$= \sum_j \left(\mathbf{v}_{j,0} \cdot \nabla_j \hat{\mathbf{p}}_i(t) + \mathbf{f}_{j,0} \cdot \nabla_{\mathbf{p}_j} \hat{\mathbf{p}}_i(t) \right), \tag{36}$$

where $\mathbf{v}_{j,0} = \mathbf{p}_j / m_0$ denotes the initial state velocity, $\mathbf{f}_{j,0} = -\nabla_j H_0$ is the initial state force of particle j, and the sums run over all particles $j = 1, \ldots, N$. The phase space derivatives ∇_j and $\nabla_{\mathbf{p}_j}$ can be viewed as measuring the dependence of position $\hat{\mathbf{r}}_i(t)$ in Eq. (35) and of momentum $\hat{\mathbf{p}}_i(t)$ in Eq. (36) of particle i upon changes in the initial data of particle j. Rather than forming a generic perturbation, the phase space derivatives are then weighted specifically by, respectively, the initial state velocity and initial force of particle j. Summation over all particles j then yields $\hat{\mathbf{r}}_i^\bullet(t)$ and $\hat{\mathbf{p}}_i^\bullet(t)$. In relation to the

Liouvillian splitting (5) into ideal, interparticle, and external contributions, the first term in the sum in both Eqs. (35) and (36) arises from $L_{\rm id}$ and the second term arises from the sum $L_{\rm int} + L_{\rm ext}$.

The initial state derivative of a general Heisenberg observable $\hat{A}(t)$ then follows from using the chain rule, which gives

$$\hat{A}^{\bullet}(t) = \sum_i \left[(\nabla_i \hat{A})(t) \cdot \hat{\mathbf{r}}_i^{\bullet}(t) + (\nabla_{\mathbf{p}_i} \hat{A})(t) \cdot \hat{\mathbf{p}}_i^{\bullet}(t) \right], \quad (37)$$

where we use the notation $(\nabla_i \hat{A})(t) = \mathcal{U}^{\dagger}(t,0)\nabla_i \hat{A}$ and $(\nabla_{\mathbf{p}_i}\hat{A})(t) = \mathcal{U}^{\dagger}(t,0)\nabla_{\mathbf{p}_i}\hat{A}$ to respectively indicate the temporally propagated phase space position and momentum gradients of the hyperobservable $\hat{A}(\mathbf{r}^N, \mathbf{p}^N)$.

We next turn to the shift current (31), which we recall as $\hat{\mathbf{C}}(\mathbf{r}, t) = \hat{\mathbf{S}}^{\bullet}(\mathbf{r}, t)$ with $\hat{\mathbf{S}}(\mathbf{r}, t) = \beta m \hat{\mathbf{J}}(\mathbf{r}, t)$. The present formulation allows one to obtain the following trajectory-based representation: $\hat{\mathbf{C}}(\mathbf{r}, t) = \sum_i [\hat{\mathbf{r}}_i^{\bullet}(t) \cdot (\nabla_i \hat{\mathbf{S}})(\mathbf{r}, t) + \hat{\mathbf{p}}_i^{\bullet}(t) \cdot (\nabla_{\mathbf{p}_i}\hat{\mathbf{S}})(\mathbf{r}, t)]$. We express the sum of these two terms as

$$\hat{\mathbf{C}}(\mathbf{r}, t) = \nabla \cdot \hat{\boldsymbol{\tau}}_C(\mathbf{r}, t) + \hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t), \quad (38)$$

where the shift stress tensor $\hat{\boldsymbol{\tau}}_C(\mathbf{r}, t)$ and the shift acceleration current observable $\hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t)$ are given respectively by

$$\hat{\boldsymbol{\tau}}_C(\mathbf{r}, t) = -\beta \sum_i \delta(\mathbf{r} - \hat{\mathbf{r}}_i(t)) \hat{\mathbf{r}}_i^{\bullet}(t) \hat{\mathbf{p}}_i(t), \quad (39)$$

$$\hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t) = \beta \sum_i \delta(\mathbf{r} - \hat{\mathbf{r}}_i(t)) \hat{\mathbf{p}}_i^{\bullet}(t). \quad (40)$$

That the shift current $\hat{\mathbf{C}}(\mathbf{r}, t)$ has the natural splitting (38) mirrors the decomposition of the force density $\hat{\mathbf{F}}(\mathbf{r}, t)$, given below Eq. (10) in the form: $\hat{\mathbf{F}}(\mathbf{r}, t) = \nabla \cdot \hat{\boldsymbol{\tau}}(\mathbf{r}, t) + \hat{\mathbf{F}}_U(\mathbf{r}, t)$, where the potential force density observable, $\hat{\mathbf{F}}_U(\mathbf{r}, t) = \hat{\mathbf{F}}_{\rm int}(\mathbf{r}, t) + \hat{\mathbf{F}}_{\rm ext}(\mathbf{r}, t)$, combines interparticle and external contributions. Note also the structural similarity of Eq. (39) with the definition of the standard kinetic stress tensor observable $\hat{\boldsymbol{\tau}}(\mathbf{r}, t)$, given in Schrödinger form below Eq. (10), and of the shift acceleration current observable (40) with $\hat{\mathbf{F}}_U(\mathbf{r}, t)$.

Upon averaging, one can show that the mean shift stress tensor, as given by $\boldsymbol{\tau}_C(\mathbf{r}, t) = \langle \hat{\boldsymbol{\tau}}_C(\mathbf{r}, t) \rangle$, and the mean shift acceleration current, as given by $\mathbf{C}_{\rm acc}(\mathbf{r}, t) = \langle \hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t) \rangle$, are related to the *instantaneous* dynamical density profile $\rho(\mathbf{r}, t)$ and its gradient respectively via:

$$\boldsymbol{\tau}_C(\mathbf{r}, t) = -\rho(\mathbf{r}, t)\mathbb{1} \quad (41)$$
$$\mathbf{C}_{\rm acc}(\mathbf{r}, t) = \nabla \rho(\mathbf{r}, t), \quad (42)$$

where $\mathbb{1}$ denotes the $d \times d$-unit matrix. The identity (42) follows straightforwardly from writing out the average on the left hand side explicitly; we give a compact account: $\mathbf{C}_{\rm acc}(\mathbf{r}, t) = \mathrm{Tr}\, f_0 \hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t) = \mathrm{Tr}\, f(t) \mathcal{U}(t, 0) \sum_i \delta(\mathbf{r} - \hat{\mathbf{r}}_i(t)) \{\hat{\mathbf{p}}_i(t), \beta H_0\}$. Then applying the propagator and subsequently evaluating the Poisson bracket gives: $\mathrm{Tr}\, f(t) \sum_i \delta(\mathbf{r} - \mathbf{r}_i) \{\mathbf{p}_i, \mathcal{U}(t, 0)\beta H_0\} =$

$-\mathrm{Tr}\, f(t) \sum_i \delta(\mathbf{r} - \mathbf{r}_i) \nabla_i [\mathcal{U}(t, 0)\beta H_0] = \mathrm{Tr} \sum_i \delta(\mathbf{r} - \mathbf{r}_i) \nabla_i f(t) = \nabla \rho(\mathbf{r}, t)$, where we have used that $f(t) = e^{-\beta \mathcal{U}(t,0) H_0}/Z_0$ and the last step follows from integration by parts with respect to the particle positions, with $\nabla_i \delta(\mathbf{r} - \mathbf{r}_i) = -\nabla \delta(\mathbf{r} - \mathbf{r}_i)$ and then identifying the average as the gradient of the dynamical density profile. Similar steps prove Eq. (41).

The identities (41) and (42) verify explicitly the shift current sum rule (15) in the form

$$\mathbf{C}_{\rm acc}(\mathbf{r}, t) + \nabla \cdot \boldsymbol{\tau}_C(\mathbf{r}, t) = 0. \quad (43)$$

Applying the splitting (38) into transport and acceleration contributions analogously to the hypercurrent correlation function $\mathbf{C}_A(\mathbf{r}, t) = \langle \hat{\mathbf{C}}(\mathbf{r}, t)\hat{A}(t) \rangle$, as it appears in the hypercurrent sum rule (26), leads to the following decomposition:

$$\mathbf{C}_A(\mathbf{r}, t) = \langle \hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t)\hat{A}(t) \rangle + \nabla \cdot \langle \hat{\boldsymbol{\tau}}_C(\mathbf{r}, t)\hat{A}(t) \rangle, \quad (44)$$

where each term on the right had side can further be split into ideal, interparticle, and external contributions using the Liouvillian splitting (5) for the initial state derivative.

For completeness, in explicit form the classical hyperforce density observable $\hat{\mathbf{S}}_A(\mathbf{r})$, see Eqs. (24) for its emergence from applying the shifting operator to the observable $\hat{A}$, is given by

$$\hat{\mathbf{S}}_A(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\nabla_i \hat{A} + \nabla \cdot \sum_i \delta(\mathbf{r} - \mathbf{r}_i)(\nabla_{\mathbf{p}_i}\hat{A})\mathbf{p}_i. \quad (45)$$

One can decompose as $\hat{\mathbf{S}}_A(\mathbf{r}) = \hat{\mathbf{S}}_A^{\rm pos}(\mathbf{r}) + \nabla \cdot \hat{\boldsymbol{\tau}}_A(\mathbf{r})$, where the position contribution to the hyperforce density is $\hat{\mathbf{S}}_A^{\rm pos}(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)\nabla_i \hat{A}$ and the hyperstress tensor is given by $\hat{\boldsymbol{\tau}}_A(\mathbf{r}) = \sum_i \delta(\mathbf{r} - \mathbf{r}_i)(\nabla_{\mathbf{p}_i}\hat{A})\mathbf{p}_i$.

Following arguments that are very similar to those given to derive Eq. (42), one can further show that

$$\langle \hat{\boldsymbol{\tau}}_C(\mathbf{r}, t)\hat{A}(t) \rangle + \boldsymbol{\tau}_A(\mathbf{r}, t) = -\langle \hat{\rho}(\mathbf{r}, t)\hat{A}(t) \rangle \mathbb{1}, \quad (46)$$
$$\langle \hat{\mathbf{C}}_{\rm acc}(\mathbf{r}, t)\hat{A}(t) \rangle + \mathbf{S}_A^{\rm pos}(\mathbf{r}, t) = \nabla \langle \hat{\rho}(\mathbf{r}, t)\hat{A}(t) \rangle, \quad (47)$$

where $\boldsymbol{\tau}_A(\mathbf{r}, t) = \langle \hat{\boldsymbol{\tau}}_A(\mathbf{r}, t) \rangle$ and $\mathbf{S}_A^{\rm pos}(\mathbf{r}, t) = \langle \hat{\mathbf{S}}_A^{\rm pos}(\mathbf{r}, t) \rangle$. As a consistency check, adding Eq. (47) and the divergence of Eq. (46) yields the dynamical hypercurrent sum rule (26), when identifying the hypercurrent correlation function $\mathbf{C}_A(\mathbf{r}, t)$ via Eq. (44) and the hyperforce density as $\mathbf{S}_A(\mathbf{r}, t) = \mathbf{S}_A^{\rm pos}(\mathbf{r}, t) + \nabla \cdot \boldsymbol{\tau}_A(\mathbf{r}, t)$, as follows from the splitting given below Eq. (45).

G. Concrete choices of hyperobservables

To illustrate the broad range of consequences of the above laid out dynamical statistical mechanical gauge invariance, we specialize the general hypercurrent sum rule (26) for several concrete choices for the hyperobservable $\hat{A}$. We first consider both the Hamiltonian, $\hat{A} = H$,

and the interparticle potential, $\hat{A} = u(\mathbf{r}^N)$, which respectively yields:

$$\mathbf{F}(\mathbf{r},t) - \langle \hat{\mathbf{C}}(\mathbf{r},t)H(t)\rangle = 0, \qquad (48)$$

$$\mathbf{F}_{\text{int}}(\mathbf{r},t) - \langle \hat{\mathbf{C}}(\mathbf{r},t)u(t)\rangle = 0, \qquad (49)$$

where we recall $\mathbf{F}(\mathbf{r},t)$ as the total dynamical mean force density and $\mathbf{F}_{\text{int}}(\mathbf{r},t)$ as its interparticle contribution; see their descriptions below Eq. (10).

As two further specific choices, we take the total momentum, $\hat{A} = \hat{\mathbf{P}} = \sum_i \mathbf{p}_i$, and the sum of positions, $\hat{A} = \hat{\mathbf{R}} = \sum_i \mathbf{r}_i$, where the latter can be viewed as the product of the center of mass, $\hat{\mathbf{R}}/N$, and the total particle number N. The generic hypercurrent sum rule (26) then attains the following two respective forms:

$$\nabla m \mathbf{J}(\mathbf{r},t) + \langle \hat{\mathbf{C}}(\mathbf{r},t)\hat{\mathbf{P}}(t)\rangle = 0, \qquad (50)$$

$$\rho(\mathbf{r},t)\mathbb{1} + \langle \hat{\mathbf{C}}(\mathbf{r},t)\hat{\mathbf{R}}(t)\rangle = 0. \qquad (51)$$

The four sum rules (48)–(51) are exact and they are noteworthy, as in each instance a very common instantaneous observable (first term on each left hand side) is related rigorously to a specific correlation function (corresponding second term) with the shift current. Recalling the initial-time form (13) of the shift current operator $\hat{\mathbf{C}}(\mathbf{r},t)$ reveals the inherent temporally nonlocal character of these correlation functions; in contrast the corresponding hyperforce density [as realized by the respective first term in (48)–(51)] carries instantaneous dependence on time t.

On the basis of the swap identity (33) we can alternatively formulate each of the second terms in Eqs. (48)–(51) by using the scaled current $\hat{\mathbf{S}}(\mathbf{r},t) = m\beta\hat{\mathbf{J}}(\mathbf{r},t)$ instead of the shift current $\hat{\mathbf{C}}(\mathbf{r},t) = \mathbf{S}^{\bullet}(\mathbf{r},t) = L_0\beta m\hat{\mathbf{J}}(\mathbf{r},t)$. We obtain explicitly from Eq. (33) for the Hamiltonian correlation function in Eq. (48): $\langle \hat{\mathbf{C}}(\mathbf{r},t)H(t)\rangle = -\langle \hat{\mathbf{S}}(\mathbf{r},t)H^{\bullet}(t)\rangle$, for the interparticle potential in Eq. (49): $\langle \hat{\mathbf{C}}(\mathbf{r},t)u(t)\rangle = -\langle \hat{\mathbf{S}}(\mathbf{r},t)u^{\bullet}(t)\rangle$, for the total momentum in Eq. (50): $\langle \hat{\mathbf{C}}(\mathbf{r},t)\hat{\mathbf{P}}(t)\rangle = -\langle \hat{\mathbf{S}}(\mathbf{r},t)\hat{\mathbf{P}}^{\bullet}(t)\rangle$, and for the sum of particle positions in Eq. (51): $\langle \hat{\mathbf{R}}(t)\hat{\mathbf{C}}(\mathbf{r},t)\rangle = -\langle \hat{\mathbf{R}}^{\bullet}(t)\hat{\mathbf{S}}(\mathbf{r},t)\rangle$, where we recall that the superscript bold dot indicates the application of the initial state Liouvillian L_0 according to Eq. (29).

IV. APPLICATIONS

A. Equilibrium limit of hypercurrent correlations

We first demonstrate the consistency of the dynamical gauge theory formulated in Sec. III with the static hyperforce approach of Refs. [54–56]. We hence restrict the above general nonequilibrium setup, as described in detail in Sec. II A, to cases with no switching at the initial time, such that the Hamiltonian remains unchanged, $H = H_0$, at all times. We consider the

implications for the hypercurrent sum rule (26) which we recall contains for the special case $\hat{A} = 1$ the shift current balance (14). The first term on the left hand side of Eq. (26), i.e., the dynamical hyperforce density $\mathbf{S}_A(\mathbf{r},t)$, simply becomes $\mathbf{S}_A(\mathbf{r})$, independent of time. The hypercurrent correlation function is $\langle \hat{\mathbf{C}}_0(\mathbf{r},t)\hat{A}(t)\rangle$, where the subscript 0 denotes the equilibrium shift current observable $\hat{\mathbf{C}}_0(\mathbf{r},t) = \beta L_0 m_0 \hat{\mathbf{J}}_0(\mathbf{r},t)$ as is obtained from Eq. (13) for the initial state. We can re-write this on the basis of Eq. (13) as: $\beta L_0 \mathcal{U}_0^{\dagger}(t,0)m_0\hat{\mathbf{J}}_0(\mathbf{r}) = \beta \mathcal{U}_0^{\dagger}(t,0)L_0 m_0\hat{\mathbf{J}}_0(\mathbf{r}) = \beta \mathcal{U}_0^{\dagger}(t,0)\hat{\mathbf{F}}_0(\mathbf{r}) = \beta\hat{\mathbf{F}}_0(\mathbf{r},t)$, where we have first exploited that L_0 and $\mathcal{U}_0^{\dagger}(t,0)$ commute and then have identified the initial state force density observable $\hat{\mathbf{F}}_0(\mathbf{r},t)$, as corresponds to H_0; see Sec. II C.

Overall we hence obtain $\langle \hat{\mathbf{C}}_0(\mathbf{r},t)\hat{A}(t)\rangle = \langle \beta\hat{\mathbf{F}}_0(\mathbf{r},t)\hat{A}(t)\rangle = \langle \beta\hat{\mathbf{F}}_0(\mathbf{r})\hat{A}\rangle$, where the time dependence in the last step vanishes due to stationarity in equilibrium. We hence obtain the following equilibrium identity:

$$\mathbf{S}_A(\mathbf{r}) + \langle \beta\hat{\mathbf{F}}_0(\mathbf{r})\hat{A}\rangle = 0, \qquad (52)$$

which is the static hyperforce balance [54–56]. The force correlation function can thereby be decomposed into its three constituent (ideal, interparticle, and external) parts, such that alternatively to Eq. (52) we can write

$$\mathbf{S}_A(\mathbf{r}) + \langle \beta\hat{\mathbf{F}}_{\text{id},0}(\mathbf{r})\hat{A}\rangle + \langle \beta\hat{\mathbf{F}}_{\text{int},0}(\mathbf{r})\hat{A}\rangle$$
$$+ \langle \beta\hat{\mathbf{F}}_{\text{ext},0}(\mathbf{r})\hat{A}\rangle = 0, \qquad (53)$$

where the individual force density contributions, given in general form below Eq. (9), are those for the initial Hamiltonian H_0.

The above reduction to the static gauge invariance hyperforce theory constitutes an important consistency check. However, much more interesting structure is revealed by not enforcing the static limit but rather honouring the equilibrium *dynamics*. As before, we consider the situation of continuing equilibrium, such that $H = H_0$ at all times with no switching nor any further explicit time dependence occurring in H. As described above, the hyperforce density then looses its time dependence, $\mathbf{S}_A(\mathbf{r},t) = \mathbf{S}_A(\mathbf{r})$. However, such reduction does not occur in general when splitting the hypercurrent correlation function, $\langle \hat{\mathbf{C}}(\mathbf{r},t)\hat{A}(t)\rangle$, as we demonstrate in the following.

We hence forgo the operator re-ordering that led to Eq. (52) and retain the structure (13) of the shift current observable: $\hat{\mathbf{C}}(\mathbf{r},t) = \beta L_0 m\hat{\mathbf{J}}(\mathbf{r},t)$. Then the present equilibrium situation allows one to obtain from splitting the Liouvillian the result: $\hat{\mathbf{C}}_{0,\alpha}(\mathbf{r},t) = \beta L_{0,\alpha}m_0\mathbf{J}(\mathbf{r},t)$, where the subscript α = 'id', 'int', and 'ext' labels the different ideal, interparticle, and external contributions. In contrast to the above derivation of the static hyperforce sum rule (52), here no further simplification arises, as the partial Liouvillians $L_{0,\alpha}$ do *not* in general commute with the full initial state adjoint propagator $\mathcal{U}_0^{\dagger}(t,0)$.

We can conclude that two properties render the present dynamical equilibrium limit non-trivial and different from the static case: i) In general the partial hyperforce and hypercurrent correlation functions differ from each other, $\langle\beta\hat{\mathbf{F}}_\alpha(\mathbf{r})\hat{A}\rangle \neq \langle\hat{\mathbf{C}}_\alpha(\mathbf{r},t)\hat{A}(t)\rangle$, as one would expect on general grounds and which is confirmed by the concrete examples presented below. ii) Although the sum of the partial hypercurrent correlation functions is independent of time,

$$\mathbf{S}_A(\mathbf{r}) + \mathbf{C}_A^{\mathrm{id}}(\mathbf{r},t) + \mathbf{C}_A^{\mathrm{int}}(\mathbf{r},t) + \mathbf{C}_A^{\mathrm{ext}}(\mathbf{r},t) = 0, \quad (54)$$

the individual ideal, interparticle, and external contributions each retain non-trivial temporal dependence; we recall $\mathbf{C}_A^{\mathrm{id}}(\mathbf{r},t) = \langle\hat{\mathbf{C}}_{\mathrm{id}}(\mathbf{r},t)\hat{A}(t)\rangle$, *etc.* for 'int' and 'ext'. One might expect this temporal behaviour to occur on grounds of the general nontrivial setup of these correlation functions and we exemplify the temporal dependence in specific model situations in the following.

B. Nonequilibrium ideal gas of harmonic oscillators

As an initial application of the general dynamical gauge invariance framework to a conrete system, we consider an ideal gas of N particles, with vanishing interparticle potential $u(\mathbf{r}^N) = 0$, in one spatial dimension. We demonstrate explicitly that the theory allows one i) to recover the correct static hyperforce limit as described in Sec. IV A, ii) to identify non-trivial time dependence in equilibrium, iii) to discriminate between thermal equilibrium states and nonequilibrium stationary states, and vi) to retain the general theoretical structure when addressing nontrivial temporal dependence in more general nonequilibrium ideal gas setups. Furthermore, the analytical solution provides a useful reference for validation of the simulation methods.

1. Setup of the model and dynamical density profile

To facilitate analytical treatment, we choose the external potential as being harmonic: $V_{\mathrm{ext}}(x) = kx^2/2$, with strength parameter k and one-dimensional position coordinate x. The initial Hamiltonian has a positive spring constant $k_0 > 0$, particle mass m_0, and corresponding frequency $\omega_0 = \sqrt{k_0/m_0}$. At times $t \geq 0$, we allow in general the constants to be $m \neq m_0$ and $k \neq k_0$, with corresponding frequency $\omega = \sqrt{k/m}$ and again in general $\omega \neq \omega_0$. After the switching, the external potential can remain confining, $k > 0$, or vanish, $k = 0$, or become repulsive, $k < 0$. The latter case constitutes a dynamically unstable situation with the Hamiltonian being unbounded from below. The one-dimensional particle trajectories are $\hat{x}_i(t) = x_i\cos(\omega t) + p_i\sin(\omega t)/(m\omega)$ and $\hat{p}_i(t) = p_i\cos(\omega t) - m\omega x_i\sin(\omega t)$ with initial state x_i, p_i at time $t = 0$, for all $i = 1,\ldots,N$.

Straightforward algebra yields the dynamical density profile $\rho(x,t) = \langle\hat{\rho}(x,t)\rangle$ as a normalized Gaussian with temporally varying width parameter α:

$$\rho(x,t) = N\sqrt{\alpha/\pi}\,\mathrm{e}^{-\alpha x^2}, \quad (55)$$

$$\alpha = \beta k \Big/ \Big[\frac{k}{k_0} + \frac{m_0}{m} + \Big(\frac{k}{k_0} - \frac{m_0}{m}\Big)\cos(2\omega t)\Big], \quad (56)$$

with a scaled version of the expression (56) being given in Eq. (65).

We summarize several properties of this solution. Forgoing any switching and hence retaining $k = k_0$, $m = m_0$, the system remains in equilibrium at all times and the width parameter $\alpha_0 = \beta k_0/2 = \mathrm{const}$, independent of time. For general switching, $m \neq m_0$ and $k \neq k_0$, the value of α oscillates in time with doubled frequency 2ω (we comment on the unstable case at the end of the section). The temporal oscillations of α are bounded by two values, one being α_0, which is attained at times $t = n\pi/\omega$ with n being a nonnegative integer. The further bounding value of α is $\beta km/(2m_0)$, which is attained at times $t = (n + 1/2)\pi/\omega$. The midpoint value of α between the two extrema is $\beta k/[(k/k_0) + (m_0/m)]$.

As a special case one may choose parameters that satisfy $km = k_0m_0$. Then despite in general $\omega \neq \omega_0$, the density profile (55) remains independent of time and it thus forms a simple example of a nonequilibrium stationary state. The width parameter thereby retains its initial value $\alpha_0 = \beta k_0/2$, unperturbed by this specific switching, over the course of time.

Figure 3 displays results for dynamical density profiles $\rho(x,t)$ (first row) obtained from the analytical solution (55) for $m = m_0$ and the three representative cases of no switching: $k = k_0$, switching to a softer spring: $k = k_0/4$, and switching to an unstable situation: $k = -k_0$ (from left to right).

2. Ideal shift current balance

Turning to the gauge theory we first consider the static force density balance (12), which for the one-dimensional ideal gas simplifies to:

$$F_{\mathrm{id},0}(x) + F_{\mathrm{ext},0}(x) = 0, \quad (57)$$

where we use scalar notation and the interparticle force density vanishes, $F_{\mathrm{int},0}(x) = 0$, for the present ideal gas. The static ideal and external force fields follow from respective explicit calculation as

$$\frac{\beta F_{\mathrm{id},0}(x)}{\rho_0(x)} = \beta k_0 x, \quad (58)$$

$$\frac{\beta F_{\mathrm{ext},0}(x)}{\rho_0(x)} = -\beta k_0 x, \quad (59)$$

where we have normalized by the initial density profile $\rho_0(x)$; recall that this is a Gaussian defined by Eq. (55) with $\alpha_0 = \beta k_0/2$, such that $\rho_0(x) = N\sqrt{\beta k_0/(2\pi)}\exp(-\beta k_0 x^2/2)$. The results (58) and (59) satisfy the static force sum rule (57) explicitly.

FIG. 3. Correlation functions for the ideal gas in a harmonic trap in equilibrium (left column) and nonequilibrium (middle and right columns). Shown are space-time plots of the scaled dynamical density profile $\rho(x,t)a_0$ (top row) and for the different types of hypercurrent correlation functions that arise from dynamical gauge invariance, namely the ideal (second row) and external (third row) hypercurrent correlation functions for the center of mass, $\langle \hat{C}_{\mathrm{id}}(x,t)\hat{R}(t)\rangle a_0$ and $\langle \hat{C}_{\mathrm{ext}}(x,t)\hat{R}(t)\rangle a_0$, with the lengthscale $a_0 = 1/\sqrt{\beta k_0}$. The nonequilibrium sum rule Eq. (69) constrains the sum of these three contributions to vanish. Left column: Equilibrium dynamics where $H = H_0$ and $k = k_0$ at all times; despite the density profile being stationary both hypercurrent correlation functions oscillate in time. Middle column: At the initial time the value of the spring constant is reduced, $k = k_0/4$, such that temporal density oscillations are induced and these affect the hypercurrent correlation functions. Right column: At the initial time the sign of the harmonic potential is flipped, $k = -k_0$, which creates an unstable situation without corresponding equilibrium; pronounced structuring is apparent in both hypercurrent correlation functions.

The dynamic shift current balance (15) simplifies for the present system to consist of merely two contributions:

$$C_{\mathrm{id}}(x,t) + C_{\mathrm{ext}}(x,t) = 0, \qquad (60)$$

as the interparticle shift current vanishes, $C_{\mathrm{int}}(x,t) = 0$. Both the ideal and the external partial shift current is conveniently normalized by the dynamic density profile $\rho(x,t)$, which leads to the following odd cubic polynomials in position x:

$$\frac{C_{\mathrm{id}}(x,t)}{\rho(x,t)} = b_1 x + b_3 x^3, \qquad (61)$$

$$\frac{C_{\mathrm{ext}}(x,t)}{\rho(x,t)} = -b_1 x - b_3 x^3. \qquad (62)$$

The coefficients b_1 and b_3 are time-dependent and, when scaled by the appropriate powers α and α^2 of the width

parameter, possess the following forms:

$$\frac{b_1}{\alpha} = \frac{2(\kappa + \kappa_0)\cos(2\omega t) + (\kappa - \kappa_0)[5 - \cos(4\omega t)]/2}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}, \qquad (63)$$

$$\frac{b_3}{\alpha^2} = -\frac{(\kappa - \kappa_0)[1 - \cos(4\omega t)]}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}. \qquad (64)$$

Here we use the shorthand $\kappa = km$ to denote the product of spring constant k and mass m; correspondingly $\kappa_0 = k_0 m_0$ for the intial state parameters at time 0. Using these variables allows one to express the width parameter (56) succinctly as:

$$\alpha = \frac{\beta k_0 \kappa}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}. \qquad (65)$$

It is noteworthy that Eqs. (63)–(65) share a common denominator; this structure is a consequence of the scal-

FIG. 4. Comparison of results obtained analytically (left column) and from molecular dynamics simulations via autodifferentiation (center column) and via finite-difference differentiation (right column) with respect to the initial state of molecular trajectories. Shown is the scaled dynamical density profile $\rho(x,t)a_0$ (top row), the ideal part, $\langle \hat{C}_{\mathrm{id}}(x,t)\hat{R}(t)\rangle a_0$, and the external contribution, $\langle \hat{C}_{\mathrm{ext}}(x,t)\hat{R}(t)\rangle a_0$, of the hypercurrent correlation function for the scaled center of mass $\hat{R}$. The results are shown as a function of the scaled distance x/a_0, with natural lengthscale $a_0 = 1/\sqrt{\beta k_0}$ and scaled time $t\omega/\pi$. The switching protocol implies $k = k_0/4$ and $m = m_0$. Except for small numerical sampling artifacts, the results from all three methods agree with each other to high precision and they satisfy the sum rule (69).

ing with $\rho(x,t)$ in Eqs. (61) and (62) and the scaling with powers of α in Eqs. (63) and (64).

Considering the limit $t \to 0^+$ leads to the coefficients (63) and (64) attaining respective values $b_1 = \beta k_0$ and $b_3 = 0$, with the width parameter (65) becoming $\alpha_0 = \beta k_0/2$. Thus both the ideal and the external contribution to the dynamic shift current $C(x,0)$ reduce correctly to their corresponding thermally scaled initial partial force density: $C_{\mathrm{id}}(x,0) = \beta F_{\mathrm{id},0}(x)$ and $C_{\mathrm{ext}}(x,0) = \beta F_{\mathrm{ext},0}(x)$.

We emphasize that the cubic spatial dependence of the (scaled) external shift current $C_{\mathrm{ext}}(x,t)/\rho(x,t)$ given by Eq. (62) is very different from the behaviour of the time-dependent external force field, which remains linear, $F_{\mathrm{ext}}(x,t)/\rho(x,t) = -kx$, for all times $t \geq 0$.

3. Ideal hypercurrent balance

As a representative hyperobservable, we consider the sum of positions $\hat{R} = \sum_i \hat{x}_i$; we recall the general hyper-

current sum rule Eq. (51) for $\hat{\mathbf{R}}(t)$. We first consider the corresponding static hyperforce balance, which possesses the following initial state form:

$$\rho_0(x) + \langle \beta \hat{F}_{\mathrm{id},0}(x)\hat{R}\rangle + \langle \beta \hat{F}_{\mathrm{ext},0}(x)\hat{R}\rangle = 0. \quad (66)$$

The two static correlation functions in Eq. (66) follow from explicit algebra in the respective forms:

$$\frac{\langle \beta \hat{F}_{\mathrm{id},0}(x)\hat{R}\rangle}{\rho_0(x)} = \beta k_0 x^2 - 1, \quad (67)$$

$$\frac{\langle \beta \hat{F}_{\mathrm{ext},0}(x)\hat{R}\rangle}{\rho_0(x)} = -\beta k_0 x^2, \quad (68)$$

where the initial state density profile $\rho_0(x)$ is given below Eq. (59) in its explicit Gaussian form with width parameter α_0. Again Eqs. (67) and (68) satisfy the static sum rule (66), as can be seen directly by summing up Eqs. (67) and (68), adding unity and multiplying by $\rho_0(x)$.

The hypercurrent sum rule (51) for the one-dimensional ideal gas attains the form:

$$\rho(x,t) + \langle \hat{C}_{\mathrm{id}}(x,t)\hat{R}(t)\rangle + \langle \hat{C}_{\mathrm{ext}}(x,t)\hat{R}(t)\rangle = 0, \quad (69)$$

FIG. 5. Molecular dynamics results for the hypercurrent correlation functions of a one-dimensional system of particles that mutually repel via the Weeks-Chandler-Andersen pair potential. Shown is the dynamical density profile $\rho(x,t)a$ (top row) and the three contributions to the hypercurrent correlation function for the (scaled) center of mass, $\hat{A} = \hat{R}$, namely the ideal part $\langle \hat{C}_{\text{id}}(x,t)\hat{R}(t)\rangle a$ (second row), the external part $\langle \hat{C}_{\text{ext}}(x,t)\hat{R}(t)\rangle a$ (third row), and the interparticle interaction contribution $\langle \hat{C}_{\text{int}}(x,t)\hat{R}(t)\rangle a$ (bottom row), shown as a function of the scaled distance x/a and scaled time t/t_{MD}, where a is the particle size and $t_{\text{MD}} = a\sqrt{m\beta}$ is the time scale. The results are shown for a harmonic trap that is switched narrower (first, second, and third column) and wider (fourth column), as obtained from finite-difference differentiation (first column) and via automatic differentiation (second, third, and fourth columns). The results from both methods agree with each other for identical conditions (first and second columns) and the one-dimensional form (75) of the dynamical sum rule (51) is satisfied with high accuracy.

where $\hat{C}(x,t) = \hat{C}_{\text{id}}(x,t) + \hat{C}_{\text{ext}}(x,t)$ is the one-dimensional version of the shift current operator with $\hat{C}_{\text{int}}(x,t) = 0$, as the particles are ideal. The two hypercurrent correlation functions in Eq. (69) follow by explicit calculation as two even fourth-order polynomials

in x, when normalized by the dynamic density profile:

$$\frac{\langle \hat{C}_{\text{id}}(x,t)\hat{R}(t)\rangle}{\rho(x,t)} = -1 + b_0 + b_2 x^2 + b_4 x^4, \quad (70)$$

$$\frac{\langle \hat{C}_{\text{ext}}(x,t)\hat{R}(t)\rangle}{\rho(x,t)} = -b_0 - b_2 x^2 - b_4 x^4. \quad (71)$$

Again, the solution (70) and (71) satisfies the dynamical sum rule (69), as follows straightforwardly.

FIG. 6. Splitting of the hypercurrent correlation function into transport and acceleration contributions. Shown is the scaled dynamical density profile $\rho(x,t)a$ (top row) obtained from histogram filling together with its gradient $\nabla\rho(x,t)a^2 = \partial\rho(x,t)a^2/\partial x$ (second row), as obtained from numerical differentiation with respect to position x. The scaled negative mean shift stress $-\tau_C(x,t)a$ (third row) is identical, on the scale of the plot, to the density profile (first row), which verifies the sum rule (41): $\tau_C(x,t) = -\rho(x,t)$. The scaled shift acceleration current $C_{\mathrm{acc}}(x,t)a$ (bottom row) is numerically identical to the density gradient (second row), thus verifying the identity (42): $C_{\mathrm{acc}}(x,t) = \nabla\rho(x,t)$, as is demonstrated by high-quality averaging over 10^7 trajectories (left column). Reduced averaging over only 10^5 trajectories (right column) reveals reduction of statistical noise of $C_{\mathrm{acc}}(x,t)a$ (right column, bottom panel) as compared to the direct result for $\nabla\rho(x,t)a^2$ (right column, second panel from top), which renders the shift acceleration current a potential candidate for nonequilibrium reduced-variance sampling.

The coefficients b_0, b_2, and b_4 in Eqs. (70) and (71) are time-dependent in general. Scaling by the respective appropriate powers $1, \alpha$, and α^2 of the time-dependent Gaussian width parameter α, see Eq. (65) for its explicit

form, yields:

$$b_0 = \frac{\kappa_0[1 - \cos(2\omega t)]}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}, \tag{72}$$

$$\frac{b_2}{\alpha} = \frac{2(\kappa + \kappa_0)\cos(2\omega t) + (\kappa - \kappa_0)[3 - \cos(4\omega t)]}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}, \tag{73}$$

$$\frac{b_4}{\alpha^2} = -\frac{(\kappa - \kappa_0)[1 - \cos(4\omega t)]}{\kappa + \kappa_0 + (\kappa - \kappa_0)\cos(2\omega t)}, \tag{74}$$

where as before $\omega = \sqrt{k/m}$ is the oscillator frequency after switching and the commonality of denominators, as observed in Eqs. (63)–(65), is retained. Comparison of Eq. (74) to the coefficient (64) for the shift current reveals that $b_3 = b_4$.

As a consistency check of the static and dynamical solutions, at time $t = 0$ and hence $\alpha_0 = \beta k_0/2$, the dynamical coefficients (72)–(74) reduce to: $b_0 = 0, b_2 = \beta k_0$, and $b_4 = 0$. Inserting these results into the hypercurrent correlation functions (70) and (71) reduces these expressions correctly to the respective static hyperforce correlation functions (67) and (68), such that indeed $\langle \hat{C}_{\mathrm{id}}(x,0)\hat{R}(0)\rangle = \langle \beta \hat{F}_{\mathrm{id},0}(x)\hat{R}\rangle$ and $\langle \hat{C}_{\mathrm{ext}}(x,0)\hat{R}(0)\rangle = \langle \beta \hat{F}_{\mathrm{ext},0}(x)\hat{R}\rangle$.

The ideal and external parts of the hypercurrent correlation function, $\langle \hat{C}_{\mathrm{id}}(x,t)\hat{R}(t)\rangle$ and $\langle \hat{C}_{\mathrm{ext}}(x,t)\hat{R}(t)\rangle$, as respectively given by Eqs. (70) and (71), are depicted graphically in the second and third row of Fig. 3. We recall the above description of the three considered cases of no switching (first column), switching to softer confinement (second column), and switching to an unstable situtation by reversing the sign of the force constant (third column); we return to the latter case at the end of Sec. IV B 4. As anticipated in the discussion given in Sec. IV A on the basis of general arguments, the hyperforce correlation funtions indeed display nontrivial time dependence, and they provide arguably much deeper insight into the dynamical structuring than does the dynamical density profile (first row in Fig. 3).

4. Reduction to limiting cases of the ideal gas

As a specific simple situation, it is interesting to consider the dynamics when the model parameters are kept constant at the initial time, $k = k_0$ and $m = m_0$, and hence no switching occurs. Then $b_1 = 2\alpha_0 \cos(2\omega_0 t) = \beta k_0 \cos(2\omega_0 t)$ from Eq. (63) and $b_3 = 0$ from Eq. (64). Hence the ideal and external mean shift current both follow from Eqs. (61) and (62) as $C_{\mathrm{id}}(x,t) = -C_{\mathrm{ext}}(x,t) = \beta k_0 x \cos(2\omega_0 t)\rho_0(x)$, where we recall $\alpha_0 = \beta k_0/2$ as the initial width parameter and $\omega_0 = \sqrt{k_0/m_0}$ as the initial frequency and note the period doubling effect. Furthermore, for the present case of switching being absent, the hypercurrent identity (69), reduces from the general nonequilibrium solution (72)–(74) similarly to $b_0 = [1 - \cos(2\omega_0 t)]/2$, $b_2 = \beta k_0 \cos(2\omega_0 t)$, and $b_4 = 0$,

which simplifies the ideal (70) and external (71) hypercurrent correlation functions. Hence the occurring oscillation is characterized by a doubled initial frequency $2\omega_0$. It is remarkable that even in this arguably simplest setup of noninteracting harmonic oscillators in thermal equilibrium, there is nontrivial temporal dependence exposed by the hypercurrent approach; we recall the illustration of the ideal and external hypercurrent correlation functions in the first column of Fig. 3.

Turning to the case of nonequilibrium steady states with constrained parameter choices $km = k_0 m_0$, the coefficients of the shift current are $b_1 = \beta k_0 \cos(2\omega t)$, $b_3 = 0$ and hence $C_{\mathrm{id}}(x,t) = -C_{\mathrm{ext}}(x,t) = \beta k_0 x \cos(2\omega t)\rho_0(x)$, where the frequency is twice that *after* switching: recall $\omega = \sqrt{k/m}$ and that this is in general different from the doubled frequency $2\omega_0$ identified above in equilibrium. We recall that the density profile itself remains stationary in the present case. The hypercurrent coefficients are $b_0 = [1 - \cos(2\omega t)]/2$, $b_2 = 2\alpha_0 \cos(2\omega t) = \beta k_0 \cos(2\omega t)$, and $b_4 = 0$.

As a final case, the presented general solution for the contributions to the shift current identity (Sec. IV B 2) and to the hypercurrent balance (Sec. IV B 3) remains valid when the external potential is no longer confining and thus $k < 0$. Then the trigonometric dependence becomes hyperbolic, such that e.g., $\cos(2\omega t) = \cosh(2|\omega|t)$, where $|\omega| = \omega/i = \sqrt{-k/m} \geq 0$ with imaginary unit i, which again results in non-trivial time dependence in the dynamical correlation functions (70) and (71), see the last column of Fig. 3 for the graphical representations. Taking the limit $k \to 0$, such that the system is free at $t > 0$, allows one to make further simplifications to the above analytical results.

C. Molecular dynamics simulations

1. Simulation methodology

We turn to simulations to gain further and deeper physical insight into the nature of the hypercurrent correlation structure beyond the simple nonequilibrium ideal gas setup of Sec. IV B. We base our methodology on the trajectory level, as described in Sec. II D and in particular on the initial state differentiation laid out in Sec. III F.

We use two different methods to realize the initial state time differentiation. As a basis for the time evolution we use in both methods the velocity Verlet algorithm [7, 8, 77], see Sec. II D, but we see no reason why one should not be able to use other molecular dynamics time integrators [8]. First, to realize the initial state derivative, we use automatic differentiation to keep track of the effect of differential changes in the initial microstate of each molecular dynamics trajectory. Results for the shift current and for the hypercurrent correlation functions are then obtained according to the trajectory-based picture described in Sec. III F. Implementation-wise, the method requires the molecular dynamics to be performed in an

FIG. 7. Dynamical gauge correlation functions of the ideal gas (left column) and of mutually interacting particles (right column) inside of an external double-well potential. Shown is the scaled dynamical density profile $\rho(x,t)a$ (top row) and the hypercurrent correlation function for the total position, consisting of the ideal part, $\langle C_{\mathrm{id}}(x,t)R(t)\rangle a$ (second row), external part, $\langle C_{\mathrm{ext}}(x,t)R(t)\rangle a$ (third row), and interparticle contribution, $\langle C_{\mathrm{int}}(x,t)R(t)\rangle a$ (bottom row). The system is in equilibrium and thus the density profile is stationary. Note the spatiotemporal hypercurrent structure that is indicative of barrier crossing.

environment that provides ready access to the automatic differentiation functionality; we use a custom molecular dynamics implementation in the programming language Julia [80] and note recent progress in differentiable molecular dynamics [63], where libraries are available.

As a second and alternative route, we perform differentiation via finite differences, which can be realized in any molecular dynamics environment. Here the initial state differentiation is based on dynamically propagating one additional trajectory that differs from the corresponding original trajectory by a single, additional time step that is performed on the initial microstate and with respect to the dynamics generated by the initial state Hamilto-

nian H_0. The magnitude of this time step can be chosen freely, as described below. The thus altered initial microstate is then propagated forward in time, in an identical way as the original corresponding trajectory, i.e., on the basis of the given form of H for $t \geq 0$. At (each) target time t, the finite difference with the unperturbed trajectory then provides the numerator for the finite difference ratio; the denominator is the time step used for the perturbation of the initial state. To perform the dynamical evolution, we use a standard value of the size of the time step, $\Delta t = 5 \times 10^{-3} t_{\mathrm{MD}}$, with microscopic time scale $t_{\mathrm{MD}} = a\sqrt{m\beta}$, where a is the particle size (as specified below). The initial time derivative is performed

with a single, much smaller initial time step of $10^{-4} t_{\mathrm{MD}}$.

To first validate both simulation methods we use the harmonic oscillator setup, as described above in Sec. IV B, and compare against the analytical solution. The use of the simulation methods in this case also serves to demonstrate and illustrate the arguably more intuitive access that the trajectory-based picture provides for the dynamical gauge theory. Despite the simplicity of the noninteracting sytem, we deem the test to be nontrivial, due to the significant conceptual differences between the three methods (analytical solution, automatic and finite-difference trajectory differentiation) to obtain the respective results. We recall the explicit phase space manipulations, described in Sec. IV B, to obtain the analytical solution and the trajectory-based formulation of the initial state time derivative in Sec. III F.

In Fig. 4 we use the analytical solution as a reference (left column) to compare the results from both simulation methods: automatic differentiation (center column) and finite-difference differentiation (right column). As expected, the results for the dynamical density profile $\rho(x, t)$ are numerically identical. We recall that based on the simulation, mere filling of a position- and time-resolved histogram of particle positions is required. Averaging over the initial ensemble, as is realized via Monte Carlo simulations on the basis of the initial state Hamiltonian H_0 and initial inverse temperature β, then yields the numerical results.

Obtaining simulation results for the ideal and external hypercurrent correlation functions, $\langle \hat{C}_{\mathrm{id}}(x, t) \hat{R}(t) \rangle$ and $\langle \hat{C}_{\mathrm{ext}}(x, t) \hat{R}(t) \rangle$, requires to carry out explicitly the initial state time differentiation, as is apparent from the definition (13) of the shift current observable. Figure 4 demonstrates excellent agreement of the results from both simulation methods with each other and with the analytical solution. Besides a strong validation of the computational methodology (we compare both simulation methods against each other for mutually interacting particles below) this also confirms the successful mirroring of the dynamical gauge invariance theory, as developed in Sec. III on the basis of phase space differential operator methods, using the arguably more intuitive trajectory picture; see the description in Secs. II D and III F.

2. Interacting particles in harmonic confinement

Both molecular dynamics simulation methods allow one to go beyond noninteracting systems in a relatively straightforward way, as the required time evolution remains based on temporal discretization, here via the velocity Verlet algorithm, and the initial state time derivative remains identical. As a crucial difference to the ideal gas, the interparticle shift current observable $\hat{C}_{\mathrm{int}}(x, t)$ no longer vanishes, and all further shift and hypercurrent observables are naturally affected by the altered dynamics.

We consider mutually repulsive particles that interact with a pair potential $\phi(r)$ as a function of (one-

dimensional) interparticle distance r. Figure 5 displays results for the Weeks-Chandler-Andersen pair potential as a prototypical short-ranged, strongly repulsive model. Explicitly the pair potential is thereby given by $\phi(r) = 4\epsilon[(r/a)^{-12} - (r/a)^{-6} + 1/4]$ for $r \leq 2^{1/6} a$ and zero otherwise, where a is a lengthscale, ϵ is an energy scale, and we choose temperature such that $\beta\epsilon = 1$.

The general center-of-mass sum rule (51) attains the following one-dimensional split form:

$$\rho(x, t) + \langle \hat{C}_{\mathrm{id}}(x, t) \hat{R}(t) \rangle + \langle \hat{C}_{\mathrm{int}}(x, t) \hat{R}(t) \rangle$$
$$+ \langle \hat{C}_{\mathrm{ext}}(x, t) \hat{R}(t) \rangle = 0. \tag{75}$$

Figure 5 displays results for the density profile and the further three hypercurrent correlation functions on the left hand side of Eq. (75) using various different switching setups; see the caption of Fig. 5. The simulation results from automatic differentiation agree with those from the finite-difference method within high numerical accuracy. Besides providing a consistency check, this agreement validates each method as being fit for applying the hypercurrent framework to systems of mutually interacting particles. Despite our test system being only one-dimensional, the fact that both differentiation methods perform well can also serve as a vindication of the implied concept of initial state differentiation, as it arises from the dynamical gauge invariance.

3. Towards reduced-variance sampling

We have thus far presented simulation results based on the splitting of the hypercurrent correlation function into ideal, interparticle, and external contributions as they arise from the splitting of the initial state Liouvillian L_0; we recall Eq. (5). We next address the splitting of the hypercurrent observable into acceleration and transport contributions presented in Sec. III F; see specifically Eq. (38) therein. For the present situation, we express the general relationships (41) and (42) in the following one-dimensional forms:

$$\tau_C(x, t) = -\rho(x, t), \tag{76}$$
$$C_{\mathrm{acc}}(x, t) = \nabla\rho(x, t), \tag{77}$$

where $\nabla = \partial/\partial x$ and the general d-dimensional shift current sum rule (43) simplifies to $C_{\mathrm{acc}}(x, t) + \nabla\tau_C(x, t) = 0$.

We consider the case of switching to a stiffer trap and present the corresponding dynamical density profile $\rho(x, t)$ together with its numerically differentiated gradient, $\nabla\rho(x, t)$, in the first and second rows of Fig. 6. Thereby the results for the density profile are obtained from histogram-resolved counting of particle positions. Then calculating the numerical position derivative of the data gives results for the density gradient, which displays the typical effect of amplification of the statistical noise.

The third panel in Fig. 6 shows results for the mean shift stress tensor (39), which we spell out in one dimension as: $\tau_C(x, t) = -\beta\langle \sum_i \delta(x - \hat{x}_i(t)) \hat{x}_i^{\bullet}(t) \hat{p}_i(t) \rangle$; we

recall that the bold dot denotes the initial-time derivative (29). The simulation results for $\tau_C(x,t)$ are numerically nearly identical to those in the first row of Fig. 6, which validates the sum rule (76). The general definition of the acceleration part (40) of the shift current, as is relevant for Eq. (77), attains the form: $C_{\mathrm{acc}}(x,t) = \beta\langle\sum_i \delta(x - \hat{x}_i(t))\hat{p}_i^{\bullet}(t)\rangle$. The corresponding simulation results are shown in the fourth row of Fig. 6. It is noteworthy that these results i) are consistent with those presented in the second row for $\nabla\rho(x,t)$, as expected from the sum rule (77), and ii) that they show reduced statistical noise – see the right column of Fig. 6 where the number of trajectories has been reduced to demonstrate the effect.

Hence, strikingly, each of the sum rules (76) and (77) provides an independent means of *practical* access to the dynamical density profile $\rho(x,t)$. (Using Eq. (77) requires spatial integration to undo the gradient operation; see e.g. Refs. [81–83] that address this point in equilibrium.) When using the finite-difference method to carry out the initial time derivative, the computational overhead involves a mere factor 2. The thus obtained results display reduced statistical variance over the counting method. Hence the present methodology offers a way forward to generalize equilibrium reduced-variance (force-sampling) methods [81–83] to *nonequilibrium* situations. This could potentially be very general, when starting with the hypercurrent identities (46) and (47) that hold for general observables $\hat{A}$. We have verified numerically both of these identities in the present system and for the present choice of hyperobservable (results not shown).

4. Double-well potential and barrier crossing

To demonstrate that genuine physical insight can be gained from the present framework, we turn to the classical double-well problem and show in Fig. 7 results for both the ideal gas and for repulsive Weeks-Chandler-Andersen particles. As a choice for a specific situation, we keep the external double-well potential unchanged over time, $V_{\mathrm{ext}}(x) = \epsilon(x^2 - x_d^2)^2/x_d^4$, with constant distance $x_d = 2.5a$ between the maximum and each minimum; ϵ is the energy scale of the pair interaction potential and a is the particle size and we consider $\beta\epsilon = 1$ as before. The density profile remains stationary in this situation and it is identical to its initial equilibrium form (see the panels in the top row of Fig. 7). Both the ideal and external parts of the hypercurrent correlation function exhibit pronounced spatiotemporal structure; we recall the sum rule (51), which we find again to be satified with high numerical accuracy.

The spatiotemporal structuring of the hypercurrent correlation functions shows clear signs of barrier crossings, see the diagonal streaks in Figs. 7. Comparing the behaviour of the interacting system with that of the ideal gas reveals that the presence of the repulsive interparticle interactions leads to more immediate barrier crossing, as

is arguably consistent with intuition. We conclude that the present setup offers fresh insight into the coupled motion in complex energy landscapes and leave more detailed investigations to future work.

V. CONCLUSIONS

In conclusion, we have explored the consequences of dynamical gauge invariance against phase space shifting in the nonequilibrium statistical mechanics of many-body systems. The shifting transformation has been identified previously for systems in thermal equilibrium [55, 56], where it was shown to induce exact static identities, ranging from global force [47] and variance identities [49], force-force correlation '3g'-sum rules [51], to locally resolved quantum [50] and classical [53] force balance relationships. The framework also allows one to obtain very general hyperforce correlation identities [54] which are embedded, via an associated generalized ensemble, in hyperdensity functional theory for the behaviour of general observables in equilibrium [67, 68].

The derivation of these prior equilibrium results was based primarily on variational calculus and in particular on exploiting the properties of functional derivatives with respect to the shifting field that parameterizes the transform on phase space. As the shifting field itself plays the role of a mere gauge function [55, 56], here we have rather worked with the equivalent differential operator formalism [55, 56]. The higher level of abstraction that the operator method provides over the variational method leads to significant simplification of the complexity of the required algebra. We have shown that the static shifting operators $\boldsymbol{\sigma}(\mathbf{r})$ acquire time dependence via the standard embedding inside of a propagator 'sandwich' as given by the time evolution (19) for $\boldsymbol{\sigma}(\mathbf{r},t)$. This particular propagator structure might be more familiar from quantum theory [78] than it is within the present classical physics, but it indeed also constitutes a general property of (classical) differential operators, as laid out in Sec. II B.

The mechanism of temporal nonlocality that is inherent in the dynamical gauge invariance is markedly different from the more common memory integral formalism, as is central in modelling via generalized Langevin equations [6, 7, 10] and power functional theory [18, 84–86]. Rather than a temporal integral, the gauge framework features a time differential structure, which arises from a specific differential change according to the time evolution of the initial equilibrium ensemble. While this operation can be seen technically as a perturbation, it is also arguably the most natural one, as it is inherent in the initial ensemble and the initial thermal distribution function remains invariant under the change; we recall the illustration shown in Fig. 1.

Our primary general results are the shift current balance (14) and the hypercurrent identity (26), which are both exact. These sum rules acquire via decomposing into ideal (kinetic), interparticle, and external contribu-

tions the respective forms (15) and (28). We have considered four concrete examples of hyperobservables, where in particular the correlation function of the sum of all positions with the shift current equals the (negative) dynamical density profile, see Eq. (51).

The harmonically confined ideal gas served as a toy model to illustrate some of the properties of the shift and hypercurrent correlation function. An initial thermal ensemble is thereby set into motion by switching both the spring constant and the particle mass in the most general case. Given the simplicity of the setup, the resulting rich gauge correlation structure is striking, as it displays, e.g., period doubling effects and polynomial contributions in position of higher order than one might expect naively to find in a system of harmonic oscillators.

Our simulation results have confirmed the above picture via reproducing the analytical ideal gas solution, which provides a consistency check, and via enabling us to address the effects of interparticle interactions on the gauge correlation behaviour. It is noteworthy that the effects of interparticle interaction are incorporated effortlessly into the setup. These effects are contained in an interparticle contribution to the shift current, which upon averaging forms the mean interparticle shift current and furthermore consitutes the correlator contribution in the hypercurrent sum rule (28). All contributions to the shift current observable $\hat{\mathbf{C}}(\mathbf{r}, t)$ are thereby accessible via automatic differentiation; we recall our description of the virtues of the method in the introduction (Sec. I). Nevertheless, working with finite differences is entirely feasible, as we have demonstrated; see the comparison of both simulation methods presented in Fig. 4 and in Fig. 5 (see its two leftmost columns). The finite-difference method requires only very moderate overhead over standard molecular dynamics work, as one merely needs to analyze the differences of pairs of trajectories that differ by a small finite change in their initial microstates.

It is worth pointing out several differences of the dynamical gauge invariance theory with several established methods in nonequilibrium statistical mechanics. The present framework is exact and no approximations are involved in the treatment of the many-body physics as set up in Sec. II. Yet, investigating the combination with approximate closure relations could be worthwhile. The central concept of initial state time differentiation, as described in Sec. III E, is different from standard dynamical perturbation analysis against generic changes in the initial conditions. Here the change in initial conditions is generated by the application of the initial state Liouvillian L_0, which is natural, as the initial thermal ensemble is invariant under its action to generate the time evolution of the initial state.

The dynamical gauge invariance leads to shift current and hypercurrent sum rules that are structurally different from the Jarzynski equation [87] and the further fluctuation theorems of stochastic thermodynamics [13]. Yet, exploring connections and possible cross-fertilization with stochastic thermodynamics constitutes an interesting topic for future work. While the present treatment is based genuinely on dynamical averages, as is standard procedure [6], the dynamical gauge invariance framework is distinct from the projection operator formalism and from mode-coupling theory. Investigating the consequences for these approaches is a valuable point for future work. In particular tracing out the connections with the Yvon theorem [7, 35], as used in Götze's seminal account of mode-coupling theory [11], constitutes an intereresting topic.

The mapped averaging framework developed by Kofke and coworkers [88–100] is a highly efficient sampling scheme for equilibrium properties of complex systems. Whether the present methodology could help to generalize the mapped averaging to nonequilibrium situations is an interesting question. Furthermore, possible generalizations of force sampling [81–83, 100–105], as used previously for local transport coefficients and mobility profiles within the Green-Kubo formalism [81, 104], could be very interesting. The reduced-variance effect described in Sec. IV C 3 points to the practical feasibility. Finally, the connection to power functional theory [18, 19] is worth exploring and whether the present formalism can shed further light on the limitations of the dynamical density functional theory [20]. Potential applications of the dynamical gauge sum rules include the development of convergence tests and sampling schemes for simulations and to provide consistency checks for dynamical neural functionals [18, 20, 21].

DATA AVAILABILITY

The data that support the findings of this study are openly available [80].

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